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Efficient and stable low-cost perovskite solar cells enabled by using the surface passivated carbon as counter electrode

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Methods

Chemicals: Dimethyl sulfoxide (DMSO), N,N-dimethylformamide (DMF, 99.99%), chlorobenzene and FTO substrates were purchased from Liaoning Youxuan New Energy Technology Co., Ltd. Lead iodide (PbI_2, 99.999%) was purchased from Liaoning Huite Technology Corp., Ltd. Lead bromide ($PbBr_2$), cesium iodide, (CsI, 99.9%), methylammonium iodide (MABr), formamidine hydroiodide (FAI, 99.99%) purchased from Xi'an Polymer Light Technology Corp., Ltd. Low-temperature carbon paste was purchased from Ningbo Borun New Material Corp., Ltd. $TiCl_4$ (98%) and PEG-2000 were purchased form Sigma Aldrich.

TiO_2 ETL Fabrication: Laser patterned FTO substrates were ultrasonically cleaned by deionized water, acetone and isopropanol for 15 min, respectively, followed by the treatment with using a plasma cleaning machine for 20 min before use. Then FTO substrates were immersed into 40 mM $TiCl_4$ at 70 ℃ for 1 h, afterwards they were rinsed with ultrapure water and ethanol, respectively. After drying, heat on a hot plate at 200 ℃ for 30 min. Before preparing perovskite, the TiO_2 substrates were treated by plasma cleaning machine for 15 min.

Perovskite Film Fabrication: 1.12 mol of PbI_2, 0.2 mol of $PbBr_2$, 0.2 mol of MABr and 1.1 mol of FAI were dissolved in 1 mL mixed solvent of DMF/DMSO (4:1 in volume ratio). After full dissolution, 5 vol% of preprepared CsI/DMSO solution was added to have a mix-cation perovskite precursor solution. Perovskite solution (70 μL) was spin-coated on the TiO_2 film at 10 s at 1000 rpm and 30 s at 5000 rpm. During the last 10 s of the second spin-coating step, the substrates were treated by dropping of chlorobenzene (150 μL) on the spinning films. Then the perovskite precursor films were heated at 100 °C for 40 min.

Carbon Electrode Fabrication: Carbon paste was coated onto the glass slide by doctor-blade, then soaked the wet-films in ethanol or PEG/ethanol solution (1 wt%). After full immersion, the carbon films fall off spontaneously and forms an independent wet carbon film. After drying at 100 ℃ for 5 min, the carbon electrode films were obtained. Then the carbon films were directly adhered to the top surface of perovskite by hot pressing transfer method (80 ℃, 0.5 MPa).

Characterization:

The square resistance of carbon films was performed using four-point probe resistivity tester. Current density-voltage (J-V) of the cells were obtained by using a solar simulator with a source meter (Keithley 2400) at one sun light intensity (AM1.5G, 100 mW cm^{-2}). While measuring photocurrent and voltage, the device was covered with a 0.1 cm^2 black mask. The incident photon to current efficiency (IPCE) spectra were recorded using an IPCE measurement system. Prior to starting the measurement, the light intensity at each wavelength was adjusted by using a standard

Si solar cell. The surface and cross-sectional morphology of the samples were obtained with a field emission scanning electron microscope (FE-SEM, JSM 7001F). The electrochemical impedance spectra of devices were measured by using an electrochemical workstation (Zahner IM6) in dark condition with the frequency ranging from 10^5 to 10^{-1} Hz (perturbation amplitude is 10 mV).

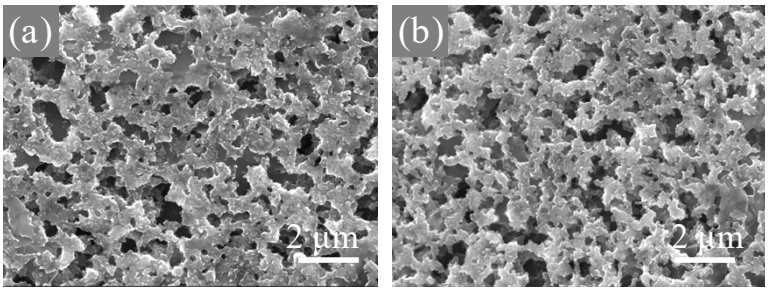

Figure S1. The SEM images of carbon films without (a) and (b) with PEG.

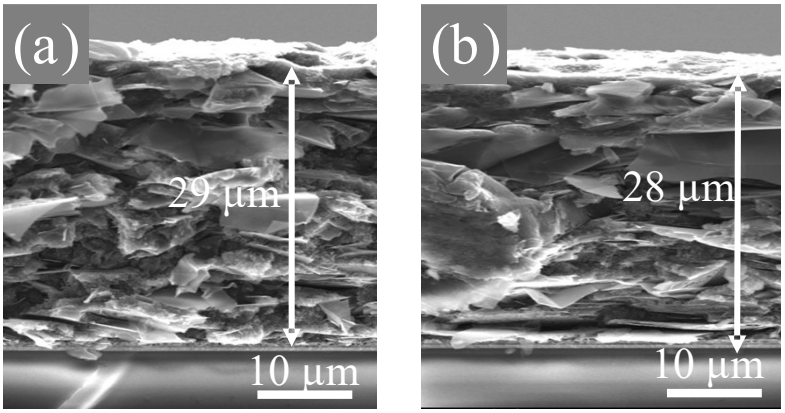

Figure S2.The cross-sectional SEM images of C_{Ref} and C_{PEG} films.

