

## **Intrinsically Stretchable, Semi-transparent Organic Photovoltaics with High Efficiency and Mechanical Robustness via Full Solution Process**

*Jiaming Huang<sup>1</sup>, Zhen Lu<sup>1</sup>, Jiaqi He<sup>1,2</sup>, Hong Hu<sup>3</sup>, Qiong Liang<sup>1</sup>, Kuan Liu<sup>1</sup>, Zhiwei Ren<sup>1</sup>, Yaokang Zhang<sup>2</sup>, Hongyu Yu<sup>2</sup>, Zijian Zheng<sup>3</sup> and Gang Li<sup>1\*</sup>*

J. Huang, Z. Lu, J. He, Q. Liang, Dr. K. Liu, Dr. Z. Ren, and Prof. G. Li

Department of Electronic and Information Engineering, Research Institute for Smart Energy (RISE),

The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong, China.

The Hong Kong Polytechnic University Shenzhen Research Institute, Shenzhen 518057, China.

E-mail: gang.w.li@polyu.edu.hk

J. He, Prof. H. Yu

School of Microelectronics, Southern University of Science and Technology, Shenzhen, China

Dr. H. Hu, Dr. Y. Zhang, Prof. Z. Zheng

Laboratory for Advanced Interfacial Materials and Devices, Institute of Textiles and Clothing, The

Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong, China.

## Methods

### Materials

The silver nanowires (AgNWs) dispersion in isopropyl alcohol (IPA) (D: 30 nm, L: 30  $\mu$ m, 10 mg/ml) is purchased from Beijing Deke Daojin Science And Technology Co., Ltd. PM6, BTP-eC9, PY-IT, N2200 were purchased from Solarmer Materials Inc, Beijing. TPU Elastollan 685 A was purchased from Basf. Al-doped Zinc oxide, chlorobenzene, chloroform (CF), N,N-dimethylformamide (DMF), 1,8-diiodooctane (DIO), and 1-chloronaphthalene (CN) were purchased by Sigma-Aldrich. Poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PVP AI4083) was purchased from Heraeus Deutschland GmbH & Co. KG. Patterned ITO/glass substrates were purchased from Huananxiangcheng Ltd. All materials and solvents were used as received without further purification.

### Device Fabrication

For the device based on AgNWs@TPU, AgNWs were diluted into IPA and were put into a sonication bath for 5-10 min until good dispersion. The AgNWs were spin-coated onto a pre-cleaned glass with 2000 rpm, then thermal annealing at 200  $^{\circ}$ C for 5 min and 150  $^{\circ}$ C for 15 min. The pattern was achieved by polyimide tape. The AgNWs film was then soaked in deionized water (DI-water) to remove the surfactant and dry at 110  $^{\circ}$ C for 10 min. Then the TPU (200 mg/ml in DMF) was spin-coated on AgNWs at 1000 rpm for 1 min and dry at 80  $^{\circ}$ C for 5 h. Then the TPU@AgNWs were prepared after peel-off from the glass substrate. The silver paste was painted at the edge of AgNWs for contact.

Before spin-coating PEDOT:PSS, the AgNWs@TPU were UV-ozone treated for 5 min. PEDOT:PSS was spin-coated with 3000 rpm for 30 s and then annealed at 110  $^{\circ}$ C for 15 min. Then the samples were moved into the glovebox. Afterwards, for the active layer, the PM6:BTP-eC9 (22 mg/ml in CB, D:A = 1:1.2, 0.5% DIO, 2000 rpm/30s), PM6:PY-IT (14 mg/ml in CF, D:A = 1:1, 1% CN, 2000 rpm/30s), and PBDB-T:N2200 (10 mg/ml in CB, D:A = 2:1, 1% DIO, 2000 rpm) were deposited onto the PEDOT:PSS films and annealing at 100 $^{\circ}$ C for 10 min. Then, AZO dispersion (0.8% wt in IPA) was spin-coated onto active layers at 2000 rpm for 30 s. After annealing at 60 $^{\circ}$ C for 8 min, the AgNWs (5 mg/ml) were spin-coated onto AZO at 1000 rpm for 30 s and annealed at 100  $^{\circ}$ C for 10 min. For the AAA back electrode, another AZO (2.5% wt in IPA) was spin-coated onto AgNWs at 1000 rpm for 30 s. The pattern was achieved by PDMS. The silver paste was painted at the edge of AgNWs for contact. For the devices based on Ag electrode, Ag (100 nm) was evaporated through a shadow mask. The effective area of the device is 0.04 cm<sup>2</sup>.

For the device based on ITO/glass, the patterned ITO was sonically cleaned in detergent aqueous solution, DI-water, acetone, and IPA for 40 min subsequently. After UV-Ozone treatment for 20 min, the PEDOT:PSS was spin-coated. The following procedures are the same as the details above. For OLED devices, the active layer material is spiro co-polymer with a concentration of 8 mg/ml in toluene.<sup>1</sup> Then spin-coating with 2000 rpm for 40 s followed by thermal annealing at 100 $^{\circ}$ C for 10 min. Other steps are similar to *is*-OPV.

## Simulation

### 1 Details of the adhesion failure under tensile strain

The model was performed in the commercial software Abaqus with the dynamic/explicit solver. All the materials were assumed isotropic and linearly elastic. To ensure the model is a quasi-static process, the total kinetic energy was strictly constrained to only 1% of the total strain energy by tuning the time period. When the two contact surfaces separated from each other, they experienced attractive traction, which increased linearly to the maximum of  $\sigma = \sigma_m$  at the separation of  $\delta = \delta_m$ . Afterward, the traction decayed linearly and finally vanished. The failure degree of the CZM (D) can define the detachment of the NWs on the substrate. When the CZM started to decay (*i.e.*,  $\delta = \delta_m$ ) D was equal to 0. As D increased to unity with the increase of  $\delta$ , no attractive attraction was applied to the interface, indicating the complete detachment of the NWs. The area underneath the traction-separation curve was defined as the toughness (or namely, interfacial binding energy) of the interface ( $\Gamma$ ). To reveal the geometrical confinement effect of the upper AZO in preventing the detachment of nanowires, we assumed that there was also an interface between the upper AZO and underlying AZO ETL. Differing from the nanowire-AZO interface (interface 1), the material at the two sides of this interface (interface 2) was the same (AZO). Regarding neglectable material mismatch, interface 2 was assumed to be much stronger than interface 1 (*i.e.*,  $\Gamma_2$  is much larger than  $\Gamma_1$ ). With these definitions, a displacement boundary condition was applied on the plane of the right-hand side of the substrate to stretch the part. The right-hand side plane is symmetrical, while the back- and front sides were periodic boundaries.<sup>2</sup>

The parameters used in our model are included in the following table.

| Item                                                                      | Value |
|---------------------------------------------------------------------------|-------|
|                                                                           | e     |
| Young's modulus of nanowires, $E_{NW}$ /GPa                               | 120   |
| Young's modulus of AZO, $E_{AZO}$ /GPa                                    | 80    |
| The radius of nanowires, $R_{NW}$ /nm                                     | 15    |
| The thickness of upper AZO coverage film $t_{AZO}$ / nm (only in model b) | 25    |
| $\delta_m$ for CZM / nm                                                   | 1.5   |
| $\sigma_m$ for CZM / GPa                                                  | 0.2   |
| Toughness of interface 1, $\Gamma_1$ / J/m <sup>2</sup>                   | 0.05  |
| Toughness of interface 2, $\Gamma_2$ / J/m <sup>2</sup>                   | 0.5   |

### 2 Details of the electric potential simulation by COMSOL Multiphysics

The COMSOL Multiphysics commercial software package is used for the electrical characterization of the AAA STE system in this study. The program simulates electrical components (conductivity and permittivity) and devices used in electrostatic applications, particularly in terms of the effects of other physical properties.

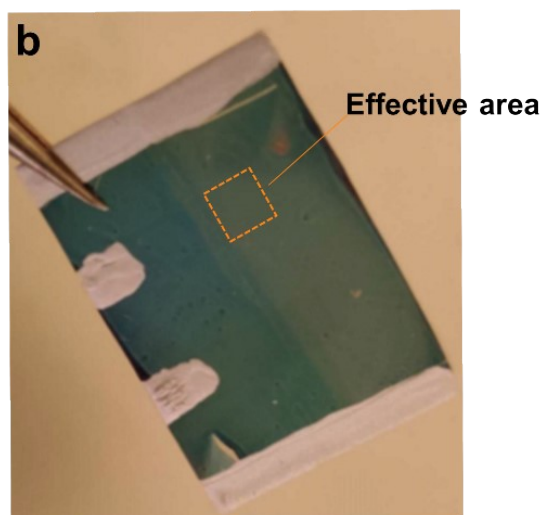
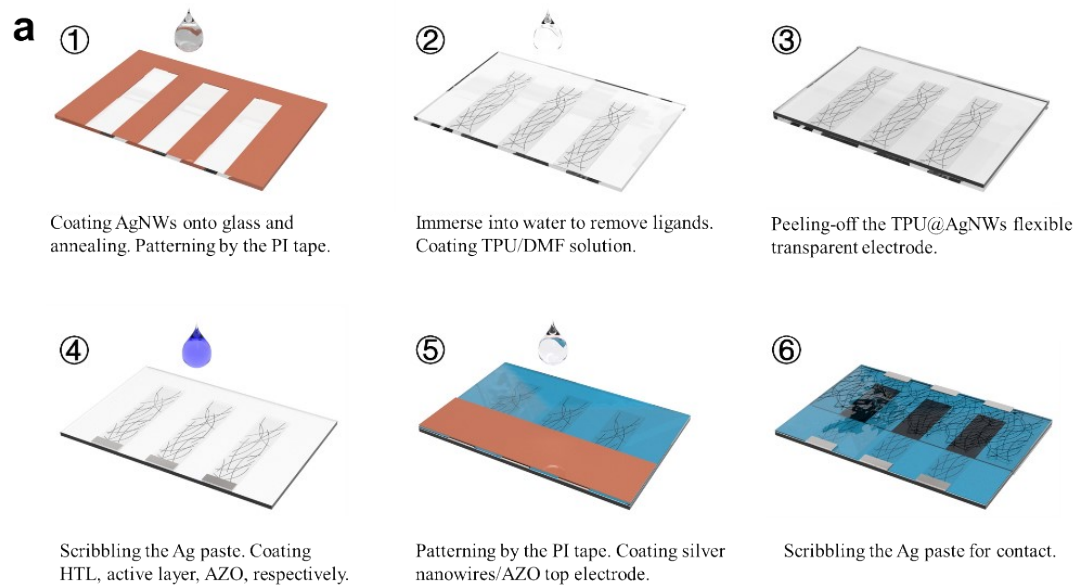
The three-dimensional numerical model used in this study incorporates an AC/DC module (Electric Currents, Stationary). The width, length, and height of the AZO substrate are 1  $\mu\text{m}$ , 1  $\mu\text{m}$ , and 50 nm, respectively. The origin of the coordinate is in the corner of the AZO substrate. Two Ag nanowires cross electrodes (radius: 20 nm) with a central overlap are set on the AZO surface. The conductivity of Ag nanowires and AZO are  $10^4$  S/cm and  $10^{-3}$  S/cm, respectively. The boundary conditions used are meant to represent a connection to a DC source of voltage. The bottom of the AZO substrate is grounded, representing a zero potential point, and the upper surfaces of Ag nanowires are connected to a constant voltage source of  $8 \times 10^{-5}$  V, using the Terminal boundary condition.

