Perovskite Photovoltachromic Supercapacitor With All-Transparent Electrodes

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Supporting information:

Figure S1 shows the fabrication process flow of a PSC. The absorption spectra and SEM image of the perovskite thin film are presented in Figure S2a and S2b. The 300-nm-thick perovskite thin film is well crystallized with good coverage and exhibits excellent light absorption over the wavelength ranging from 400 nm to 800 nm. Figure S2c shows the *J-V* curve of a typical PSC device with Au top electrode, which exhibits a high efficiency of 16.4%.

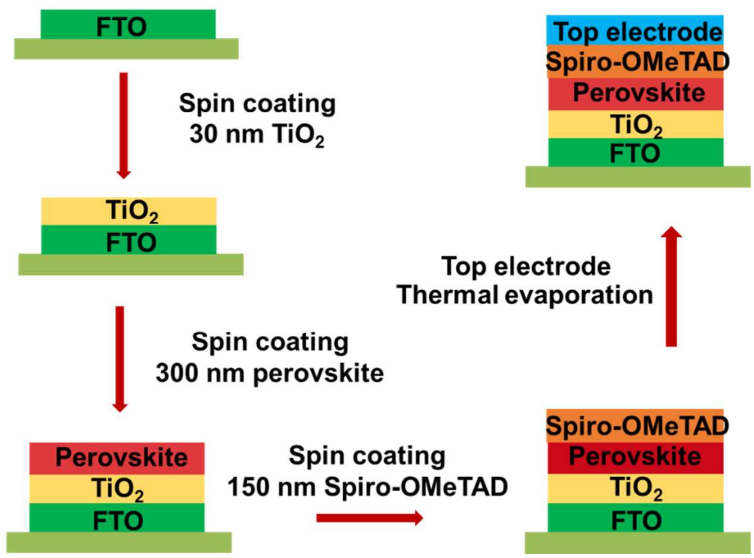

Figure S1. Fabrication process flow of the PSC.

