Supporting Information

Ultra-flexible organic solar cells based on eco-friendly cellulose substrate with efficiency approaching 19%

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Materials:

All materials were used as obtained unless otherwise indicated. Silver nanowires (AgNWs) (10 wt% in H_2O with an average length of ~25 μm and diameter of ~35 nm) were purchased from Zhejiang Kechuang Adv. Mater. Co., Ltd. PM6, BO-4Cl, Y6, PBDB-T and ITIC-4F were purchased from Solarmer Materials (Beijing) Inc. L8-BO was purchased from Jiaxing Hyper Inc. Poly (sodium 4-styrenesulfonate) (PSSNa, Mw ~500,000, powder), 1,8-diiodooctane (DIO) were purchased from Sigma-Aldrich. Ethyl cellulose (EC), Ethoxylated trimethylolpropane triacrylate (ETPTA) and 2-Hydroxy-2-methylpropiophenone were purchased from Adamas Reagent Co., Ltd. The indium-doped tin oxide (ITO)-coated glass and PET/ITO, PEN/ITO were purchased from Liaoning Advanced Election Technology. ZnO nanoparticles (NPs) were synthesized according to the literature.[1]

Electrode Fabrication:

The EC solution was prepared by dissolving 0.05 g of EC into 1 mL of methanol (Kemiou, 99.8%). After EC solution was filtered, different contents of ETPTA with 2-Hydroxy-2-methylpropiophenone (3wt%, as initiator) were added to obtain EC@ETPTA solution. EC@ETPTA solution were deposited on the glass by spin coating to obtain the EC@ETPTA films (7-8 μm). For preparing EC@ETPTA/AgNWs electrode, the EC@ETPTA substrate was treated by plasma for 8 min and a thin layer (~ 20 nm) ZnO NPs was deposited on the flexible substrate, then AgNWs was spinning coated following our previous report.[2]

Electrode characterization.

Sheet resistances of the glass/ITO, PET/ITO, PEN/ITO and EC@ETPTA/AgNWs were measured with a ST-2258C four-probe instrument (Suzhou Jingge Electronic). Atomic force microscope (AFM) investigation was performed using Bruker MultiMode 8 in tapping mode. AFM-IR experiments were carried out using a commercial AFM-IR setup (Bruker nanoIR 3) that consists of an AFM microscope operating in contact mode and a Quantum Cascade Laser (QCL laser). The surface and cross-section morphologies of EC@ETPTA and EC@ETPTA/AgNWs were measured by SEM (ZEISSMERLIN Compact). The stretch ability was measured by cupping machine (Transcell Technology BAB-10MT). The UV-Vis spectra were obtained by a Cary

5000 UV-Vis spectrophotometer. The thermal expansion coefficient measurements were performed using a Thermal Mechanical Analysis (TMA) (PE DMA8000). The Stress-strain measurements were performed using a mechanical tester (CTM-2GD) with a pre-strain of 5% in the temperature range of 25°C at an environmental RH of 35%.

Device fabrication:

The OSCs devices were fabricated using an inverted structure of Electrode/ZnO /active layers/MoOx/Ag. For rigid devices, ITO was ultrasonically cleaned with detergent, deionized water, acetone, isopropanol and UV-treated for 15 min. ZnO nanoparticle (ZnO NPs)[3] layer (~ 20 nm) was deposited by spin coating on top of precleaned ITO substrates and annealed at 120 °C for 10 mins. Different active layers were deposited following procedures in previous reports[4-8]. Then, MoO_x (~5 nm) and Ag (~100 nm) were successively evaporated onto the active layer through a shadow mask (pressure ca. 10^{-4} Pa). The device area is ~4 mm^2. The flexible devices based on the EC@ETPTA/AgNWs and PET/ITO and PEN/ITO electrodes were fabricated following the same procedure to that of the rigid devices.

Device characterization.

The J-V characteristics of photovoltaic devices were measured using a Keithley 2400 source measure unit, with a fixed scan speed of 0.02 V s-1 and dwell times of 20 ms for the J-V curves. The photocurrent was measured under illumination simulated 100 mW cm^{-2} AM1.5G irradiation using an Enli SS-F5-3A solar simulator, calibrated with a standard Si solar cell (Enli Technology Co., Ltd., Taiwan, and calibrated report can be traced to NREL). The external quantum efficiency (EQE) spectrum was measured using a QE-R Solar Cell Spectral Response Measurement System (Enli Technology Co., Ltd., Taiwan). The thickness of film and device was measured using a Veeco Dektak 150 profilometer.