To improve the electric potential distribution of impedance measurement, a series of additional AZO coverage with different thicknesses have been proposed in this study. The AZO coverage is set to partially surround or fully surround the cross electrodes and fill the gap between the two Ag nanowires, and the electrical properties are the same as the AZO substrate. The mesh is set to Free Tetrahedral and the predefined accuracy is set to Extra Fine.

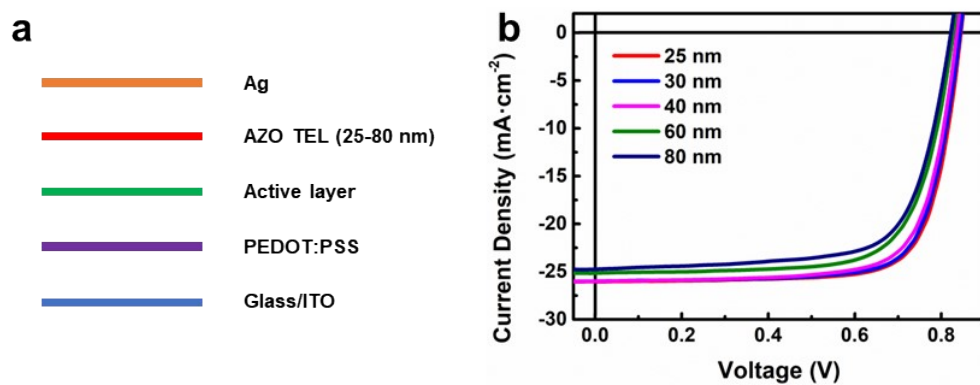
After the computing, the 3D electric potential mapping can be extracted from the convergent model. The 1D potential distribution in the Ag nanowires/AZO substrate interface along the vertical and lateral directions is plotted and the integrated intensity of the whole interface can be calculated by the Boundary Probe.

### **Characterization**

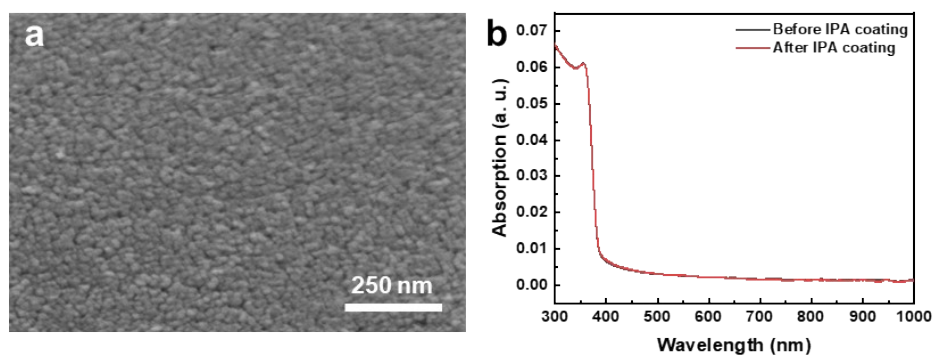
The  $J$ - $V$  curves were measured in the glovebox with a Keithley 2400 unit under 1 sun, AM 1.5G spectra ( $100 \text{ mW} \cdot \text{cm}^{-2}$ ) from a solar simulator (Enli Tech. Co., Ltd., Taiwan). The light intensity was calibrated with a 20 mm  $\times$  20 mm monocrystalline silicon reference cell with a KG5 filter (Enli Tech. Co., Ltd., Taiwan). The EQE spectra were measured by QE-R3-011 (Enli Tech. Co., Ltd., Taiwan). The light intensity at each wavelength was calibrated with a standard single-crystal Si photovoltaic cell. The OPV device used for the mechanical stability test is the translucent device with the bottom illumination under AM 1.5G ( $100 \text{ mW} \cdot \text{cm}^{-2}$ ). The mechanical stability (bending, twisting, and stretching) was performed by a linear reciprocating motor in the glovebox without encapsulation. The sheet resistance was conducted by four-terminal measurement. Transmittance and absorption were measured by Varian Cary® 300 UV-Vis spectrophotometers (Agilent Technologies). Conductive AFM was conducted by a Bruker Dimension ICON. Resistance variation is conducted by an electrochemical workstation (AUTOLAB, PGSTAT 302N).



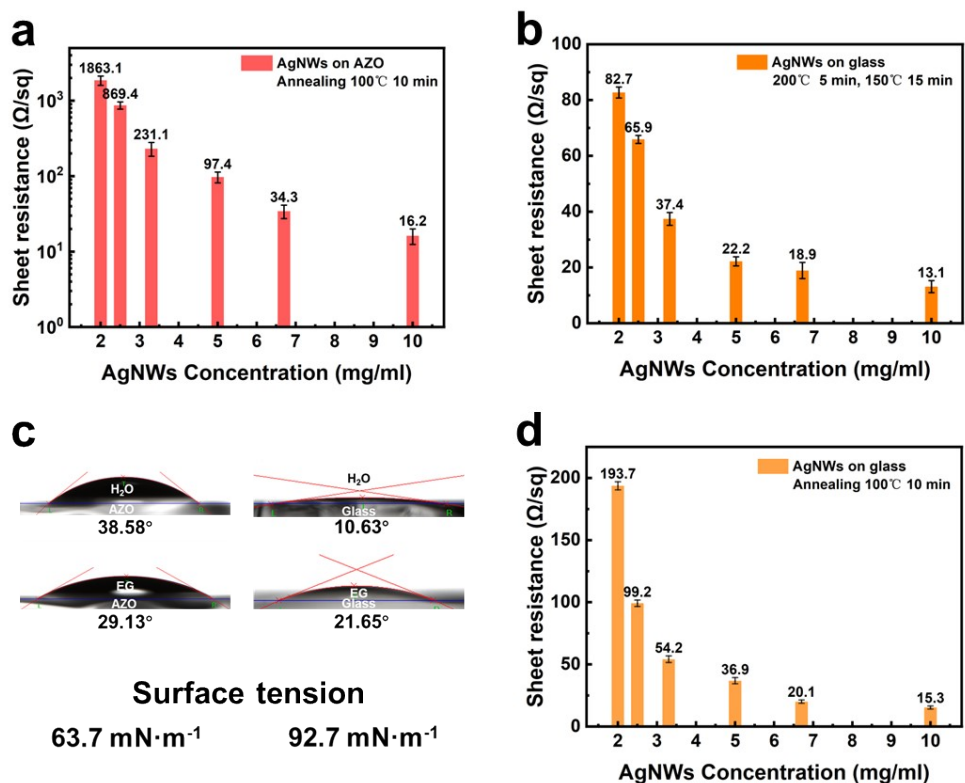
**Figure S1** a) Fabrication of *is*-OPV by full solution process; b) Photograph of the device with the marked effective area.



**Figure S2** a) Device configurations and b)  $J$ - $V$  curves of the AZO-based ETL

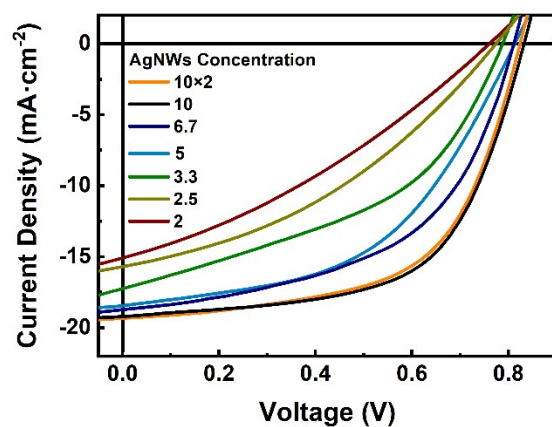


**Figure S3** a) SEM image of the AZO ETL after coating IPA; b) Absorption spectra of AZO films before and after IPA coating.

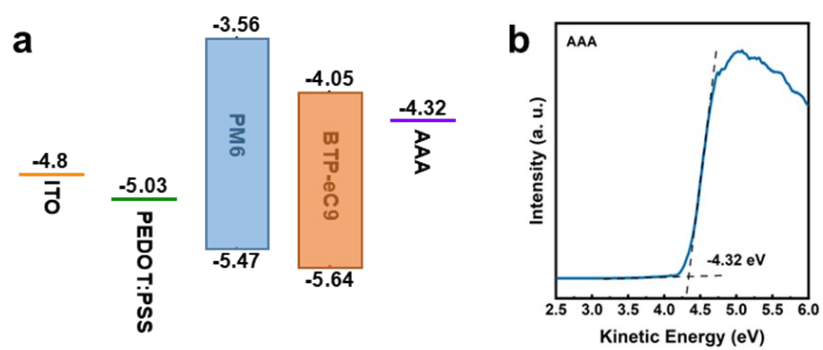


**Figure S4** Sheet resistance of the AgNWs networks coating on a) AZO with annealing condition (100°C/10 min), b) neat glass with annealing condition (200°C/5 min and 150°C/15 min), d) neat glass with annealing condition (100°C/10 min); c) contact angle of H<sub>2</sub>O and ethylene glycol on AZO and neat glass and their surface potential.