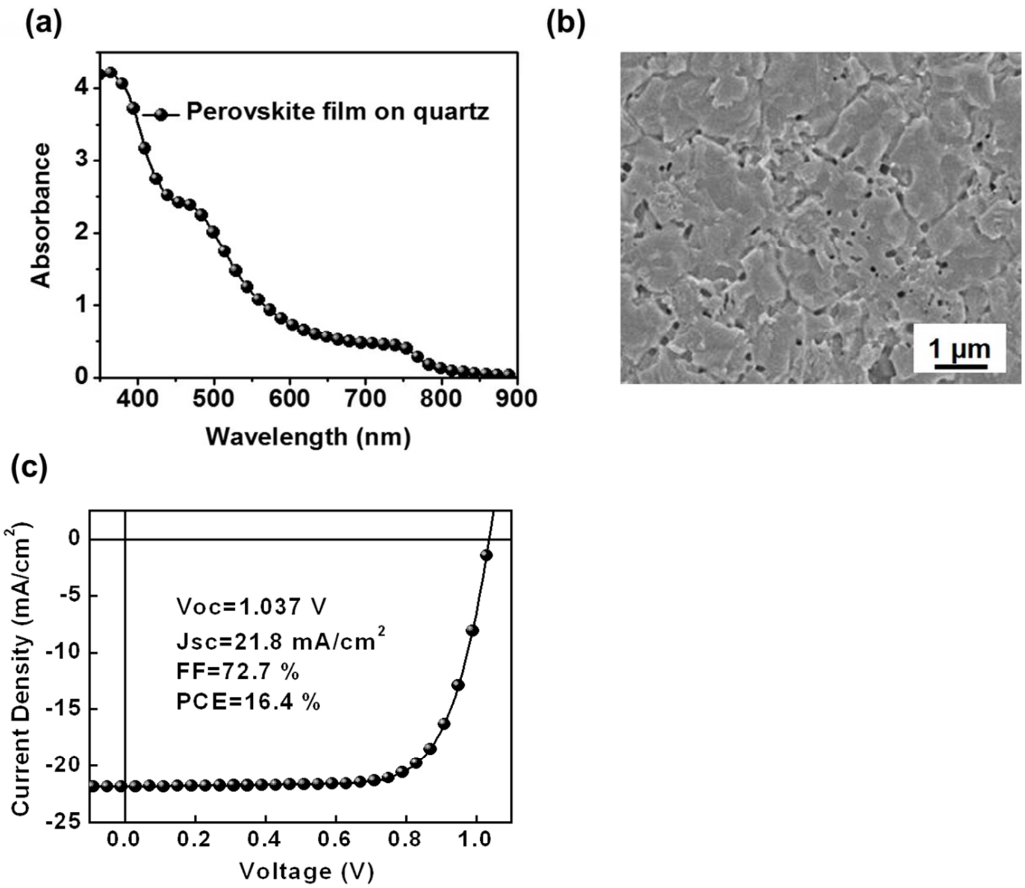

Figure S2. (a) Absorption spectra of perovskite film on quartz. (b) SEM image of perovskite film. (c) A J-V curve of a typical PSC device with Au top electrode, showing a PCE of 16.4 %.

Figure S3a shows the schematic diagram of a symmetric WO_3 electrochromic supercapacitor (ECS). A 200-nm-thick WO_3 thin film was deposited onto an fluorine-doped tin oxide (FTO) substrate as supercapacitor electrode. Polyvinyl alcohol (PVA) doped with 1 M H_2SO_4 was used as the electrolyte. The symmetric ECS was assembled by two identical WO_3 electrodes coated with PVA. Figure S3b and S3c are representative atomic force microscopy (AFM) image and scanning electron microscopy (SEM) image of the as-deposited WO_3 thin film, respectively. The nearly spherical grains are observed with a grain size of 50-200 nm and a roughness of 22.9 nm. Lots of voids are presented between the grains, providing large surface area and effective paths for ion storage and redox reaction.Figure S3d and S3e show the X-ray photoelectron microscopy (XPS) spectra of the WO_3 thin film. Four peaks are separated from the W 4f peak, corresponding to W^{6+} and W^{5+} oxidation states, respectively. The strong W^{6+} peaks suggest W^{6+} oxidation sate is the dominating state in the as-prepared WO_3 thin film, which is in agreement with the transparent state.