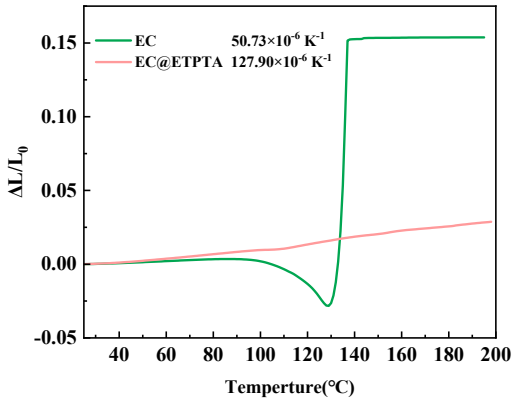

Fig. S1. The values of thermal expansion coefficients (TEC) for the EC and EC@ETPTA.

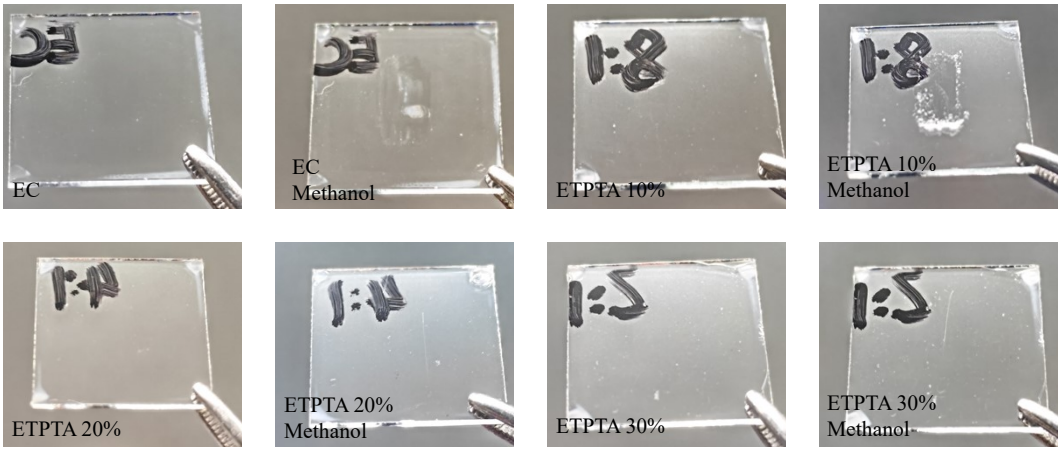

Fig. S2. Pure EC and EC@ETPTA films with different ETPTA contents were subjected to solvent resistance testing against methanol following ASTM D5402-19.

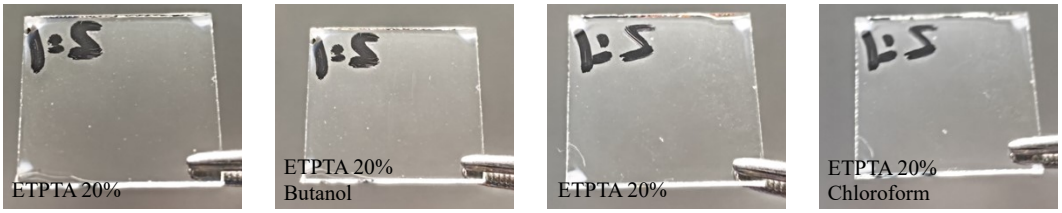

Fig. S3. Stability of EC@ETPTA films with 20% ETPTA content were subjected to solvent resistance testing against butanol and chloroform following ASTM D5402-19.

