

Supplementary Information

A facile route of preparing nickel(II) oxide thin films for high-performance inverted perovskite solar cells

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Experimental section

1. Materials

Indium-tin oxide (ITO) glasses were purchased from OPVtech (China) with the size of 1.5 cm×1.5 cm and patterned by laser etching. Nickel(II) nitrate hexahydrate was obtained from Beijing Tong Guang Fine Chemicals (China) Co., Ltd. Polymethyl methacrylate (450-550k) was obtained from Aladdin (China). Lead(II) iodide (PbI₂) was purchased from Alfa Aesar. MAI and C₆₀ were purchased from Xi'an Polymer Light Technology (China) Co., Ltd. Chlorobenzene (CB), N, N-dimethylformamide (DMF), and dimethyl sulfoxide (DMSO) were obtained from J&K (China) Co. All reagents were directly used without further purification.

2. Preparation of NiO film

2.1. PAD NiO film

An optimum PAD NiO precursor solution was prepared by dissolving 0.6 mmol $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and 30 mg PMMA in 6 ml acetone. The solution was filtered using a 0.45 μm PTFE syringe filter after being stirred at room temperature for 12 h. PAD NiO precursor solution was spin coated on the ITO substrates at 5000 rpm for 20 s. Then the precursor film was transferred to a muffle furnace with the setting temperature of 275 $^\circ\text{C}$ for 3 h at a heating rate of 5 $^\circ\text{C}/\text{min}$. All the above procedures were performed in air atmosphere. After cooling to room temperature, the NiO films were transferred into a glove box for further process.

2.2. Control NiO film

The control NiO films were prepared according to the procedures described in previous reports. In short, 291 mg (1 mmol) $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and 67 μL (1 mmol) ethylene diamine was dissolved in 1 mL ethylene glycol solution. Afterwards, the solution was spin-coated on ITO substrates at a speed of 3000 rpm for 30 s. The substrates were annealed at 300 $^\circ\text{C}$ for 1 h in a muffle furnace.

3. Device preparation

ITO substrates were washed by ultrasonic cleaning and rinsed with an aqueous detergent solution, followed by deionized water, then acetone, and finally isopropanol.

After drying in a baking oven, the substrates were then treated with an oxygen plasma for 5 min.

The perovskite precursor solution was prepared from the mixture of 1.2 M PbI_2 and 1.2 M MAI in DMF/DMSO with a ratio of 7:3 by volume. The solution was deposited on NiO layers by spin-coating, which included two steps of 1000 rpm for 10 s and 5000 rpm for 25 s. 300 μL CB was dropped onto the precursor film as anti-solvent after 17 s. Then the films were annealed at 60 °C for 5 min and at 100 °C for 10 min. After cooling to room temperature, C_{60} was thermally evaporated above the perovskite layer to form a 26 nm electron transport layer, a 6.5 nm blocking layer of BCP and a 100 nm Ag electrode were deposited by evaporation to finish the device.

4. Characterization

J-V characteristics of the PSCs were measured on a system equipped with a Keithley 2400 SourceMeter under a simulated AM 1.5G spectrum (100 mW/cm^2) calibrated by a standard silicon reference cell. During the measurement, the solar cells were covered with a shadow mask with a defined active area of 0.1 cm^2 . The EQE was measured on an Enli Technology (Taiwan) EQE measurement system (QE-R), and the light intensity was calibrated using a silicon reference solar cell.

Raman scattering spectra were obtained using a Horiba Xplora plus Raman spectrometer with a 532 nm laser. The crystallinity of all the films were characterized using XRD (Shimadzu XRD-70000) at a 2 °/min scan rate. Thermogravimetric analysis (TGA)- Differential scanning calorimetry (DSC) were carried out using an

SDT Q600 V20.9 Build 20. UPS was measured using a Thermo Scientific ESCALab 250Xi spectrometer with a monochromatic He I light source (21.22 eV). The surface morphology images of the perovskite films were obtained with a scanning electron microscope (Hitachi SU8010). A Cary 50 spectrometer was used to obtain UV-vis absorption and transmission spectra of the films. Photoluminescence was recorded using an Edinburgh FLS 980 spectrometer under 510 nm xenon lamp excitation light. The time-resolved PL (TRPL) spectra were obtained using an UltraFast Lifetime Spectrofluorometer (Horiba Jobinyvon IBH Inc, Delta flex) with a Hamamatsu 320 nm diode laser excitation source.