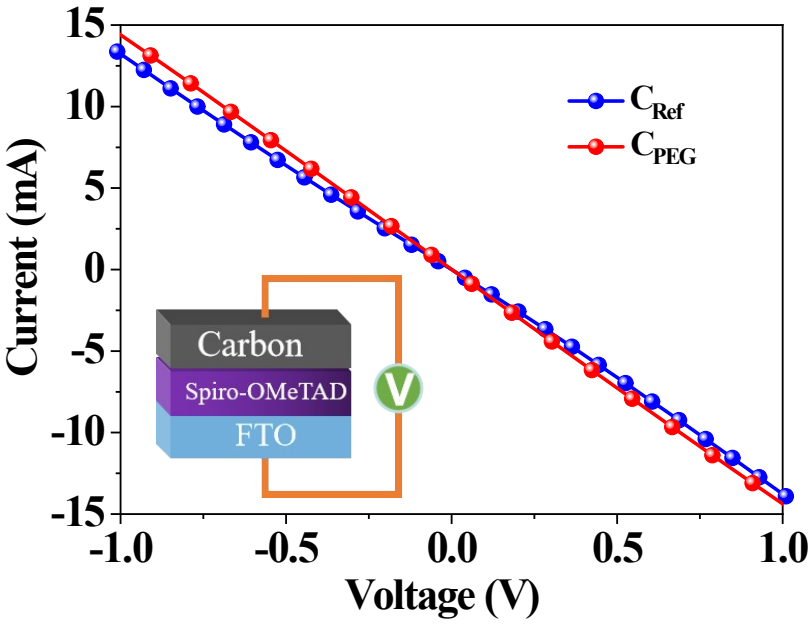

Figure S3. Direct current I-V measurement curves of FTO/Spiro-OMeTAD/Carbon.

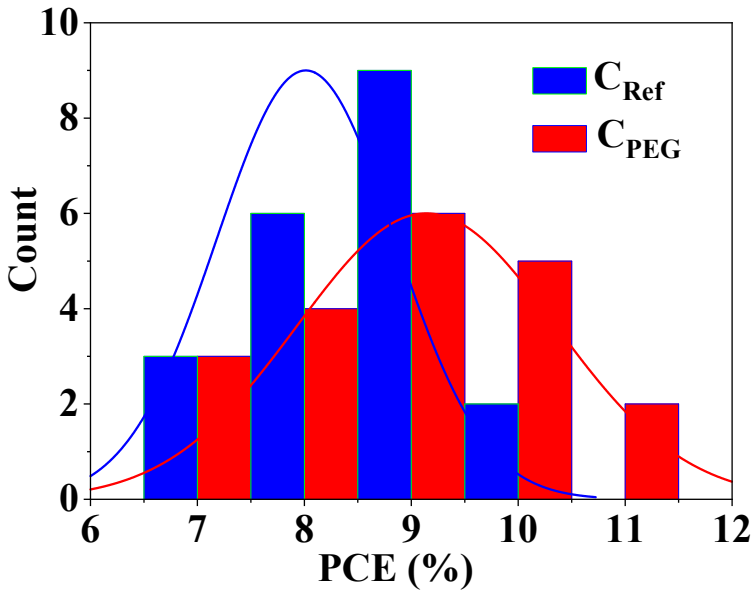

Figure S4. Power conversion efficiency histogram of C_{Ref} and C_{PEG} based devices.

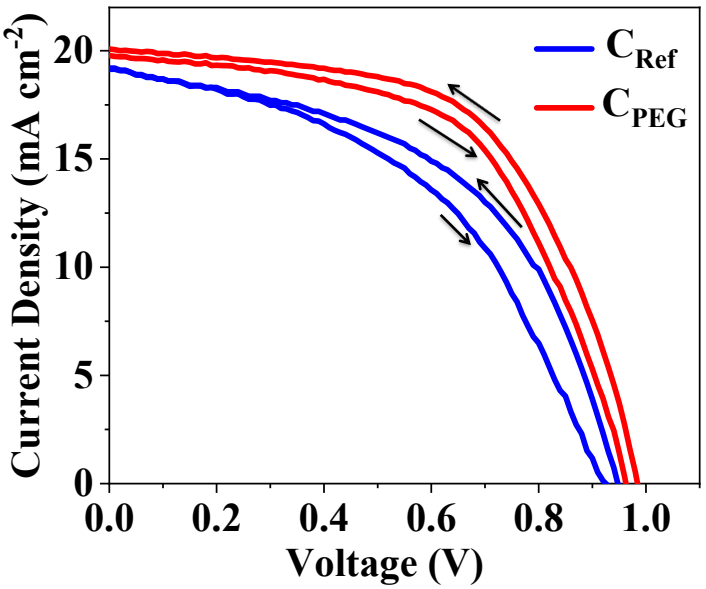

Figure S5. Reverse and forward J-V curves of the PSCs based on C_{Ref} and C_{PEG} electrode.

Table S1. Photovoltaic parameters of the PSCs based on C_{Ref} and C_{PEG} electrode.

Samples	Direction	V_{OC}(V)	J_{SC}(mA cm^{-2})	FF (%)	PCE (%)	Hysteresis Index (%)
C_{Ref}	Reverse	19.22	0.95	0.50	9.17	10.79
	Forward	19.14	0.93	0.46	8.18	
C_{PEG}	Reverse	20.07	0.98	0.58	11.52	5.64
	Forward	19.78	0.97	0.56	10.87	

Table S2. EIS parameters of the PSCs with different carbon electrodes.

Samples	R_s/Ω	R_{rec}/Ω
C_{Ref}	103	285
C_{PEG}	91	594