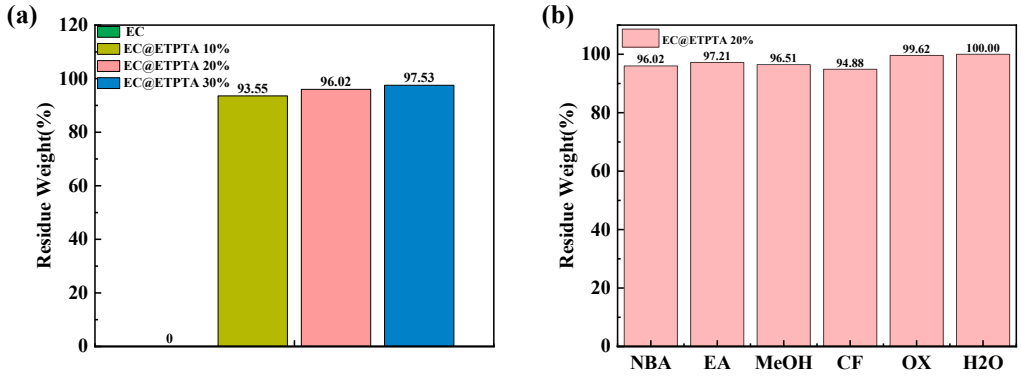

Fig. S4. Residue weight of the a) dried EC and EC@ETPTTA containing different ETPTA content films after soaking in n-butanol and b) EC@ETPTTA containing 20% ETPTA after soaking in different solvents for 24 h.

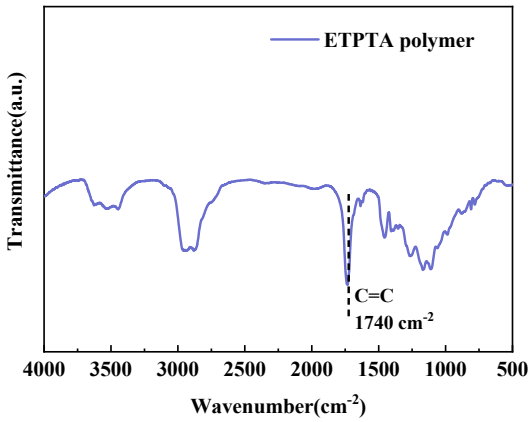

Fig. S5. FT-IR spectra of ETPTA polymer.

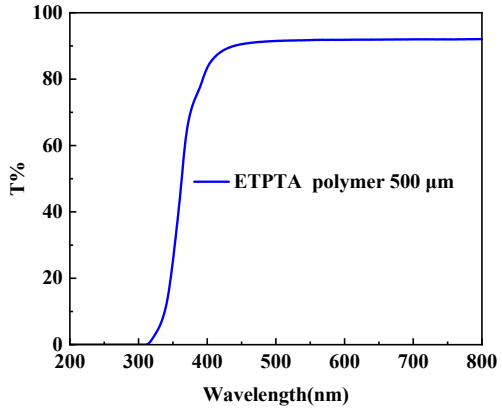

Fig. S6. Transmittance spectra of ETPTA.

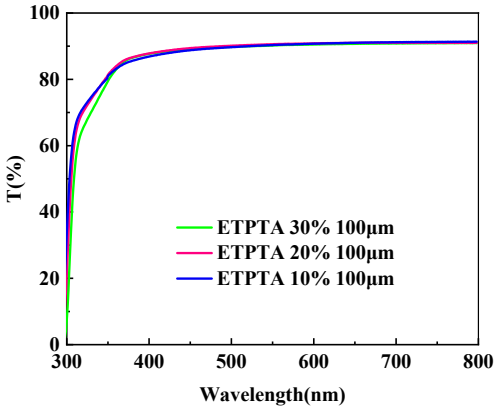

Fig. S7. Transmittance spectra of EC@ETPTA films with different ETPTA contents.

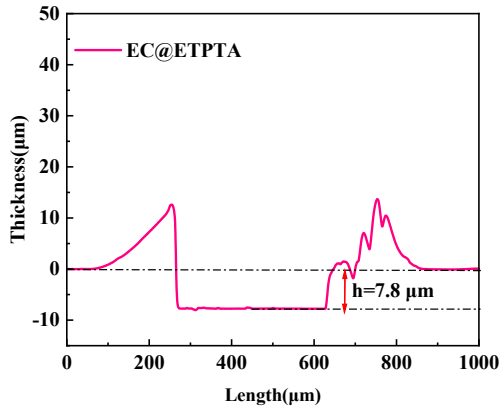

Fig. S8. The thickness of EC@ETPTA film was measured by a profiler.