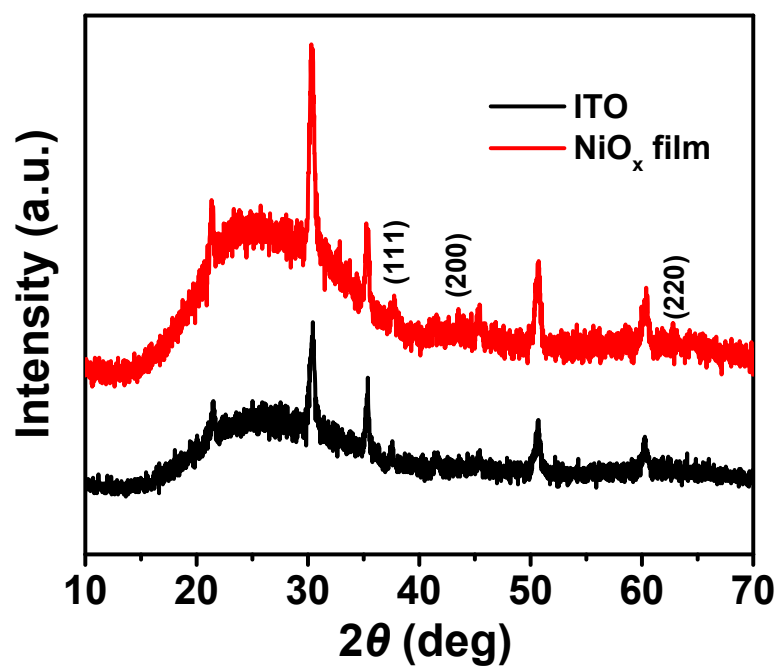


Fig. S1. XRD pattern of pristine ITO and PAD NiO film on the ITO substrate.

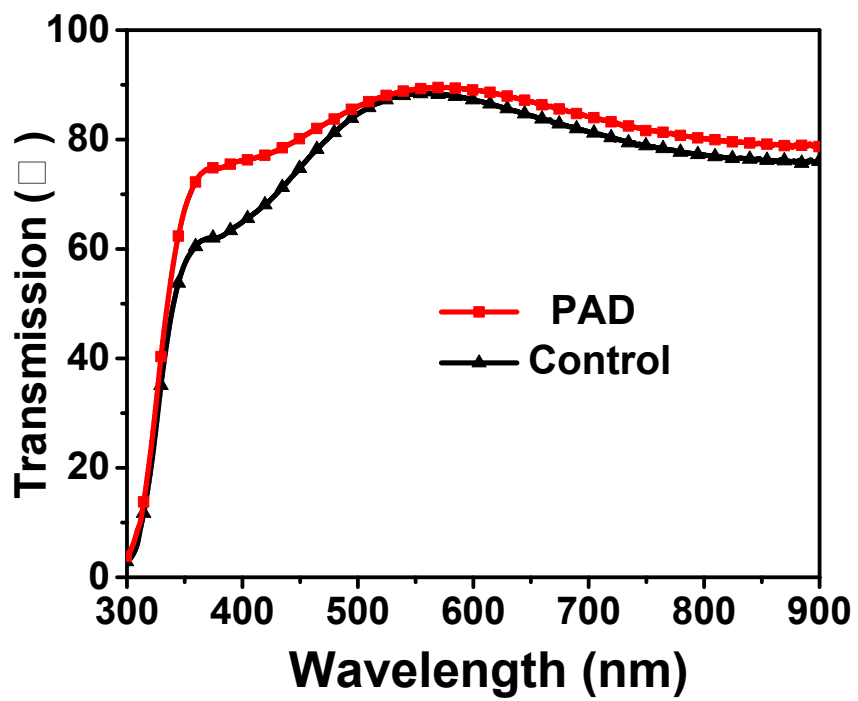


Fig. S2. Transmittance spectra of the Control and PAD NiO films on ITO substrates.