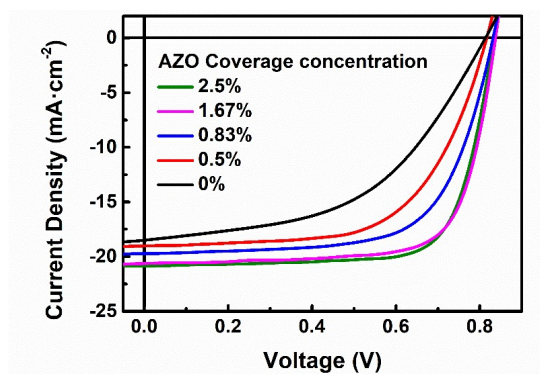




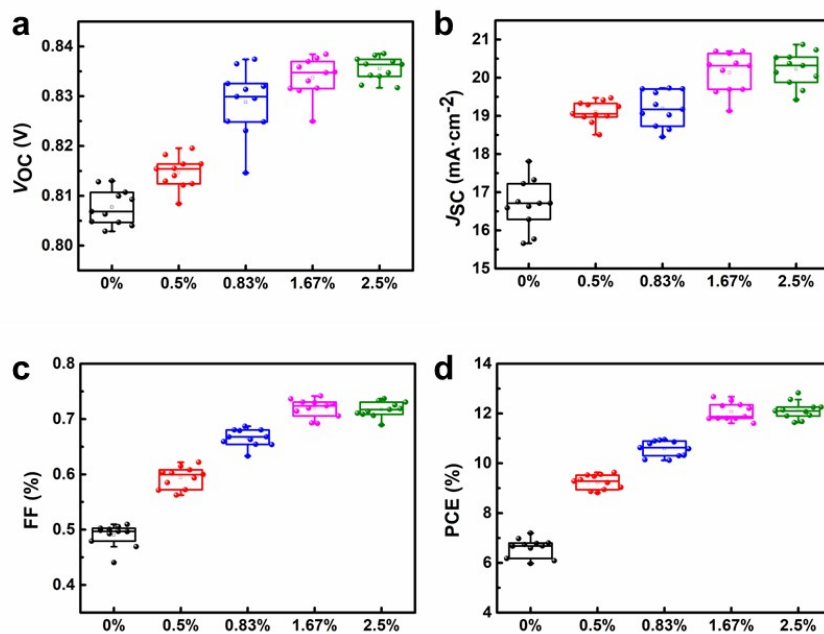
**Figure S5**  $J$ - $V$  curves of the OPV devices based on bare AgNWs back electrode with different AgNWs concentrations



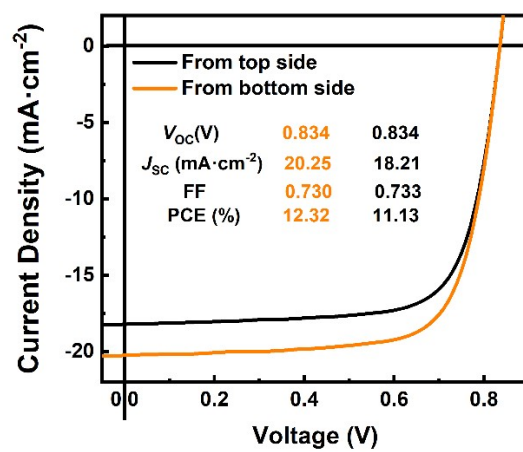
**Figure S6** a) Energy diagram of the device; b) UPS spectra of the AAA electrode.



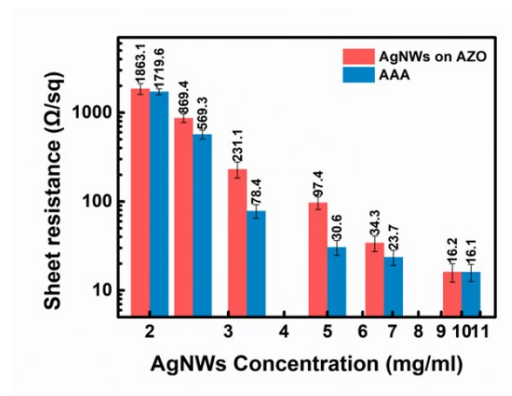
**Figure S7**  $J$ - $V$  curves of the devices based on AAA STE based on AZO coverage with different concentrations.



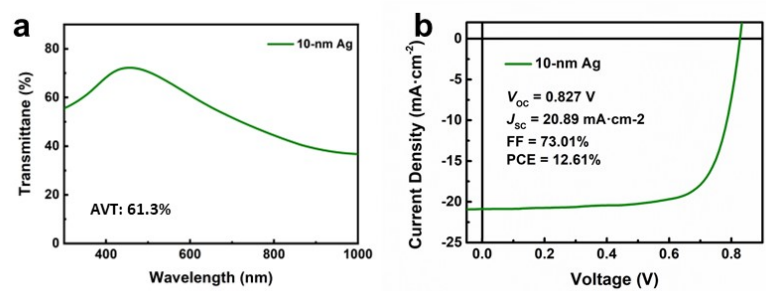
**Figure S8** Device performance. (a)  $V_{OC}$ , (b)  $J_{SC}$ , and (c) FF and (d) PCE parameters comparisons of the devices that deposited with different AZO concentrations with 1000 rpm.



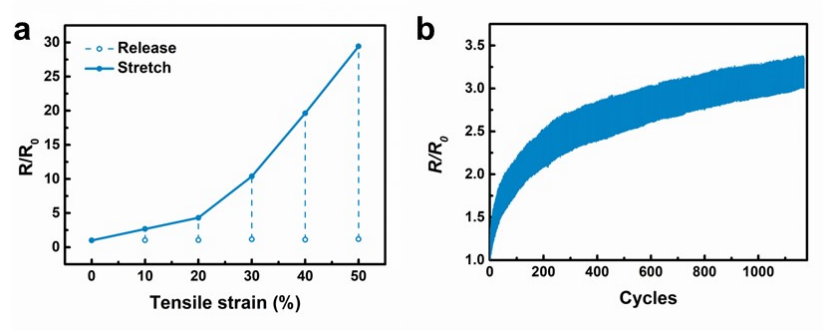
**Figure S9**  $J$ - $V$  curves of the device with the structure of ITO/PEDOT:PSS/PM6:BTP-eC9/AAA when illuminated from the top and the bottom side under simulated AM1.5G, respectively.



**Figure S10** Sheet resistance of AgNWs/AZO and AAA electrodes with different AgNWs concentrations (AZO coverage with a concentration of 2.5%)

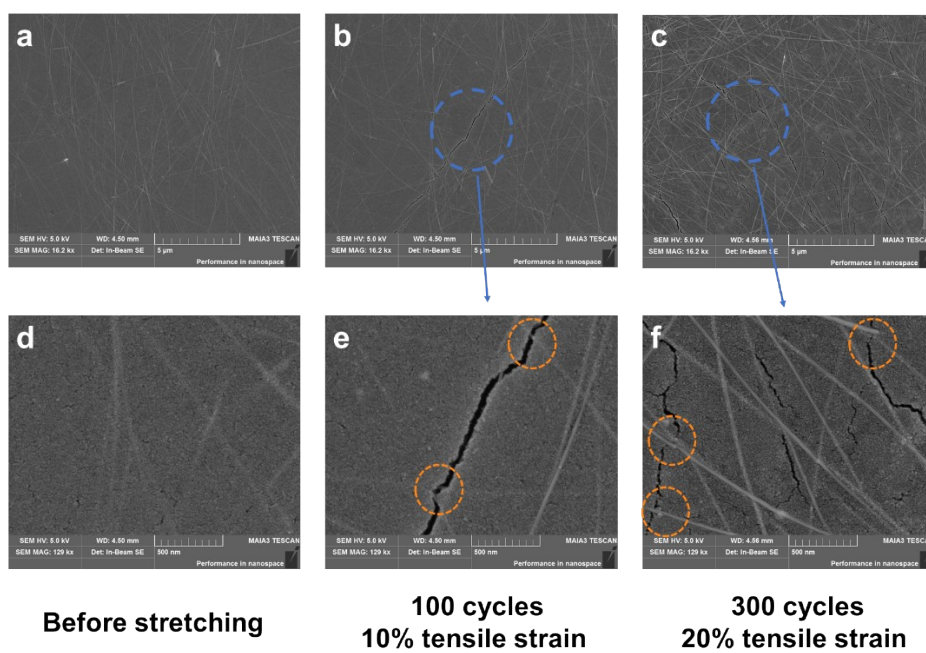


**Figure S11** a) Optical transmittance of the 10-nm Ag electrode fabricated by thermal evaporation;  
b)  $J$ - $V$  curves of the semi-transparent OPV devices based on 10-nm Ag as the back electrode

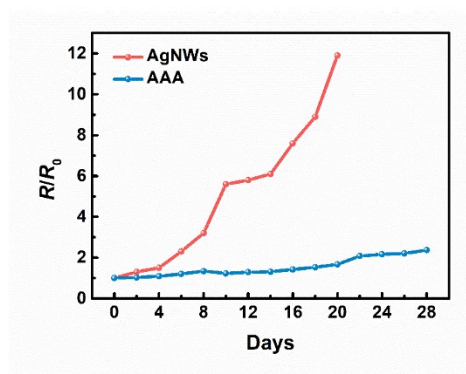


**Figure S12** a) Sheet resistance variation of AAA STE under different tensile strains; b) Long-term mechanical stability under continuous bending with a tensile strain of 20%.

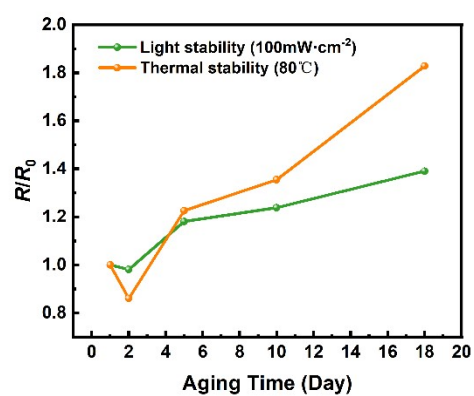




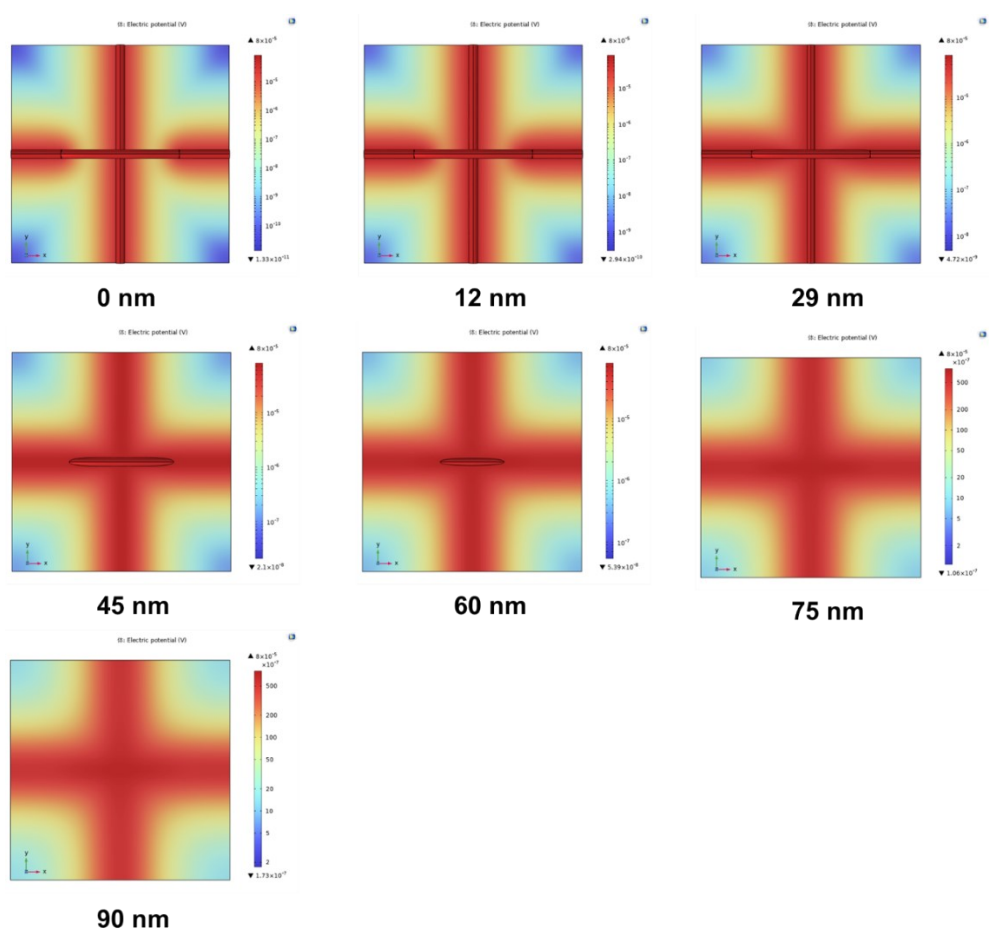
**Figure S13** SEM micrographs of AAA electrode before and after stretch-release cycles. The AgNWs marked by organic circles is not broken after continuous stretch-release cycles.