Fig. S9. Authentic pictures of EC@ETPTA (23×13 cm).

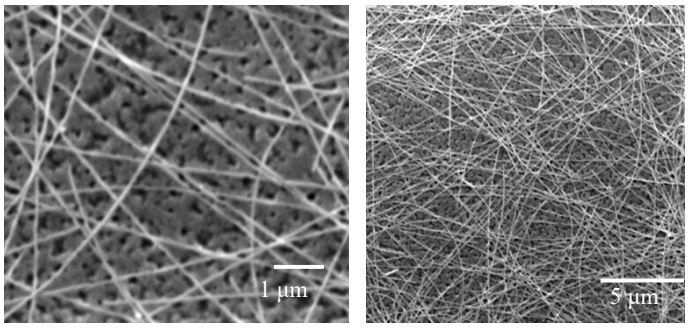

Fig. S10. SEM image of EC@ETPTA/AgNWs film.

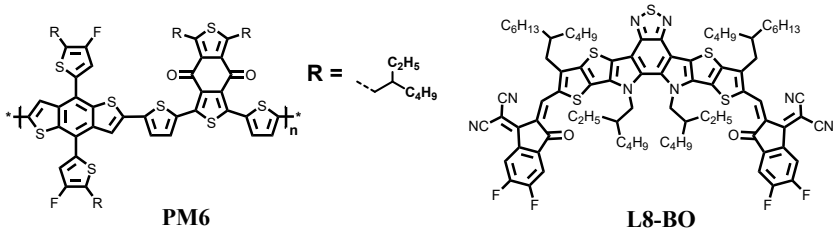

Fig. S11. Chemical structure of PM6, L8-BO.

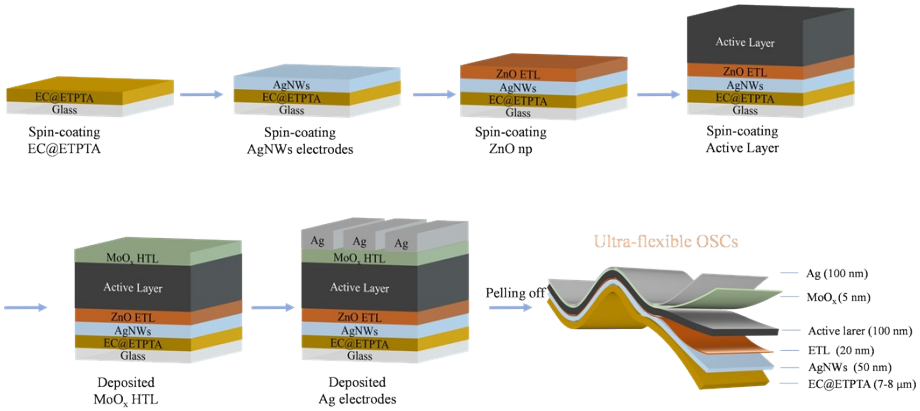

Fig. S12. The schematic diagram of the detailed preparation process of ultra-flexible OSCs.

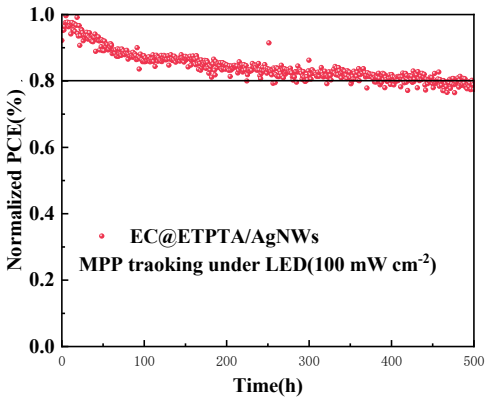

Fig. S13. illumination stability at maximum power point (MPP) tracking of the flexible devices based on EC@ETPTA/AgNWs.

