

Supporting Information

for

Low-temperature solution-processed p-type vanadium oxide for perovskite solar cells

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Experiments

Synthesis of VO_x sols: The VO_x sols were prepared by a melt quenching process. V₂O₅ powder (20 g) was heated to 800 °C in a ceramic crucible and was kept for 20 min. Then, the molten liquid was quickly poured into 1.0 L distilled water while stirring and a brownish solution was formed immediately. The solution was allowed to heat to the boiling point and then cool down to room temperature. After filtration and aging for more than 3 days, brownish VO_x sols were obtained. The concentration of the VO_x sols was calculated by weighting the residual mass after drying the sols in 180 °C for 24 hrs.²²

Device fabrication: The VO_x sol was spun on cleaned ITO substrates at 4000 rpm for 30 s after UV-ozone treatment for 15 min, and dried at 80~150 °C for 20 min. The perovskite precursor solution was prepared by dissolving 1.25 M CH₃NH₃I and PbI₂ mixture (molar ratio 1:1) in a DMF/DMSO mixed solvent at 1:3 volume ratio at 70 °C. The CH₃NH₃PbI₃ precursor solution was spun on top of VO_x layer at 1000 rpm for 10 s and 5000 rpm for 30 s. 400 µl toluene was dripped at the center of the wet film 15 s after the 5000 rpm spinning process started.²³ Then, the films were transferred onto a hotplate and annealed at 100 °C in N₂ (thermal annealing) or DMF vapor (solvent annealing) for 15 min. After cooling down to room temperature (RT), 40 mg ml⁻¹ dichlorobenzene (DCB) solution of PCBM was spun on top of perovskite films at 1800 rpm for 30 s, and dried at 80 °C for 20 min. Finally, 8 nm-thick bathocuproine (BCP) and 100 nm-thick Al were thermal evaporated with a mask under high vacuum (2×10^{-6} Pa). The active device area is 0.16 cm².

Measurement and characterization: The X-ray diffraction (XRD) spectra were recorded using a Rigaku diffractometer with Cu K α radiation (1.5405 Å). The transmittance spectra and ultraviolet-visible (UV-Vis) absorption spectra were recorded using an Agilent spectrophotometer (Cary 60). X-ray photoelectron spectroscopy (XPS) measurements were performed using a VG MultiLab 2000 instrument. Atomic Force Microscopy (AFM) tests were performed using the Bruker

Innova-IRIS AFM system in tapping mode. The work function of VO_x layers were measured on ITO coated glass substrates by Kelvin Probe in air. The current-voltage (*I-V*) curves of the VO_x layer on glass substrates were measured by depositing two parallel silver electrodes and providing a bias between -1.0 and 1.0 V. The current density–voltage (*J-V*) characteristics were measured using a Keithley 2400 source-meter under AM1.5G illumination at 100 mW cm⁻² from a Newport solar simulator calibrated with a Si diode (KG-5 filter). The *J-V* curves were obtained through reverse scan (1.2 V to -0.2 V) and forward scan (-0.2 V to 1.2 V) with a step of 20 mV and a delay time of 50 ms.

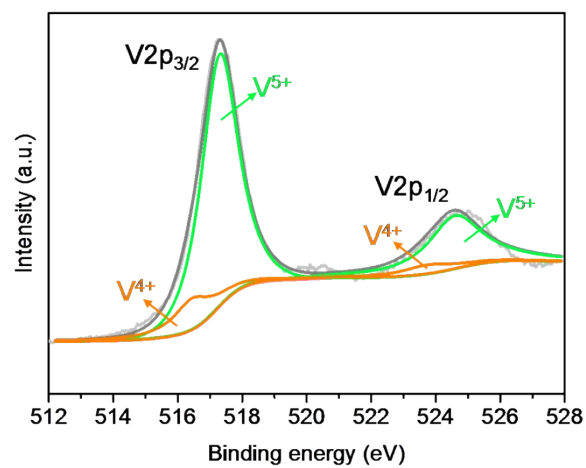


Fig. S1. XPS spectra of the as-prepared VO_x layer. The calculated V⁴⁺/V⁵⁺ ratio is 16.8%, corresponding to VO_x with x = 2.428.

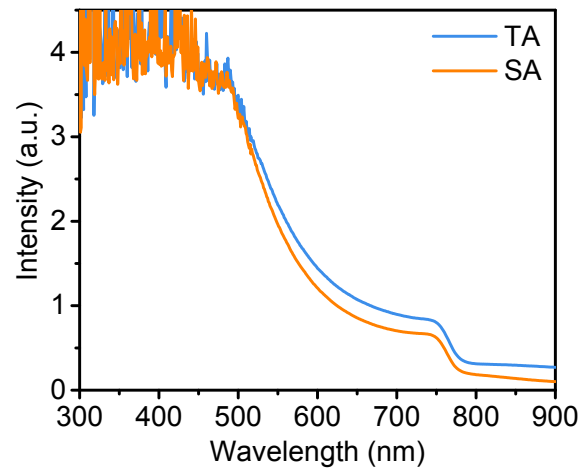


Fig. S2. Absorption spectra of CH₃NH₃PbI₃ layers (*ca.* 300 nm) processed with thermal annealing (TA) and solvent annealing (SA) process.