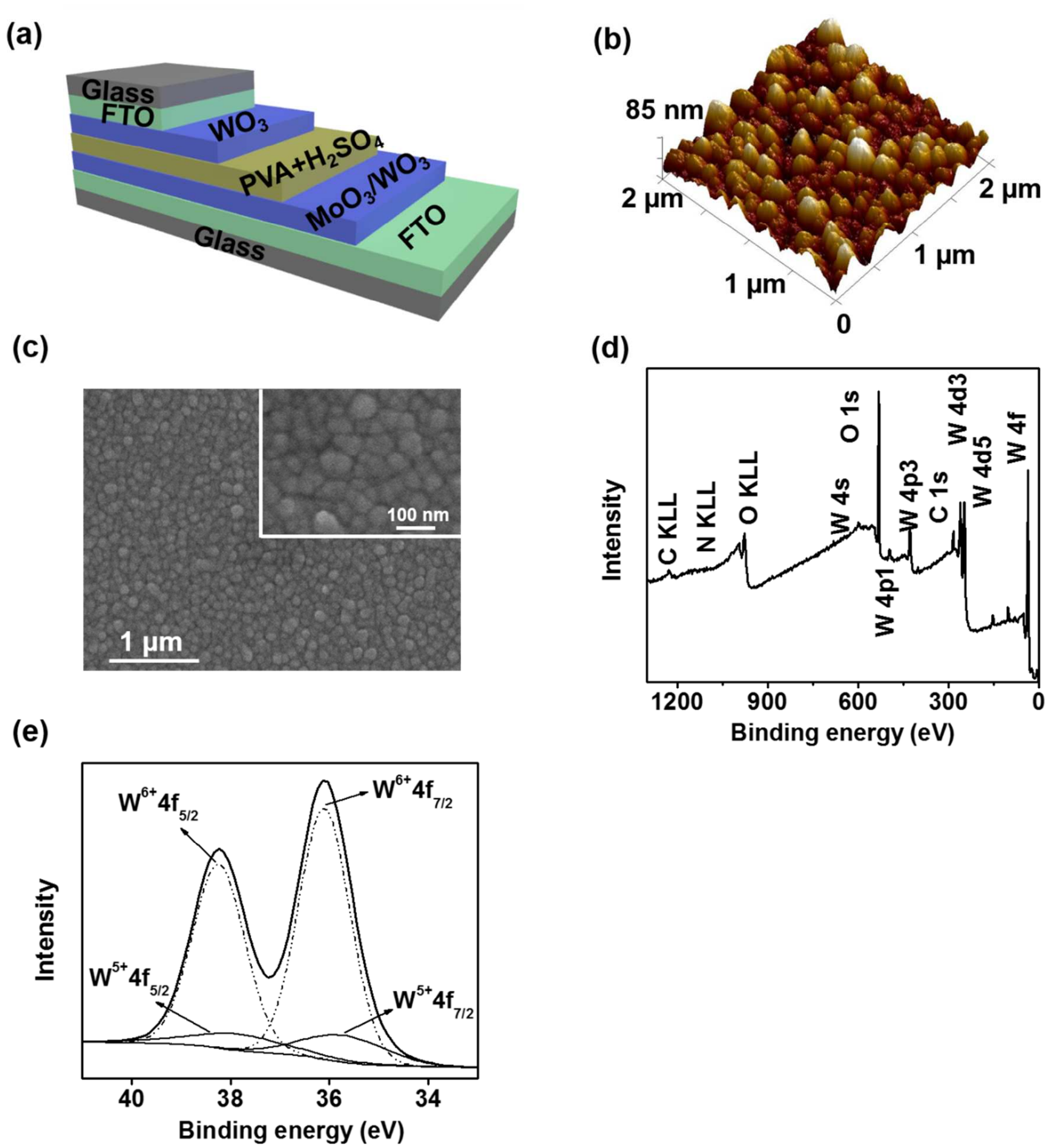

Figure S3. (a) Schematic diagram of a symmetric WO_3 ECS. (b) AFM and (c) SEM image of the as-deposited WO_3 thin film. (d) and (e) XPS spectra of the WO_3 thin film.

Figure S4 (a), (b) and (c) shows the three-dimensional schematic, cross-section schematic and photo of co-anode PVCS. We use PDMS and epoxy to seal the PSC part in order to not damage the films in PSC. Figure S4 (d) and (e) show the three-dimensional schematic and photo of co-cathode PVCS.