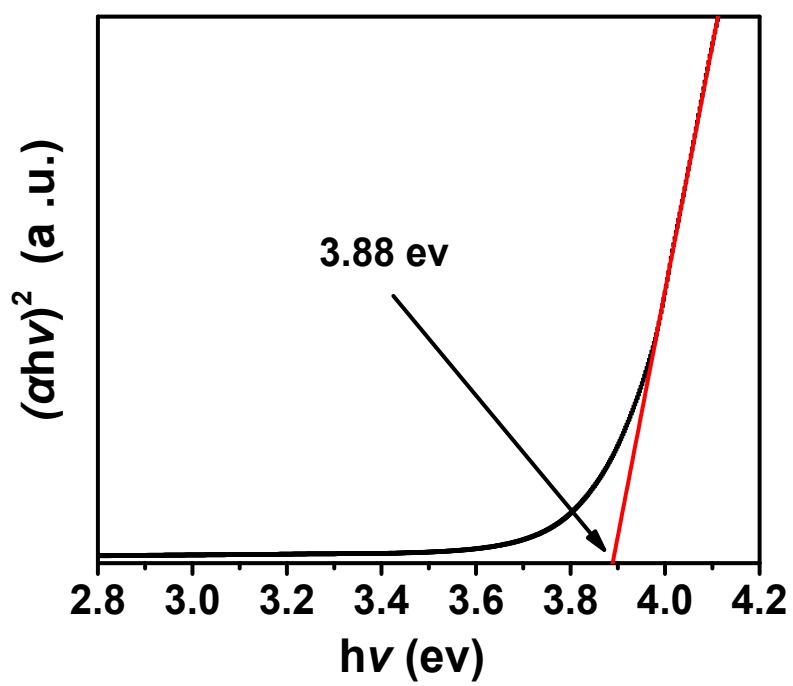


Fig. S3. Tauc plots for the PAD NiO film

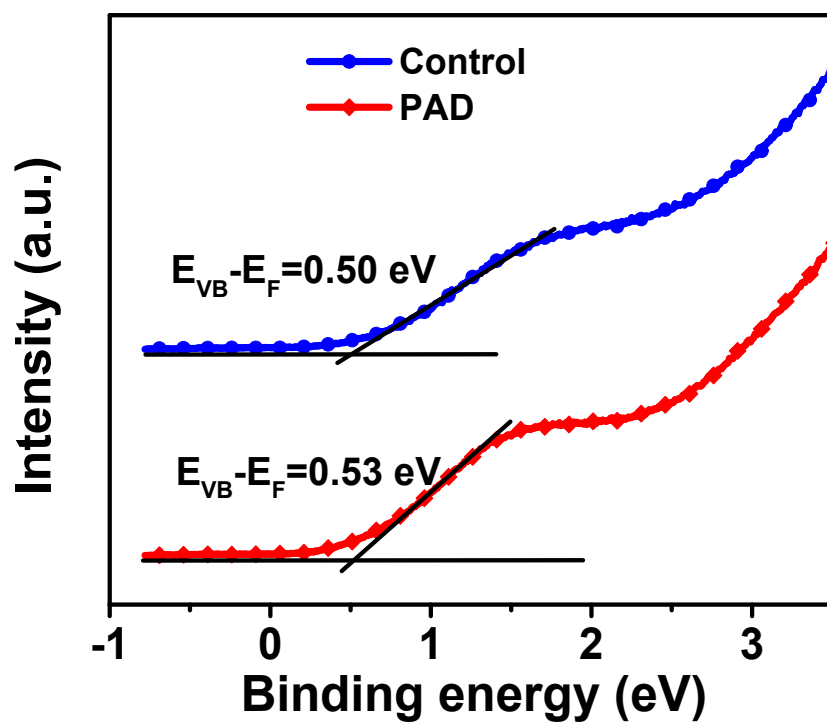


Fig. S4. UPS spectrum of Control and PAD NiO films.