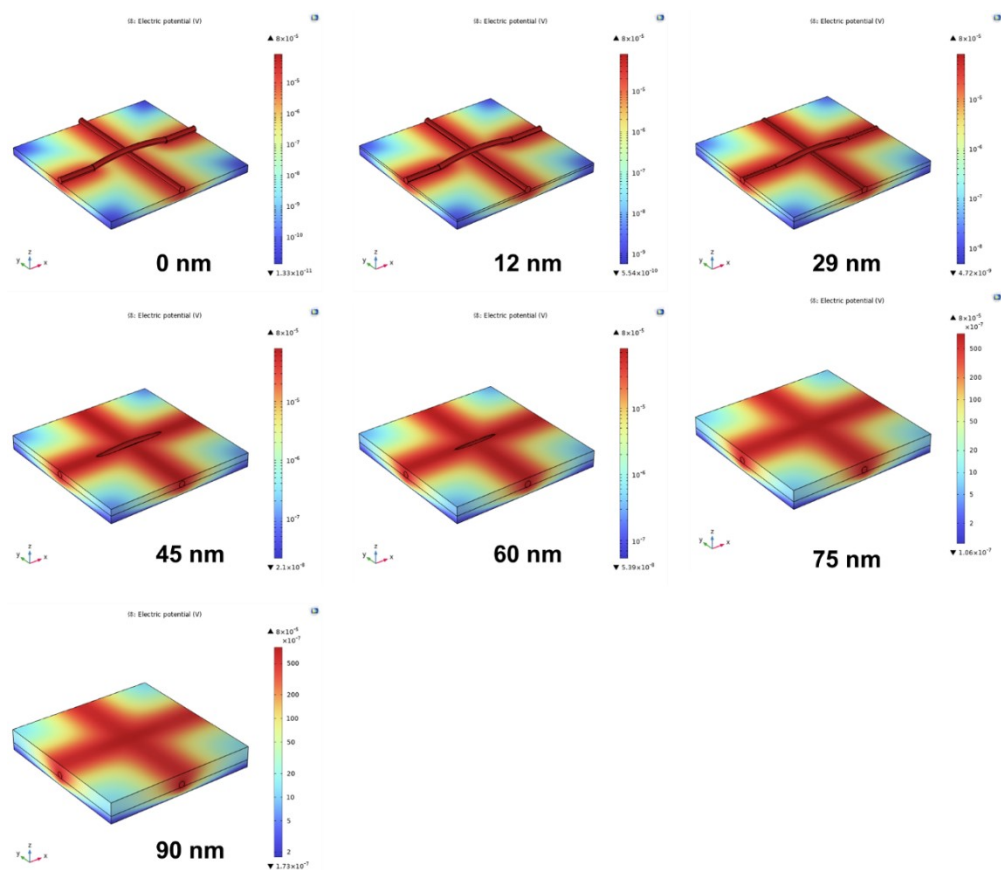
**Figure S14** Air ambient stability of the bare AgNWs and AAA composited STE



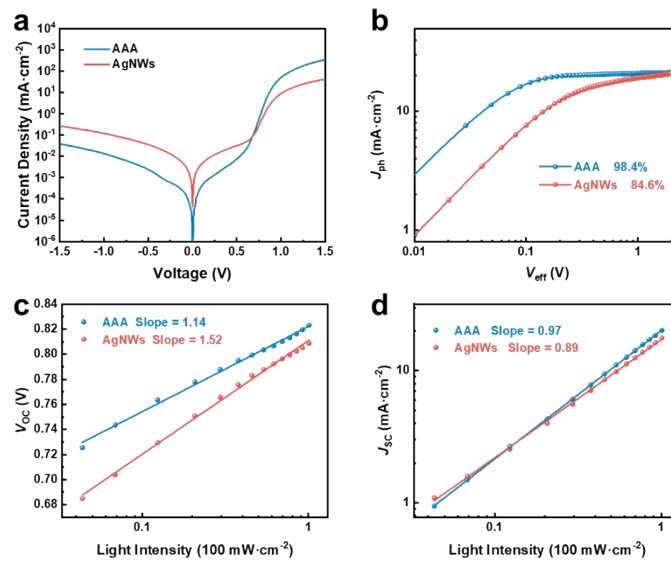
**Figure S15** Light stability and thermal stability of the AAA electrode in ambient conditions.



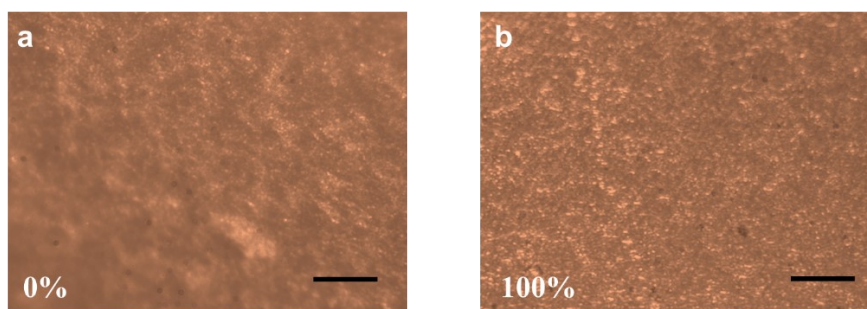
**Figure S16** Top view of the COMSOL simulation of the AAA STE with different thicknesses of AZO coverage



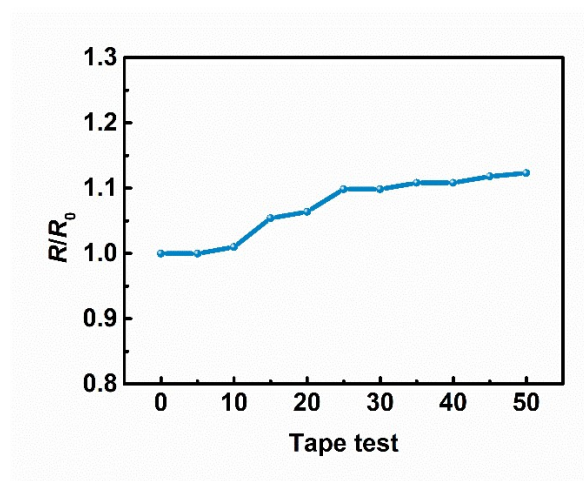
**Figure S17** Perspective view of the COMSOL simulation of the AAA STE with different thickness of AZO coverage



**Figure S18** Photophysical processes of the devices based on AAA and AgNWs back STE. a)  $J$ - $V$  curves under dark conditions; b) Photocurrent *versus* effective voltage; c) Light intensity-dependent  $V_{oc}$  and d) Light intensity-dependent  $J_{sc}$ .

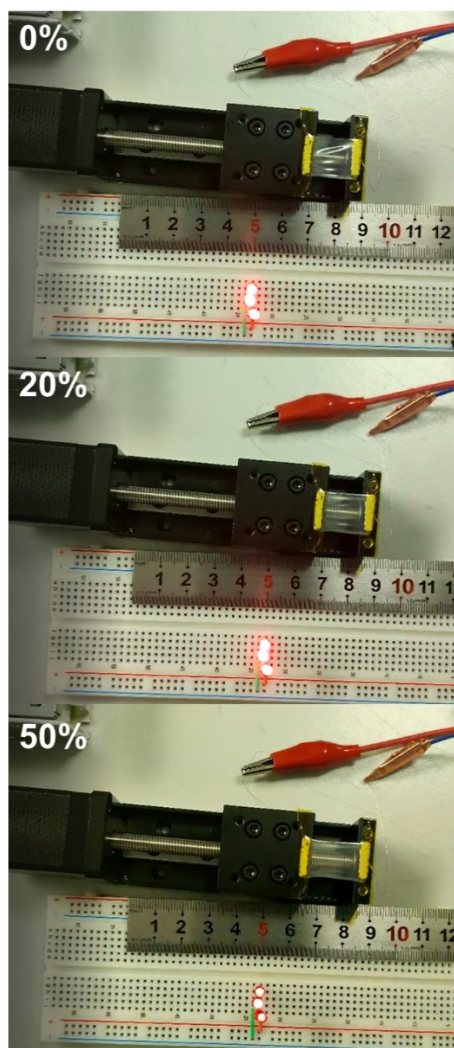


**Figure S19** Images of TPU @AgNWs in microscope under a) 0% tensile strain and b) 100% tensile strain (Scale bar: 150  $\mu\text{m}$ )

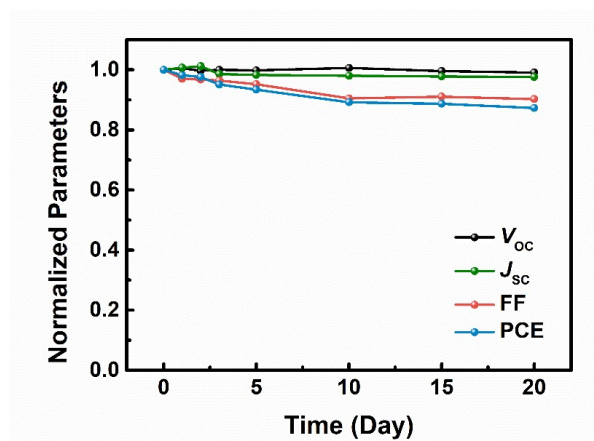


**Figure S20** Sheet resistance variation of the TPU@AgNWs after tape test

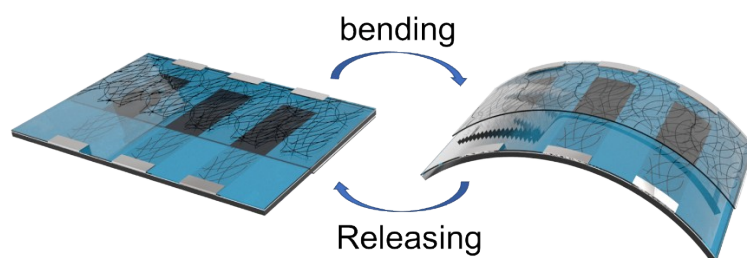




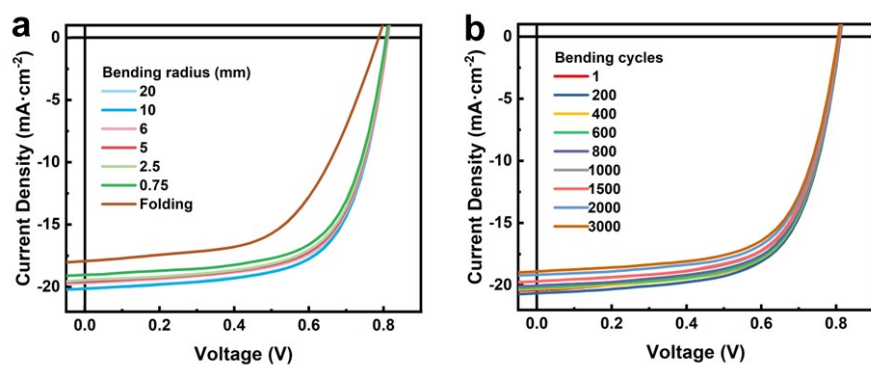
**Figure S21** Stretched TPU@AgNWs serve as the interconnect to drive commercial LED