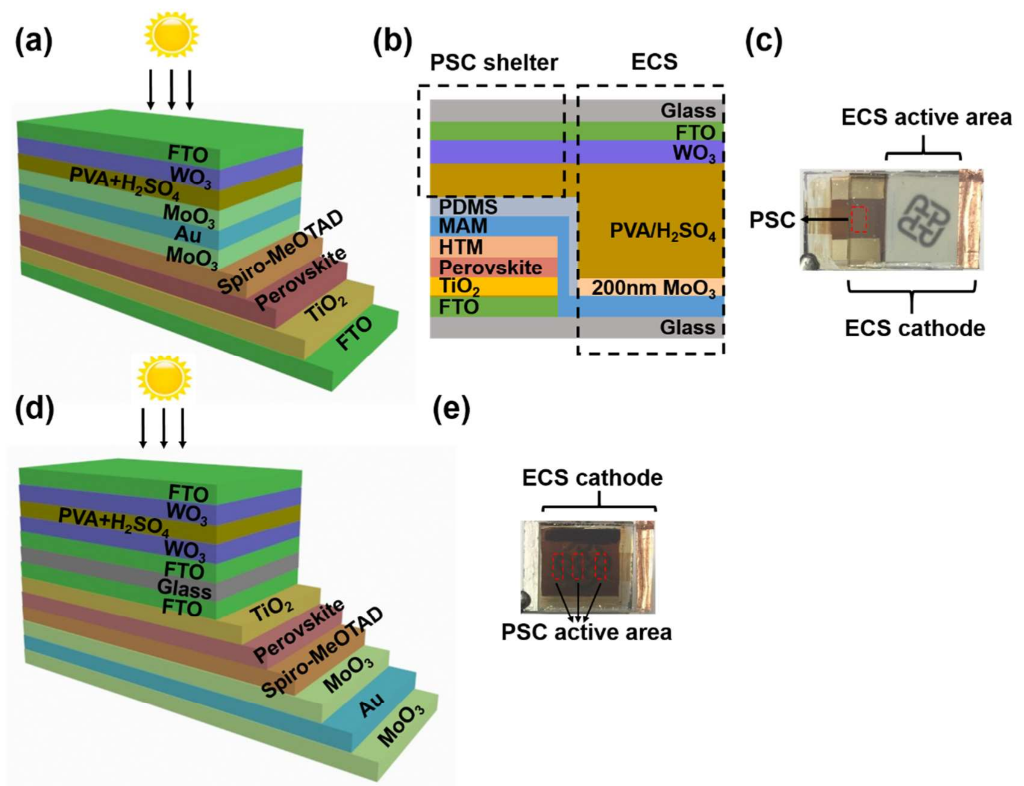

Figure S4. (a), (b) and (c) three-dimensional schematic, cross-section and photo of co-anode PVCS. (d) and (e) three-dimensional schematic and photo of co-cathode PVCS. The red dashed boxes represent the active area of PSCs.

Figure S5 shows the the integration of the PSC with MAM top electrode and symmetric WO_3 ECS connected by external wires. Figure S6a is the *J-V* characteristic of a commercial Si solar cell. The commercial Si solar cell shows a J_{sc} of 17.18 mA/cm^2 and a V_{oc} of 1.34 V. The ECS was charged by Si solar cell through external wire connection. The ECS was charged to 0.8 V within 8.5 s and reached its saturation within 35.0 s, as shown in the Figure S6b. The energy storage response is faster than the ECS charge by PSC. The energy density, power density and total photoelectric conversion efficiency of this integrated device is calculated as 61.2 mWh/m^2, 432.0 mW/m^2 and 5.51 %. These values for the device integrating PSC and ESC by external wire connection are 35.9 mWh/m^2, 461.5 mW/m^2 and 5.35%, respectively, which are comparable with the charging by the commercial solar cell. In the meantime, the PSC provides widely tunable optical transmittance. The higher energy density of ECS charged by Si solar cell can be attributed to a higher charging voltage provided by the commercial Si solar cell. Thus, the ECS and PSC integrated device is promising to be a substitute of Si based integrated energy storage device for the lower cost in the PSC fabrication.

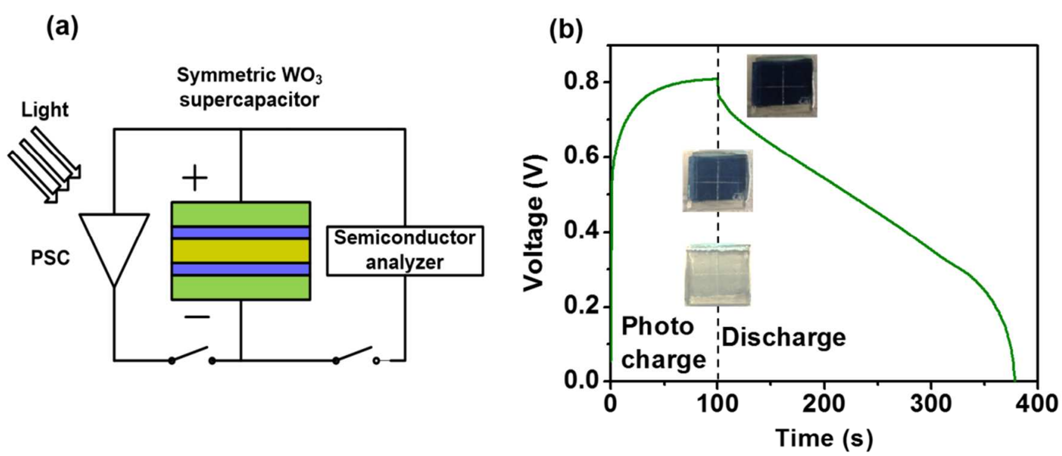

Figure S5. (a) Schematic illustration of the integration of the PSC with MAM anode and symmetric WO_3 ECS connected by external wires. (b) V-t curves of photo-charging and discharging processes and the color changes during the processes.