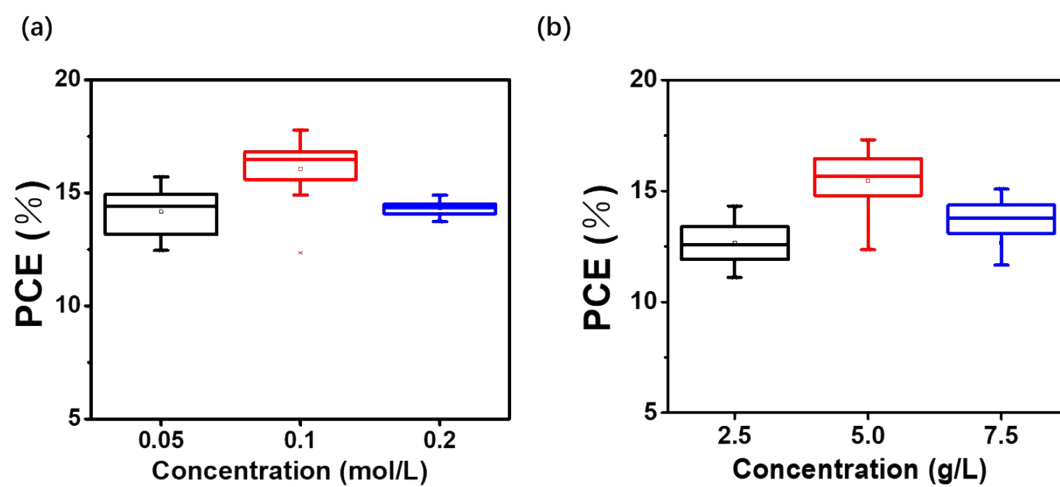


Fig. S5. Statistical PCE of devices corresponding to the precursor solutions with series concentration of (a) $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (mol/L) and (b) PMMA (g/L).

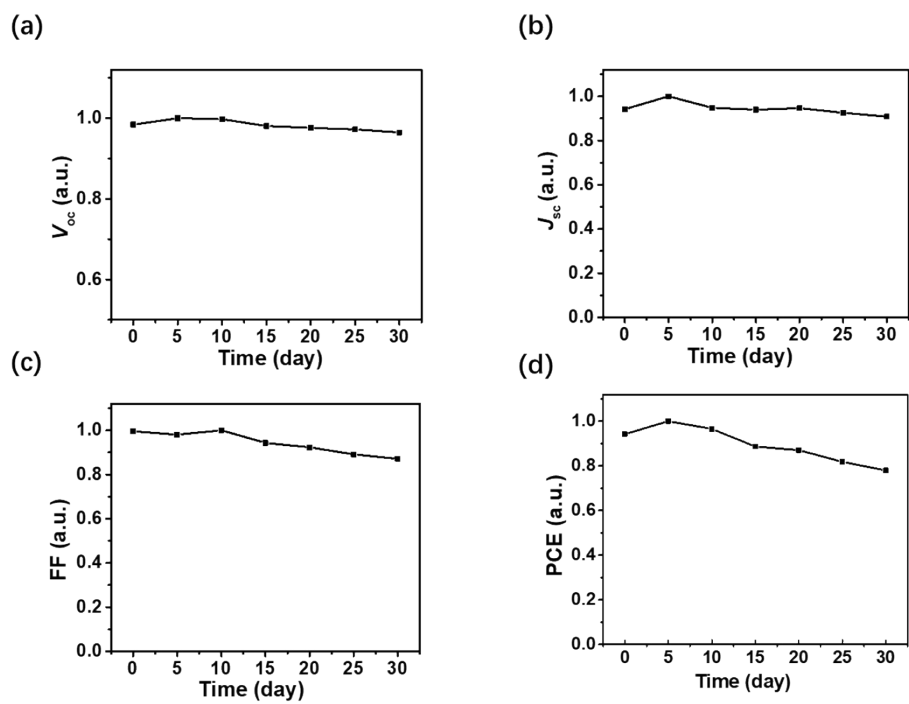


Fig. S6. Long-term J - V parameters testing of PSC based on PAD NiO film.