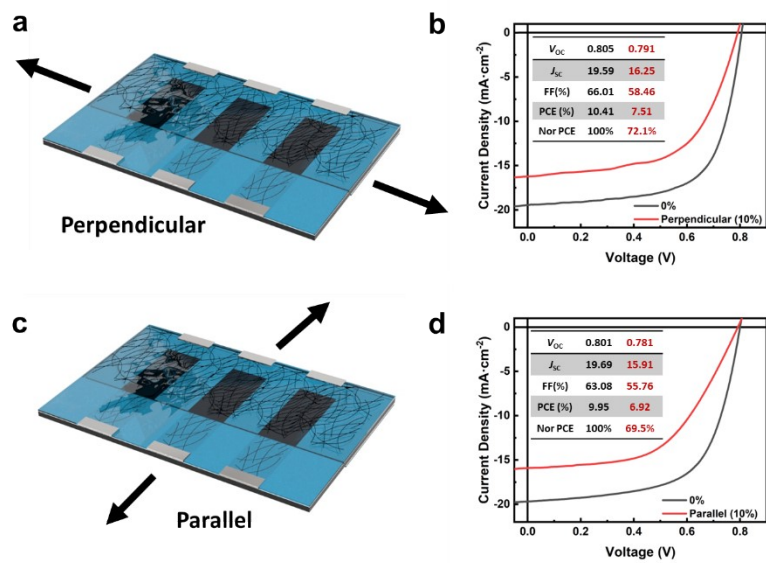
**Figure S22** The shelf lifetime of the *is*-OPV device



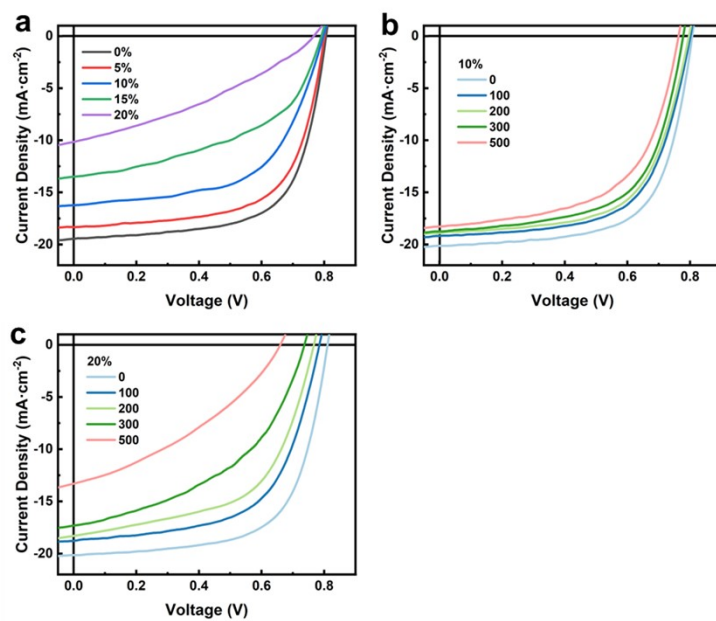
**Figure S23** The schematic diagram of the device bending direction.



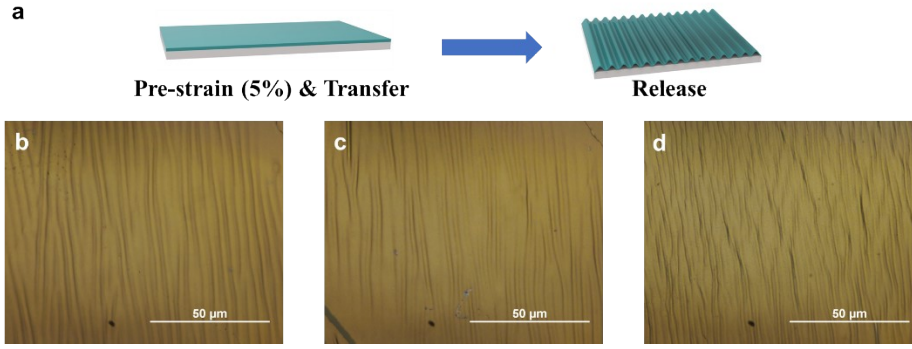
**Figure S24** The representative  $J$ - $V$  curves of the devices based on PM6:BTP-eC9 a) after 200 cycles of continuous bending with different curvature radii, b) 3000 cyclic bending-flattening processes with a 1.5 mm curvature radius.



**Figure S25** Schematic diagrams of the stretching direction. a) perpendicular and b) parallel directions; The  $J$ - $V$  curves of the devices based on PM6:BTP-eC9 under tensile strain with c) perpendicular and d) parallel directions.



**Figure S26** The representative  $J$ - $V$  curves of the devices based on PM6:BTP-eC9 a) under different tensile strains, b) after continuous stretch-release cycles with 10% tensile strain, c) after continuous stretch-release cycles with 20% tensile strain.

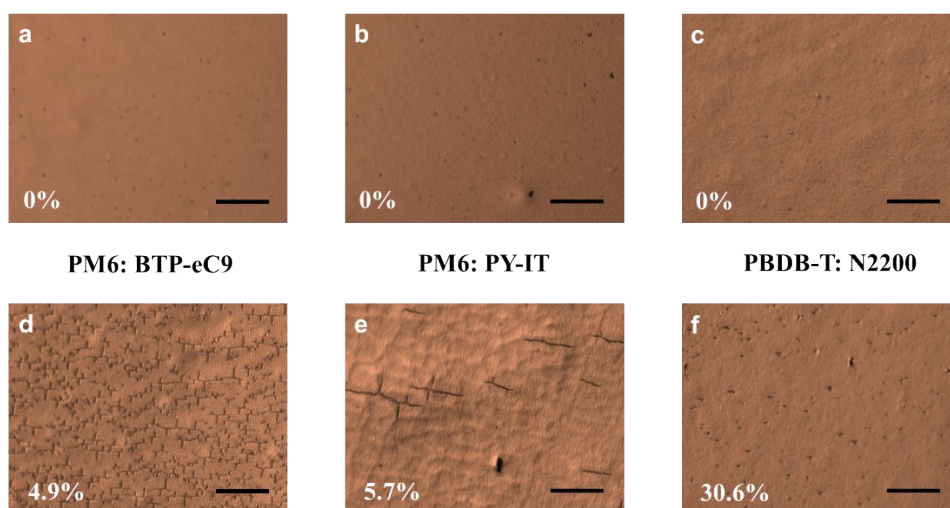


**Figure S27** a) The process of the buckling methods; Surface buckling of the active layers fabricated on PDMS b) PM6:BTP-eC9, c) PM6: PY-IT, d) PBDB-T:N2200

The organic active layers were fabricated on the elastomer PDMS by transfer. A small (<5%) compression was then applied to the PDMS-based samples and there would be buckled with a specific wavelength. Then the elastic modulus of the target films was calculated from the following equations<sup>3, 4</sup>:

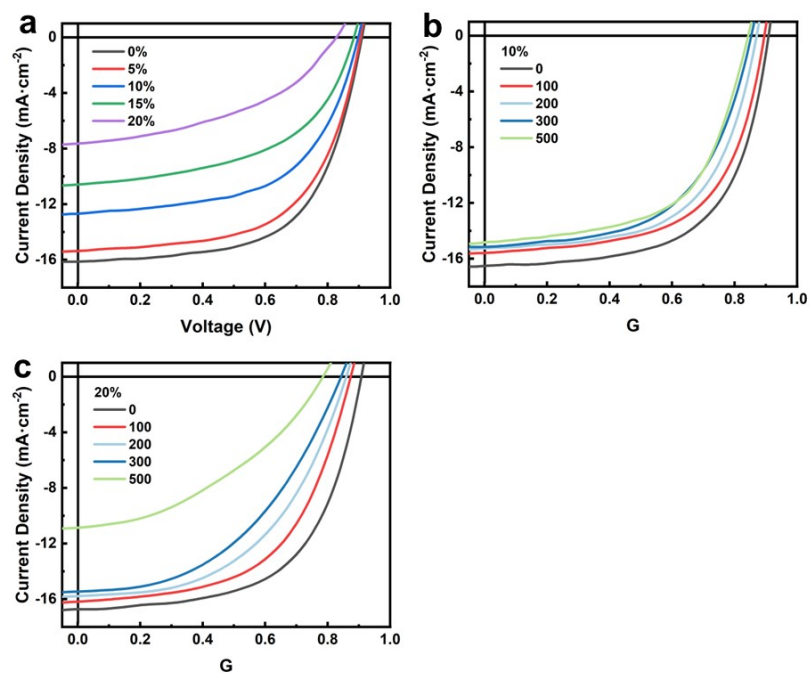
$$E_f = 3E_s \left( \frac{1 - \nu_f^2}{1 - \nu_s^2} \right) \left( \frac{\lambda}{2\pi d_f} \right)^3$$

Where the  $E_f$  is the elastic modulus of target films,  $E_s$  is the elastic modulus of substrate PDMS (1.8 MPa),  $\nu_f$  is the Poisson ratios of the target films (0.35),  $\nu_s$  is the Poisson ratios of the substrate (0.5),  $\lambda$  is the wavelength of the buckling and  $d_f$  is the thickness of films.