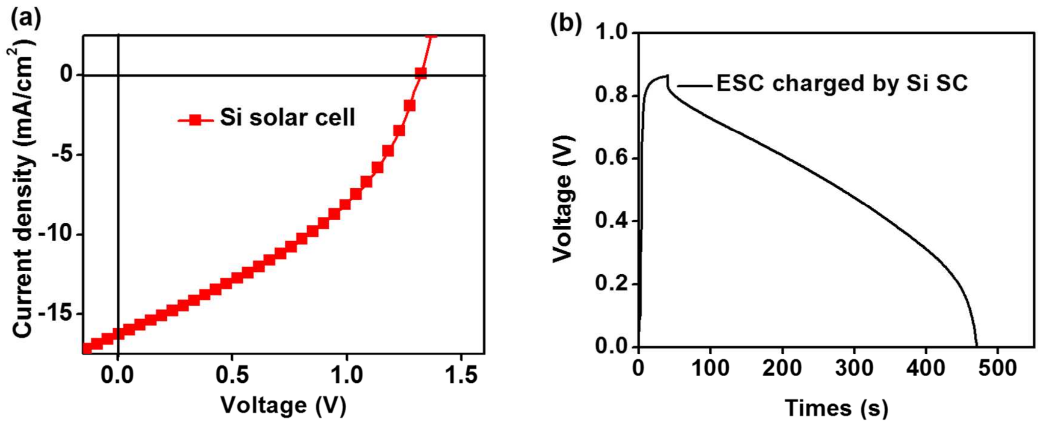

Figure S6. (a) A J-V curve of a commercial Si solar cell. (b) Photo-charging and discharging V-t curves of symmetric WO_3 ECS integrated with the commercial Si solar cell.

Figure S7a shows the areal capacitances at different current densities. The areal capacitance of the device reaches 119.5 F/cm^2 at a current density of 0.12 mA/cm^2. Figure S7b is the Ragone plot of the WO_3 ECS, which demonstrates a average power density of 6.8 W/m^2 at a charging current density of 1.20 mA/cm^2, and a energy density of 14.8 Wh/m^2 at a charging current density of 0.12 mA/cm^2.

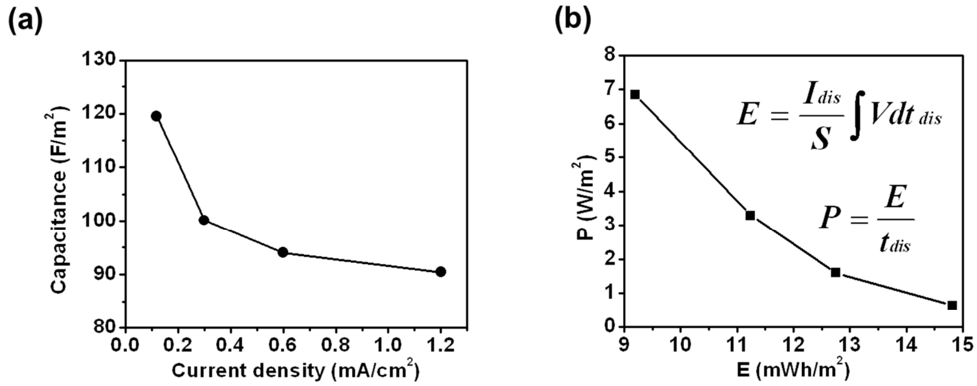

Figure S7. (a) The areal capacitances as a function of different current densities. (d) The Ragone plot of WO_3 ECS, showing the relationship between power density and energy density.

We calculated the specific capacitance and energy density according to the discharging processes. According to the discharge curves, the total energy density of device can be calculated as:

$$E_{total} = \int UdQ = \int UIdt_{dis} = I\int Udt_{dis} = IS \text{ (1)}$$

Where I is the discharge current density, U is the charged voltage, t_{dis} is the total discharge time and S is the enclosed area of the discharge curve and coordinate axis.

The average power density can be expressed as:

$$P_t = \frac{E_{total}}{t_{dis}} \text{ (2)}$$

Based on the calculated energy density, the supercapacitor areal capacitance can be calculated as:

$$E_{total} = \frac{1}{2}CU^2 \text{ (3)}$$

$$C = 2\frac{E_{total}}{U^2} \text{ (4)}$$