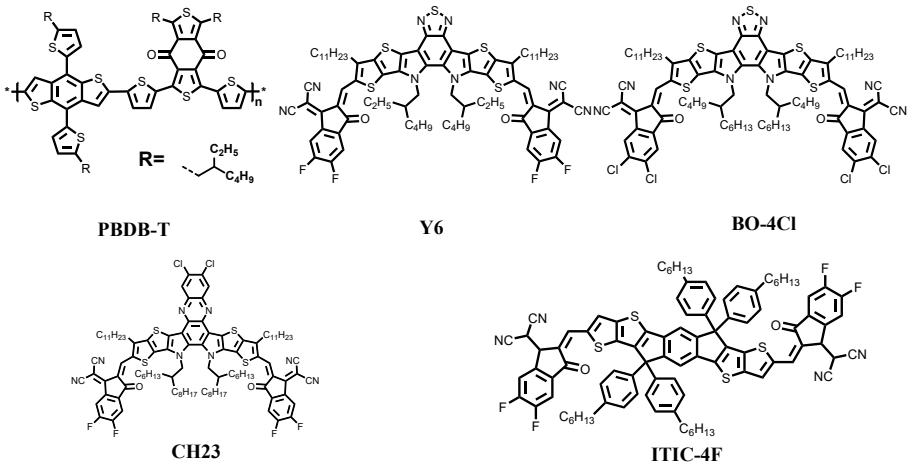

Fig. S14. Chemical structure of PBDB-T, Y6, BO-4Cl, CH23 and ITIC-4F.

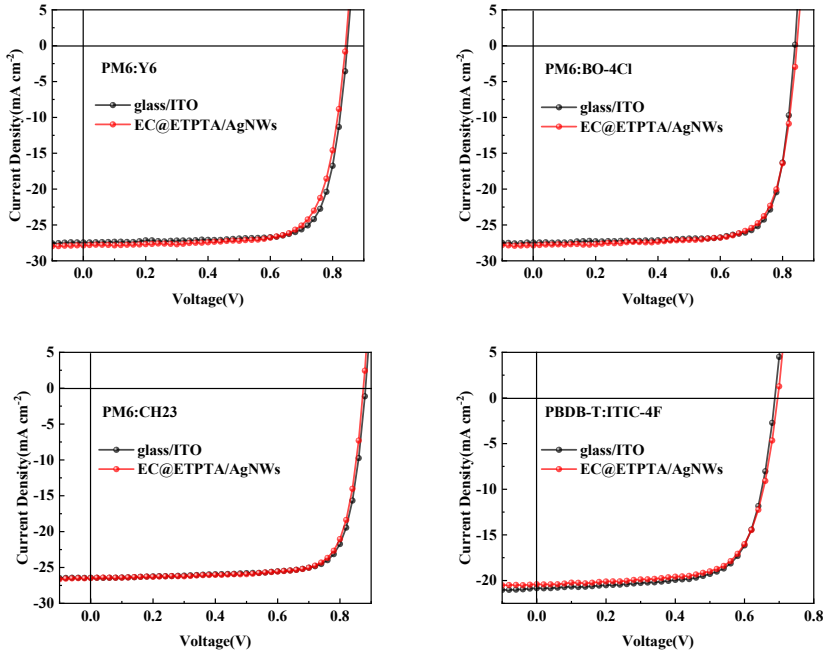

Fig. S15. J-V curves of OSCs based on EC@ETPTA/AgNWs and glass/ITO electrodes with different active layers.

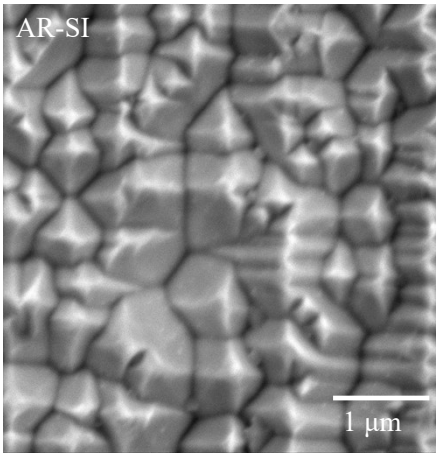

Fig. S16. SEM image of a silicon wafer with AR structure (AR-Si).