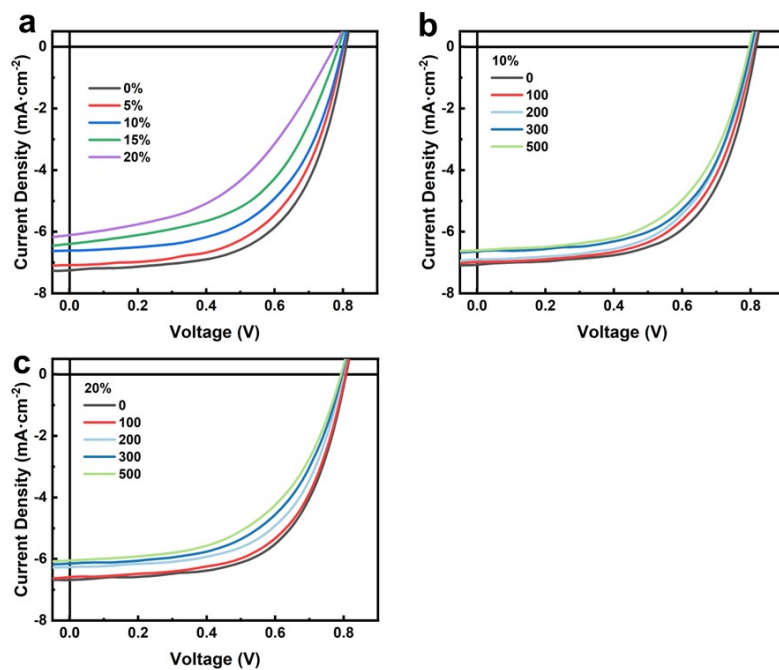


**Figure S28** COS of the active layers (Scale bar: 150  $\mu\text{m}$ )

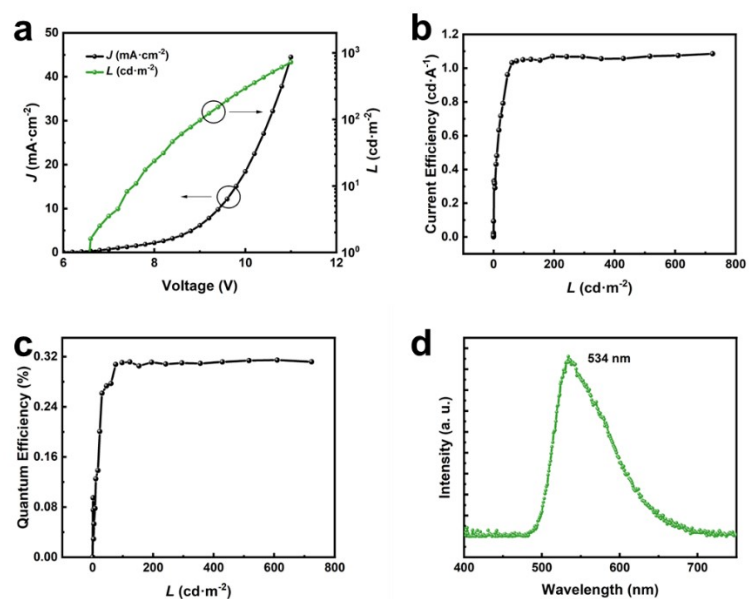




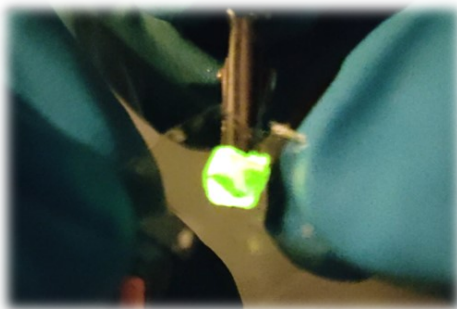
**Figure S29** The representative  $J$ - $V$  curves of the devices based on PM6:PY-IT a) under different tensile strains, b) after continuous stretch-release cycles with 10% tensile strain, c) after continuous stretch-release cycles with 20% tensile strain.



**Figure S30** The representative  $J$ - $V$  curves of the devices based on PBDB-T:N2200 a) under different tensile strains, b) after continuous stretch-release cycles with 10% tensile strain, c) after continuous stretch-release cycles with 20% tensile strain.



**Figure S31** a) Current density and luminance - voltage curves, b) current efficiency - luminance curve, c) quantum efficiency – luminance curve, d) emission spectra of the *is*-OLED device.



**Figure S32** Photograph of the OLED device at an external bias of 9V under tensile strain by a probe poking

**Table S1** Parameters of the devices based on AZO with different thickness\*.

| AZO thickness | $V_{OC}$ (V)        | $J_{SC}$ (mA·cm <sup>-2</sup> ) | FF                  | PCE (%)            |
|---------------|---------------------|---------------------------------|---------------------|--------------------|
| 25            | 0.844 (0.838±0.008) | 26.06 (25.83±0.23)              | 0.765 (0.759±0.004) | 16.83 (16.42±0.22) |
| 30            | 0.843 (0.837±0.008) | 26.01 (25.57±0.28)              | 0.763 (0.760±0.004) | 16.73 (16.27±0.26) |
| 40            | 0.838 (0.831±0.010) | 25.84 (25.18±0.41)              | 0.743 (0.739±0.003) | 16.09 (15.46±0.32) |
| 60            | 0.829 (0.820±0.007) | 25.17 (24.54±0.39)              | 0.727 (0.722±0.004) | 15.17 (14.53±0.29) |
| 80            | 0.822 (0.817±0.006) | 24.76 (23.91±0.36)              | 0.709 (0.702±0.004) | 14.43 (13.72±0.31) |

\*The average values and the standard deviation were obtained from ten independent cells.

**Table S2** Parameters of devices based on AgNWs back electrode with different concentrations\*.

| AgNWs concentration | $V_{oc}$ (V)        | $J_{sc}$ (mA·cm <sup>-2</sup> ) | FF                  | PCE (%)          |
|---------------------|---------------------|---------------------------------|---------------------|------------------|
| 2                   | 0.761 (0.742±0.012) | 15.08 (14.96±0.46)              | 0.326 (0.303±0.019) | 3.74 (3.36±0.21) |
| 2.5                 | 0.774(0.761±0.015)  | 15.71 (15.31±0.56)              | 0.375 (0.335±0.025) | 4.56 (3.90±0.28) |
| 3.3                 | 0.787(0.776±0.010)  | 17.96 (17.37±0.40)              | 0.440 (0.403±0.029) | 6.22 (5.43±0.51) |
| 5                   | 0.811(0.795±0.015)  | 18.51 (17.92±0.43)              | 0.497 (0.447±0.031) | 7.46 (6.37±0.57) |
| 6.7                 | 0.813(0.799±0.012)  | 18.72 (18.13±0.50)              | 0.527 (0.509±0.031) | 8.02 (7.37±0.02) |
| 10                  | 0.830 (0.816±0.012) | 19.20 (18.89±0.40)              | 0.603 (0.591±0.015) | 9.61 (9.11±0.29) |
| 10×2                | 0.824(0.822±0.010)  | 19.34 (19.07±0.36)              | 0.596 (0.579±0.025) | 9.50 (9.08±0.39) |

\*The average values and the standard deviation were obtained from ten independent cells.

**Table S3** Parameters of the devices based on AAA back electrode with different AZO coverage concentrations (AgNWs with 5 mg/ml)\*.

| AZO (wt. %) | $V_{oc}$ (V)        | $J_{sc}$ (mA·cm <sup>-2</sup> ) | FF                  | PCE (%)            |
|-------------|---------------------|---------------------------------|---------------------|--------------------|
| 2.5         | 0.834 (0.835±0.002) | 20.87 (20.23±0.45)              | 0.737 (0.722±0.014) | 12.83 (12.12±0.32) |
| 1.67        | 0.837 (0.834±0.004) | 20.63 (20.13±0.52)              | 0.730 (0.720±0.017) | 12.61 (12.07±0.35) |
| 0.83        | 0.832 (0.829±0.006) | 19.74 (19.19±0.46)              | 0.663 (0.666±0.016) | 10.89 (10.59±0.32) |
| 0.5         | 0.812 (0.814±0.003) | 19.04 (19.10±0.28)              | 0.615 (0.594±0.019) | 9.55 (9.25±0.29)   |
| 0           | 0.813 (0.808±0.004) | 17.82 (16.68±0.63)              | 0.497 (0.490±0.020) | 7.20 (6.61±0.28)   |

\*The average values and the standard deviation were obtained from ten independent cells.

**Table S4** Parameters of the devices based on TPU@AgNWs with different AgNWs properties\*.

| $T_{\text{avg}}$<br>(%)** | $R_{\text{sh}}$ ( $\Omega/\text{sq}$ ) | $V_{\text{OC}}$ (V)       | $J_{\text{SC}}$ ( $\text{mA}\cdot\text{cm}^{-2}$ ) | FF                        | PCE (%)                  |
|---------------------------|----------------------------------------|---------------------------|----------------------------------------------------|---------------------------|--------------------------|
| 88.2                      | 68.1                                   | 0.817 (0.812 $\pm$ 0.007) | 25.17 (24.93 $\pm$ 0.32)                           | 0.703 (0.688 $\pm$ 0.027) | 14.46 (14.02 $\pm$ 0.35) |
| 83.1                      | 27.3                                   | 0.831 (0.824 $\pm$ 0.004) | 25.83 (25.26 $\pm$ 0.41)                           | 0.739 (0.718 $\pm$ 0.019) | 15.86 (15.19 $\pm$ 0.47) |
| 70.7                      | 15.8                                   | 0.834 (0.827 $\pm$ 0.003) | 21.85 (22.19 $\pm$ 0.48)                           | 0.743 (0.725 $\pm$ 0.021) | 13.54 (13.16 $\pm$ 0.33) |

\*The device structure is TPU@AgNWs/PEDOT:PSS/PM6:BTP-eC9:PFNBr/Ag.

\*The average values and the standard deviation were obtained from ten independent cells.

\*\*Average optical transmittance between 300-1000 nm.



**Table S5** Parameters of the devices based on PM6:BTP-eC9 after 200 cycles of continuous bending with different curvature radii.

| Curvature radius<br>(mm) | $V_{OC}$ (V) | $J_{SC}$ (mA·cm <sup>-2</sup> ) | FF<br>(%) | PCE (%) |
|--------------------------|--------------|---------------------------------|-----------|---------|
| 20                       | 0.811        | 20.13                           | 65.86     | 10.75   |
| 10                       | 0.811        | 20.15                           | 66.16     | 10.81   |
| 6                        | 0.811        | 19.69                           | 66.15     | 10.57   |
| 5                        | 0.809        | 19.55                           | 65.89     | 10.43   |
| 2.5                      | 0.808        | 19.44                           | 65.72     | 10.33   |
| 0.75                     | 0.807        | 19.04                           | 65.37     | 10.04   |
| Folding                  | 0.786        | 17.81                           | 58.79     | 8.23    |

**Table S6** Parameters of the devices based on PM6:BTP-eC9 after continuous bending cycles with a 1.5 mm radius.

| Bending cycles | $V_{oc}$ (V) | $J_{sc}$ (mA·cm <sup>-2</sup> ) | FF<br>(%) | PCE (%) |
|----------------|--------------|---------------------------------|-----------|---------|
| 0              | 0.811        | 20.37                           | 65.87     | 10.88   |
| 200            | 0.810        | 20.65                           | 65.79     | 11.01   |
| 400            | 0.809        | 20.30                           | 66.07     | 10.85   |
| 600            | 0.809        | 20.21                           | 66.10     | 10.80   |
| 800            | 0.808        | 20.04                           | 65.94     | 10.68   |
| 1000           | 0.809        | 19.69                           | 65.82     | 10.48   |
| 1500           | 0.808        | 19.70                           | 65.46     | 10.41   |
| 2000           | 0.806        | 19.17                           | 65.58     | 10.13   |
| 3000           | 0.805        | 18.90                           | 65.21     | 9.92    |

