## **Supporting Information**

## Highly Conductive VO<sub>2</sub> treated with hydrogen for supercapacitors

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The specific surface area of as-grounded and H<sub>2</sub> treated VO<sub>2</sub> powder was 8.64  $\pm$ 1.60 m<sup>2</sup>/g and 4.97  $\pm$ 1.22 m<sup>2</sup>/g respectively obtained by nitrogen physisorption at cryogenic temperatures using the Brunauer-Emmett-Teller (BET) method.



Figure S1 SEM images of commercial VO<sub>2</sub> powder before grinding.



**Figure S2** (a) Cyclic voltammograms of pristine  $VO_2$  over a voltage window between 0 and 0.8 V in 1 M Na<sub>2</sub>SO<sub>4</sub> aqueous electrolyte at different scan rates from 2 to 1000 mV s<sup>-1</sup>; (b) galvanostatic charge/discharge behavior of pristine VO<sub>2</sub> with an applied constant current density of 1 A g<sup>-1</sup> in the potential window between 0 and 0.8 V.

The specific discharge capacitance and specific energy density according to the charge/discharge curves of pristine  $VO_2$  was calculated to be 76 F g<sup>-1</sup> and 1.8 W h kg<sup>-1</sup> respectively after the capacitance behavior became stable.



**Figure S3** The efficiency of the supercapacitor based on  $H_2$  treated  $VO_2$  as a function of cycle number.



**Figure S4** (a) Galvanostatic charge/discharge behavior of  $H_2$  treated  $VO_2$  with an applied constant current density of 5 A g<sup>-1</sup> in the potential window between 0 and 0.8 V; (b) Specific discharge capacitance and specific energy density of  $H_2$  treated  $VO_2$  as a function of cycle number.

The specific discharge capacitance and specific energy density was calculated to be around 250 F  $g^{-1}$  and 12 W h kg<sup>-1</sup> respectively after stabilization at around 200 cycles.



Figure S5 Raman spectra of the pristine VO<sub>2</sub> and H<sub>2</sub> treated VO<sub>2</sub>.

Dispersive Raman spectroscopy (SENTERRA, Bruker Co.) was used to examine the structural changes between pristine  $VO_2$  and  $H_2$  treated  $VO_2$  with an excitation laser of 532 nm and integration time of 5 s at the power of 2 mW.

The existence of triply coordinated  $V_3$ -O stretching mode resulting from edge-shared oxygens of three pyramids indicates slight oxidization have happened to the pristine VO<sub>2</sub>. Doubly coordinated V<sub>2</sub>-O bonds at 710 cm<sup>-1</sup> result from corner-shared oxygens of two pyramids. The peak located at 485 cm<sup>-1</sup> is attributed to the bending vibration of the bridging V-O-V. The apparent decrease in intensity of the peaks of V=O, V<sub>3</sub>-O, and V<sub>2</sub>-O bonds is observed in H<sub>2</sub> treated VO<sub>2</sub>, which suggests the H<sub>2</sub> treatment might reduce the vanadium valence states. Moreover, the H<sub>2</sub> treated VO<sub>2</sub> shows a broad band from 450 to 800 cm<sup>-1</sup>, which may be assigned to the partially structural distortion caused by hydrogen intercalation.



**Figure S6** Cyclic voltammograms of  $H_2$  treated VO<sub>2</sub> as working electrode in a three-electrode configuration with SCE as reference electrode and Pt wire as the counter electrode over a voltage window between -0.2 and 0.6 V in 1 M Na<sub>2</sub>SO<sub>4</sub> aqueous electrolyte at different scan rates from 2 to 1000 mV s<sup>-1</sup>.

The anodic and cathodic peaks were observed at around 0.04 V and 0.13 V vs. SCE respectively.