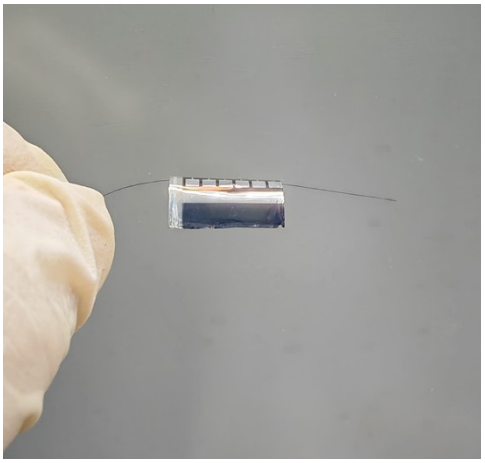

Fig. S17. The display of an ultra-flexible OSCs device on a single human hair.

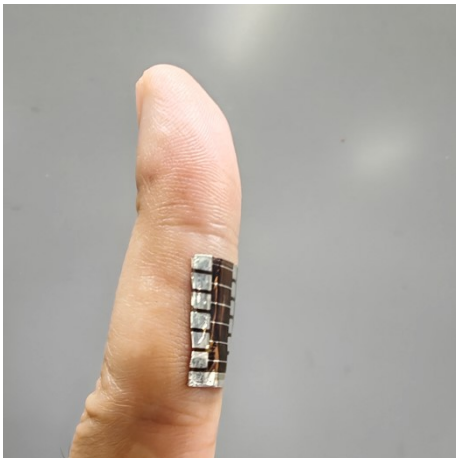

Fig. S18. Photograph of the free-standing OPVs attached to a finger.

4. Supplementary Tables

Table S1. Properties of different electrode.

Electrode	T(%) @550 nm	Rsh (Ω/□)	FoM
Glass/ITO	89.56	12.45	267.12
EC@PEGDMA/AgNWs	88.92	21.04	148.14
PEN/ITO	83.52	16.48	121.39
PET/ITO	74.20	12.67	92.46

Table S2. Photovoltaic parameters of recently reported flexible OSCs

Device structure	V_{oc} [V]	J_{sc} [mA cm^{-2}]	FF [%]	PCE [%]	Refs
PPZA/ZnO/PM6: N3: $PC_{71}BM/MoO_3$/Ag	0.840	25.00	76.50	16.10	[9]
PET/AgNWs/ZnO: $CsCO_3$/D18: Y6: $PCB_{71}M/MoO_3$/Ag	0.862	26.44	77.43	17.64	[10]
Em-AgNW FTE/ZnO/ PM6: L8-BO/MoO3/Ag	0.834	24.26	76.34	15.41	[11]
PET/Ag grids/D-PH1000/ PEDOT: PSS/D18: N3: QX-α/PDINN/Ag	0.856	27.21	77.30	18.01	[12]
PDMS/AgNWs/ZnO: $CsCO_3$/D18: Y6: $PCB_{71}M/MoO_3$/Ag	0.860	26.94	16.48	17.72	[13]
Em-Ag/AgNWs:AZO-SG/PBDB-T-2F: Y6/MoO_3/Al	0.832	25.05	72.97	15.21	[14]
PET/AgNWs@SnO_x/ZnO/PM6: L8-BO/MoO_3/Ag	0.860	25.5	75.80	16.60	[15]
PET/AgNW:PDA: AgNP /AZO/PBDB-T-2F: BTP-ec9:$PCB_{71}M/MoO_3$/Ag	0.830	27.44	74.96	17.07	[16]
AgNWs@CPI/ZnO/PM6: BTP-4Cl/MoO_3/Ag	0.780	24.77	74.35	14.37	[17]
PET/Em-Ag/AgNWs/AZO/PM6: BTP-eC9: PC71BM/MoO_3/Ag	0.835	27.37	76.90	17.52	[18]
FlexAgNEs/ZnO NPs/PFN-Br/PM6: Y6/MoO_3/Ag	0.829	25.43	74.50	15.71	[3]
EC/PI/AgNWs/ZnO NPs/NMAO/PM6: L8-BO/MoO_3/Ag	0.875	26.38	78.26	18.05	[4]
AR-EC@ETPTA/AgNWs/ZnO NPs/NMAO/PM6: L8-BO/MoO_3/Ag	0.876	27.67	77.13	18.71	This work

References

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