**Table S7** Parameters of the devices based on different active layers under different tensile strain.

| Active layers    | Tensile strain<br>(%) | $V_{oc}$ (V) | $J_{sc}$ (mA·cm <sup>-2</sup> ) | FF<br>(%) | PCE (%) |
|------------------|-----------------------|--------------|---------------------------------|-----------|---------|
| PM6:BTP-eC9      | 0                     | 0.805        | 19.59                           | 65.86     | 10.38   |
|                  | 5                     | 0.800        | 18.33                           | 64.51     | 9.46    |
|                  | 10                    | 0.797        | 16.25                           | 58.14     | 7.53    |
|                  | 15                    | 0.791        | 13.52                           | 48.06     | 5.14    |
|                  | 20                    | 0.766        | 10.79                           | 38.60     | 3.19    |
| PM6:PY-IT        | 0                     | 0.909        | 16.13                           | 61.90     | 9.08    |
|                  | 5                     | 0.905        | 15.37                           | 60.20     | 8.37    |
|                  | 10                    | 0.898        | 12.68                           | 58.38     | 6.64    |
|                  | 15                    | 0.884        | 10.58                           | 53.52     | 5.01    |
|                  | 20                    | 0.828        | 7.83                            | 43.90     | 2.84    |
| PBDB-T:<br>N2200 | 0                     | 0.808        | 7.28                            | 59.67     | 3.51    |
|                  | 5                     | 0.800        | 7.08                            | 58.01     | 3.29    |
|                  | 10                    | 0.799        | 6.70                            | 55.67     | 2.98    |
|                  | 15                    | 0.787        | 6.42                            | 52.45     | 2.65    |
|                  | 20                    | 0.775        | 6.33                            | 44.23     | 2.17    |

**Table S8** Parameters of the devices based on different active layers after continuous cycles of 10% tensile strain.

| Active layers    | Cycles | $V_{OC}$ (V) | $J_{SC}$ (mA·cm <sup>-2</sup> ) | FF (%) | PCE (%) |
|------------------|--------|--------------|---------------------------------|--------|---------|
| PM6:BTP-eC9      | 0      | 0.807        | 20.13                           | 65.53  | 10.65   |
|                  | 100    | 0.801        | 19.19                           | 63.17  | 9.71    |
|                  | 200    | 0.796        | 18.91                           | 62.66  | 9.43    |
|                  | 300    | 0.778        | 18.76                           | 61.92  | 9.04    |
|                  | 500    | 0.780        | 18.28                           | 58.17  | 8.29    |
| PM6:BTP-eC9      | 0      | 0.908        | 16.52                           | 61.85  | 9.28    |
|                  | 100    | 0.895        | 15.59                           | 60.92  | 8.50    |
|                  | 200    | 0.87         | 15.26                           | 60.32  | 8.01    |
|                  | 300    | 0.853        | 15.15                           | 58.21  | 7.52    |
|                  | 500    | 0.843        | 14.97                           | 57.40  | 7.25    |
| PBDB-T:<br>N2200 | 0      | 0.816        | 7.07                            | 61.78  | 3.56    |
|                  | 100    | 0.809        | 6.98                            | 60.18  | 3.40    |
|                  | 200    | 0.806        | 6.88                            | 59.30  | 3.29    |
|                  | 300    | 0.798        | 6.63                            | 59.87  | 3.17    |
|                  | 500    | 0.796        | 6.62                            | 58.22  | 3.07    |

**Table S9** Parameters of the devices based on different active layers after continuous cycles of 20% tensile strain.

| Active layers    | Cycles | $V_{oc}$ (V) | $J_{sc}$ (mA·cm <sup>-2</sup> ) | FF (%) | PCE (%) |
|------------------|--------|--------------|---------------------------------|--------|---------|
| PM6:BTP-eC9      | 0      | 0.810        | 20.16                           | 64.80  | 10.58   |
|                  | 100    | 0.784        | 18.81                           | 60.21  | 8.88    |
|                  | 200    | 0.764        | 18.53                           | 55.02  | 7.79    |
|                  | 300    | 0.737        | 17.29                           | 46.57  | 5.94    |
|                  | 500    | 0.659        | 14.35                           | 36.13  | 3.42    |
| PM6:BTP-eC9      | 0      | 0.907        | 16.73                           | 59.50  | 9.03    |
|                  | 100    | 0.874        | 16.18                           | 53.12  | 7.51    |
|                  | 200    | 0.854        | 15.77                           | 50.34  | 6.78    |
|                  | 300    | 0.842        | 15.46                           | 45.59  | 5.93    |
|                  | 500    | 0.782        | 10.86                           | 37.00  | 3.14    |
| PBDB-T:<br>N2200 | 0      | 0.807        | 6.82                            | 61.81  | 3.40    |
|                  | 100    | 0.805        | 6.59                            | 59.02  | 3.13    |
|                  | 200    | 0.797        | 6.26                            | 59.31  | 2.96    |
|                  | 300    | 0.796        | 6.14                            | 57.40  | 2.81    |
|                  | 500    | 0.792        | 6.05                            | 54.92  | 2.63    |

**Appendix S1** Summary of previous work about solution processed top electrode (ITO as bottom electrode)

| Year | Top electrode         | Device configuration                                                                                        | Device area (cm <sup>2</sup> ) | PCE (%) | Reference     |
|------|-----------------------|-------------------------------------------------------------------------------------------------------------|--------------------------------|---------|---------------|
| 2021 | AgNWs                 | Glass/ITO/PEDOT:PSS/PM6:Y6/PDINO/Ag NWs                                                                     | 0.0314                         | 12.1    | <sup>5</sup>  |
| 2021 | AgNWs                 | Glass/ITO/ZnO/ PBDTTT-EFT:PCBM/s-MoO <sub>3</sub> /AgNWs                                                    | -                              | 4.19    | <sup>6</sup>  |
| 2021 | AgNWs                 | Glass/ITO/PEDOT:PSS/PTB7-Th:IEICO-4F/AzO//AgNWs                                                             | 0.2                            | 8.8     | <sup>7</sup>  |
| 2019 | PH1000/AI4083/EG      | Glass/ITO/ZnO/P3HT:PCBM/ PH1000/AI4083/EG                                                                   | 0.04                           | 2.04    | <sup>8</sup>  |
| 2021 | AgNPs/HMO<br>(Opaque) | Glass/ITO/PEDOT:PSS/PM6:Y6/ZnO/AgNPs/HMO                                                                    | -                              | 15      | <sup>9</sup>  |
| 2021 | AgCNTs                | Glass/ITO/ZnO/J71:Y6:PC71CM/MoO <sub>3</sub> / AgCNTs                                                       | -                              | 9.54    | <sup>10</sup> |
| 2021 | PH1000                | Glass/ITO/PMA-Im-PTB7-Th:IEICO-4F/PH1000                                                                    | 0.04                           | 10.37   | <sup>11</sup> |
| 2020 | AgNWs                 | Glass/ITO/ZnO/PM6:Y6/PEDOT:PSS/ AgNWs                                                                       | -                              | 9.12    | <sup>12</sup> |
| 2020 | AgNWs-20              | Glass/ITO/ZnO/PBDB-T-2Cl:IT-4F/MoO <sub>3</sub> / AgNWs                                                     |                                | 6.44    | <sup>13</sup> |
| 2020 | AgNPs                 | Glass/ITO/PEDOT:PSS/PBDB-T:IT-M/ZnO/Zr(acac) <sub>4</sub> / AgNPs                                           | -                              | 9.73    | <sup>14</sup> |
| 2019 | Ag paste (Opaque)     | Glass/ITO/ZnO/SMD2:ITIC-Th/Bilayer HTLs/Ag paste                                                            | 0.04                           | 8.0     | <sup>15</sup> |
| 2018 | PH1000                | Glass/ITO/ZnO/FTAZ:IT-M/MoO <sub>3</sub> /PEDOT:PSS                                                         | 0.025                          | 5.54    | <sup>16</sup> |
| 2018 | AgNWs                 | Glass/ITO/ZnO/PTB7-Th:PC71BM/PMA/PEDOT:PSS/AgNWs                                                            | -                              | 5.01    | <sup>17</sup> |
| 2018 | PH1000                | Glass/ITO/ZnO/ PBTZT-stat-BDTT8:PCBM/ PEDOT:PSS                                                             | 0.3                            | 2.9     | <sup>18</sup> |
|      |                       |                                                                                                             | 4                              | 2.44    | <sup>18</sup> |
| 2017 | PH1000                | Glass/ITO/PEIE/PTB7:PC71BM/PEDOT:PSS/PDMS                                                                   | 0.045                          | 3.75    | <sup>19</sup> |
| 2016 | AgNWs                 | Glass/ITO/PEDOT:PSS/BDTT-S-TR:PCBM/ZnO/<br>PDINO/AgNWs                                                      | -                              | 3.62    | <sup>20</sup> |
| 2015 | PH1000:PEG-TmDD       | Glass/ITO/PEI/P3HT:ICBA/ PH1000:PEG-TmDD                                                                    | -                              | 4.1     | <sup>21</sup> |
| 2015 | PH1000                | Glass/LWF-PEDOT:PSS/P3HT:ICBA/HWF-PEDOT:PSS                                                                 | -                              | 4.0     | <sup>22</sup> |
| 2015 | PH1000                | Glass/ITO/PEI/P3HT:ICBA/PH1000/PEI/P3HT:ICBA/PEDOT-T                                                        | 0.05                           | 3.6     | <sup>23</sup> |
| 2014 | PH1000                | Glass/ITO/ZnO/P3HT:ICBA/PEDOT:PSS                                                                           | -                              | 4.0     | <sup>24</sup> |
| 2013 | AgNWs                 | Glass/ITO/PBDTT-SeDPP:PC71BM/TiO <sub>2</sub> /AgNWs                                                        | -                              | 5.5     | <sup>25</sup> |
|      |                       | Glass/ITO/ PBDTT-FDPP-C12:PC61BM/PFN/TiO <sub>2</sub> /PEDOT/<br>PBDTT-SeDPP:PC71BM/TiO <sub>2</sub> /AgNWs | -                              | 7.3     |               |

## Appendix S2 Summary of previous work about all-solution processed organic solar cells.

| Year                                                              | Substrate                    | Device configuration                                               | Device area<br>(cm <sup>2</sup> ) | PCE (%)               | Reference |
|-------------------------------------------------------------------|------------------------------|--------------------------------------------------------------------|-----------------------------------|-----------------------|-----------|
| 2022                                                              | Glass<br>(Rigid)             | Glass/AgNWs/PEI-Zn/PM6:PC <sub>71</sub> BM:Y6/PEDOT:F/AgNWs        | 0.041                             | 12.83                 | 26        |
|                                                                   |                              |                                                                    | 14.62 (with mirror)               |                       |           |
|                                                                   |                              | 12.2                                                               | 15.26                             |                       |           |
|                                                                   |                              | (modulus)                                                          |                                   |                       |           |
| Glass/AgNWs/PEI-Zn/ PM6:PC <sub>71</sub> BM:BTP-eC9/PEDOT:F/AgNWs |                              |                                                                    | 0.041                             | 15.26 (with mirror)   |           |
| 2021                                                              | TPU<br>(Stretchable/Opaque)  | TPU/PH1000/AI4083/PM6:Y7/PNDIT-F3N-Br/EGaIn                        | 0.0625                            | 11.2                  | 27        |
| 2021                                                              | TPU<br>(Stretchable/Opaque)  | TPU@AgNWs/PEDOT:PSS/PTB7-Th:IEICO-4F:PDMS/EGaIn                    | -                                 | 10.3                  | 28        |
| 2021                                                              | PDMS<br>(Stretchable/Opaque) | PDMS/PH1000/PBDB-T:PCE10:N2200/EGaIn                               | 0.04                              | 6.33                  | 29        |
| 2020                                                              | Polyimide<br>(Flexible)      | AgNWs@PI/ZnO/PBDB-T-2F:IDIC:Y6/HMoO3/PEDOT:PSS/AgNWs               | 0.04                              | 11.9<br>(with mirror) | 30        |
|                                                                   |                              |                                                                    | 1                                 | 10.3<br>(with mirror) | 30        |
| 2020                                                              | PET<br>(Flexible)            | PH1000/AI4083/PM6:Y6/ZnO/PH1000                                    | 0.06                              | 11.12                 | 31        |
| 2018                                                              | Polyurethane<br>(Flexible)   | Ag/ZnO np/ P3HT:ICBA/ PEDOT:PSS/ AgNWs                             | 0.03                              | 1.71                  | 32        |
| 2018                                                              | PET<br>(Flexible)            | PAMD-Ag/PEI/P3HT:PC61BM/PEDOT:PSS                                  | -                                 | 3.03                  | 33        |
| 2017                                                              | Glass<br>(Rigid)             | Ag ink/ZnO/PBTZT-stat-BDIT-8:PCBM/<br>PEDOT:PSS:Ag NW              | 1.04                              | 4.5                   | 34        |
|                                                                   |                              |                                                                    | 19.7<br>(Module)                  | 3.6                   |           |
|                                                                   | PET<br>(Flexible)            |                                                                    | 17.7<br>(Module)                  | 2.9                   |           |
|                                                                   |                              |                                                                    |                                   |                       |           |
| 2016                                                              | PES<br>(Flexible)            | PEDOT:PSS/PEIE/P3HT:IDT-2BR/ PEDOT:PSS                             | 0.043                             | 2.88                  | 35        |
| 2016                                                              | PET<br>(Flexible)            | Ag gird/PEDOT:PSS/ZnO/<br>PffBT4T2OD:PC61BM:PC71BM/PEDOT:PSS:AgNWs | 0.1                               | 6.6                   | 36        |
|                                                                   |                              |                                                                    | 1                                 | 5.8                   | 36        |
| 2016                                                              | Glass<br>(Rigid)             | AgNWs/ZnO/PV2000:PC70BM/ PEDOT:PSS/AgNWs                           | 1                                 | 4.3                   | 37        |
| 2016                                                              | Glass<br>(Rigid)             | PEDOT:PSS/PEI/P3HT:ICBA /PEDOT:PSS                                 | -                                 | 4.0                   | 22        |

|      |                         |                                                                                            |           |       |    |
|------|-------------------------|--------------------------------------------------------------------------------------------|-----------|-------|----|
| 2016 | PET<br>(Flexible)       | Ag gird/PEDOT:PSS/ZnO/ PTB7-<br>TH:IEIC/PEDOT:PSS/Ag gird                                  | 0.95-1.05 | 1.79  | 38 |
| 2016 | Glass<br>(Rigid)        | PEDOT:PSS/PEIE/TQ1:PC71BM/PEDOT:PSS                                                        | 0.04      | 2.54  | 39 |
| 2016 | PET<br>(Flexible)       | Ag gird/PEDOT:PSS/ZnO/<br>P3HT:ICBA/PEDOT:PSS/Ag<br>grid                                   | 57        | 2     | 40 |
| 2016 | PES<br>(Flexible)       | (PEDOT:PSS/PEI/P3HT:ICBA)*5/PEDOT:PSS (6-<br>junction)                                     | -         | 6.5   | 41 |
| 2016 | PET<br>(Flexible)       | AgNWs/ZnO/P3HT:PCBM/PEDOT:PSS/ZnO/Polymer<br>Generation 2.1:PCBM/PEDOT:PSS/Ag gird         | 0.8-1     | 2.66  | 42 |
| 2016 | IF Fabric<br>(Flexible) | AgNWs/ZnO np/ P3HT:ICBA/PEDOT:PSS/AgNWs                                                    | 0.06      | 0.14  | 43 |
| 2015 | PET<br>(Flexible)       | Ag gird/PEDOT:PSS/ZnO/<br>P3HT:F(DPP)2B2/PEDOT:PSS/Ag grid                                 | 1         | 0.65  | 44 |
| 2015 | PES<br>(Flexible)       | PEDOT:PSS/PEI/P3HT:ICBA/ PEDOT:PSS                                                         | -         | 3.3   | 45 |
| 2015 | PET<br>(Flexible)       | Ag gird/PEDOT:PSS/ZnO/ PBDTTT-C-<br>:DCIDT2T/PEDOT:PSS/Ag gird                             | 1         | 1.019 | 46 |
| 2015 | Glass<br>(Rigid)        | Glass/P_Ag/pDPP5T-2/AgNWs                                                                  | 0.15      | 4.74  | 47 |
|      |                         | Ag/PEDOT:PSS/GEN-3:PCBM/ZnO/ N-<br>PEDOT/pDPP5T2:PCBM /ZnO/AgNWs (2-junction)              | 0.15      | 5.81  |    |
| 2015 | Glass<br>(Rigid)        | Ag gird/PEDOT:PSS/ZnO/<br>P3HT:PCBM/PEDOT:PSS/Ag                                           | 1         | 4.1   | 48 |
| 2015 | PET<br>(Flexible)       | Ag grid/PEDOT:PSS/ZnO<br>np/PBDTTTz4:PCBM/PEDOT:PSS/ZnO<br>np/PDTPI:PCBM/PEDOT:PSS/Ag grid | 0.8       | 1.72  | 49 |
| 2015 | Glass<br>(Rigid)        | PEDOT:PSS/PEI/P3HT:ICBA/PEDOT:PSS                                                          | -         | 3.8   | 50 |
| 2015 | Glass<br>(Rigid)        | Ag/AZO/P3HT:PCBM/PEDOT:PSS/AgNWs                                                           | 0.104     | 2.5   | 51 |
| 2015 | PET<br>(Flexible)       | PEDOT:PSS/PEIE/TQ-1:PCBM/PEDOT:PSS                                                         | 0.1       | 0.4   | 52 |
| 2014 | PET<br>(Flexible)       | Ag gird/PEDOT:PSS/ZnO np/PBDTTT-<br>CT:PC71BM/PEDOT:PSS /Ag gird                           | 1         | 2.09  | 53 |
| 2014 | Glass<br>(Rigid)        | AgNWs/PEDOT:PSS/ DPP:PCBM/ZnO/AgNWs                                                        | 0.104     | 2.09  | 54 |
| 2014 | PET                     | Ag/PEDOT:PSS/ZnO/MH301:PCBM/PFN/PEDOT:PSS/                                                 | 52.2      | 1.76  | 55 |



|      |            |                                              |       |      |    |
|------|------------|----------------------------------------------|-------|------|----|
|      | (Flexible) | Zn                                           |       |      |    |
|      |            | O/MH306:PCBM/PFN/PEDOT:PSS/Ag grid           |       |      |    |
| 2014 | PET        | Ag gird/PEDOT:PSS/ZnO/                       | 1     | 3.8  | 56 |
|      | (Flexible) | PBDTTHDDTBTff:PCBM/PEDOT:PSS/Ag gird         |       |      |    |
| 2014 | PET        | Ag/ZnO np/PBDTT-FTTE:PC71BM/                 | 0.4   | 4.8  | 57 |
|      | (Flexible) | PEDOT:PSS/AgNWs                              |       |      |    |
| 2014 | PET        | Ag/ZnO/P3HT:PCBM/PEDOT:PSS/ZnO/              | 1     | 2.39 |    |
|      | (Flexible) | PPTBTT:PCBM/PEDOT: PSS /Ag grid              |       |      | 58 |
|      |            | Ag/ZnO/PBDTTT:PCBM/PEDOT:PSS/Ag gird         | 0.8   | 2.73 |    |
| 2014 | Glass      | PEDOT:PSS/PCDTBT:PCBM/ZnO np/Ag ink          | 0.5   | 3.13 | 59 |
|      | (Rigid)    |                                              |       |      |    |
| 2014 | Glass      | AgNWs/TiOx/ZnO/ P3HT:PCBM/PEDOT:PSS/GO       | 0.24  | 2.3  | 60 |
|      | (Rigid)    |                                              |       |      |    |
| 2014 | PET        | Ag                                           |       |      |    |
|      | (Flexible) | gird/PEDOT:PSS/ZnO/P1:PCBM/PEDOT:PSS/ZnO/P2: | -     | 2.67 | 61 |
|      |            | PCBM /PEDOT :PSS/ Ag gird                    |       |      |    |
| 2014 | PET        | Ag gird/PEDOT:PSS/ZnO/                       | 1     | 2.05 | 62 |
|      | (Flexible) | P3HT:PCBM/PEDOT:PSS/Ag                       |       |      |    |
|      |            | gird                                         |       |      |    |
| 2014 | PET        | PEDOT:PSS/ZnO np/                            | -     | 1.83 | 63 |
|      | (Flexible) | P3HT:PCBM/PEDOT:PSS/Carbon                   |       |      |    |
| 2014 | PET        | Ag gird/PEDOT:PSS/ZnO/ P3HTT-                | 1.1   | 1.36 | 64 |
|      | (Flexible) | DPP10%:PCBM/PEDOT:PSS                        |       |      |    |
| 2014 | PET        | Ag gird/PEDOT:PSS/ZnO/                       | 1     | 1.01 | 65 |
|      | (Flexible) | SM1:PC61BM/PEDOT:PSS/Ag gird                 |       |      |    |
| 2014 | Glass      | Graphene/PEDOT:PSS/PSEHTT:IC60BA/ZnO/PEDOT:  | 0.04  | 8.02 | 66 |
|      | (Rigid)    | PSS/ PBDTT-DPP:PC71BM/TiO2/AgNWs             |       |      |    |
| 2014 | PET        | Ag gird/PEDOT:PSS/ZnO/                       | 1     | 3.2  | 67 |
|      | (Flexible) | PDTSTTz4:PCBM/PEDOT:PSS/Ag gird              |       |      |    |
| 2013 | PET        | Ag gird/PEDOT:PSS/ZnO/                       | 142   | 1.82 | 68 |
|      | (Flexible) | P3HT:PCBM/PEDOT:PSS/Ag gird                  |       |      |    |
| 2013 | PET        | Ag gird/PEDOT:PSS/ZnO/                       | 1     | 3.5  | 69 |
|      | (Flexible) | PDTSTTz4:PCBM/PEDOT:PSS/Ag gird              |       |      |    |
| 2013 | Glass      | AgNWs/AZO/P3HT:PCBM:SiPCPDTBT/PEDOT:PSS/     | 0.104 | 2.2  | 70 |
|      | (Rigid)    | AgNWs                                        |       |      |    |

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