

Electronic Supplementary Information

A novel synthesis towards vanadium pentoxide porous nanodisk film as a cathode material for advanced Li-ion hybrid capacitor†

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Experimental section

Materials synthesis

Synthesis of VO₂ nanosheet array and V₂O₅ nanodisk thin-film cathode: All the chemicals were commercially supplied by the Sinopharm Chemical Reagent Co. Ltd., China and used without further purification. V₂O₅ nanodisk film was synthesized via a hydrothermal and template oxidation method. Typically, commercial V₂O₅ powder (0.1911 g) and oxalic acid (H₂C₂O₄·2H₂O; reducing agent) with a molar ratio of 1:3.4 were firstly added into a 35 mL mixed solution of acetone and H₂O (the volume ratio is 2:5), and then sealed with vigorous stirring at 40 °C for 18 h until a clear blue solution was obtained. Subsequently, the resulting solution was transferred into a Teflon-lined stainless steel autoclave with a piece of 40 mm-thickness steel mesh inside and kept at 180 °C for 24 h. After cooling to room temperature, washed with deionized water and ethanol, the steel mesh covered uniformly with VO₂ nanosheet array was attained. Finally, the V₂O₅ nanodisk film cathode was obtained by annealing VO₂ in air at 550 °C for 2 h with a heating rate of 2 °C min⁻¹.

Preparation of the activated carbon anode: The activated carbon (KYP-50, Kuraray), acetylene black, and polyvinylidene fluoride (PVDF) with a weight ratio of 7:2:1 were added in N-methylpyrrolidinone (NMP) solution to obtain a slurry. The resultant slurry was pasted on copper foil, and dried in a vacuum oven at 120 °C overnight.

Materials characterizations

Morphology and crystalline structure of the thin-film electrodes were characterized by scanning electron microscopy (SEM, JSM-6700F, 10 kV), transmission electron microscopy (TEM, JEM-2010FEF, 200 kV) and X-ray diffraction (XRD, Bruker D-8 Avance). The surface chemical composition of the V₂O₅ nanodisk film was analyzed by X-ray photoelectron spectroscopy (XPS, Thermo Electron, VG ESCALAB 250 spectrometer). The Raman spectra were recorded on a Witech CRM200 system with the excitation laser of 532 nm. Thermogravimetric analysis was done under a N₂ flow with a temperature ramp of 10 °C min⁻¹. Nitrogen sorption isotherms were measured

at 77 K on BELSORP-max analyzer after pretreated under vacuum at 200°C for 6 h.

Electrochemical measurements

Electrochemical performance of the V₂O₅ nanodisk film and activated carbon was examined in coin half cells with Li metal as the counter electrode. The solution of LiPF₆ (1 M) dissolved in ethylene carbonate (EC) and dimethyl carbonate (DMC) (volume ratio of 1:1) was used as the electrolyte (Guotai Huarong New Chemical Materials Co., Ltd, China). For the testing of full-cell LIHC, the V₂O₅ nanodisk film and the pre-lithiated activated carbon were used as the cathode and anode, respectively. To demonstrate the flexibility of the device, the full cell was encapsulated by using a thin aluminum plastic film. Both the half and full cells were assembled in an argon-filled glove box. Galvanostatic charge/discharge measurements were performed by a multichannel battery testing system (Land-CT2001A, Wuhan Jinnuo, China). Cyclic voltammetry (CV) was carried out on an electrochemical workstation (CHI-660, CH Instruments Ins.).

The gravimetric or volumetric energy/power densities (E and P) of the full-cell LIHC device were estimated according to equations $E = \int_{t_1}^{t_2} IV(t) dt$ and $P = E / \Delta t$, where I is the constant current density (A g⁻¹ or A cm⁻³), $V(t)$ is the working voltage at t , dt is time differential, t_1 and t_2 are the start and end of discharging time (s), and Δt is the total discharging time. The mass was based on all the active materials while the volume was based on all components including cathode, anode and separator.

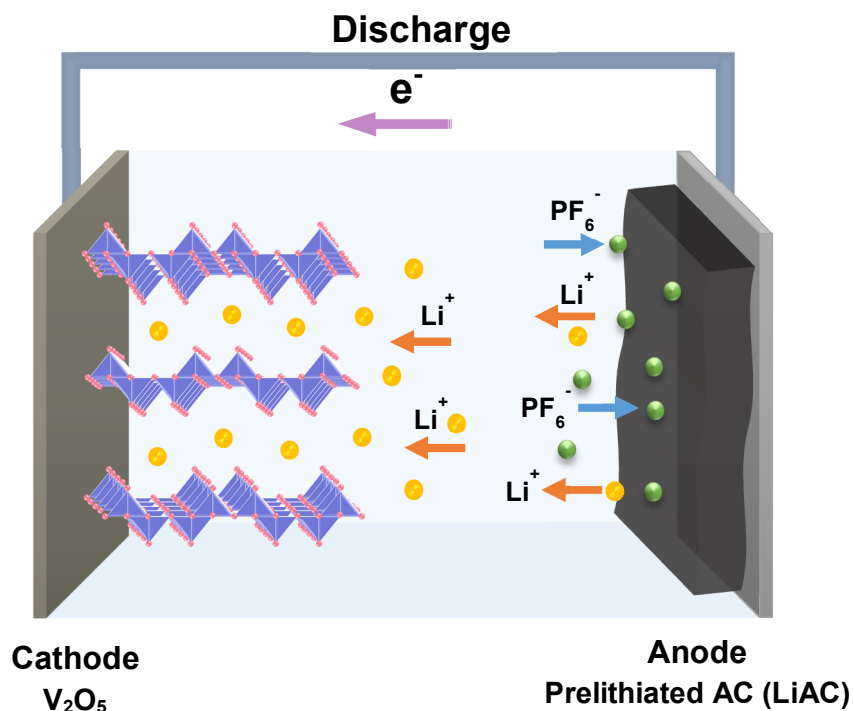


Fig. S1 The device configuration of our LIHC. The device consists of a V_2O_5 battery-type cathode and a prelithiated AC capacitive anode. During discharging process, Li^+ cations are removed from the anode and inserted into V_2O_5 cathode; at the same time, PF_6^- anions are adsorbed on the surface of anode to form electric double layer. When charged, Li^+ are deinserted from cathode and return back into anode while PF_6^- anions are desorbed from the anode. The electrons flow across the external circuit accordingly.

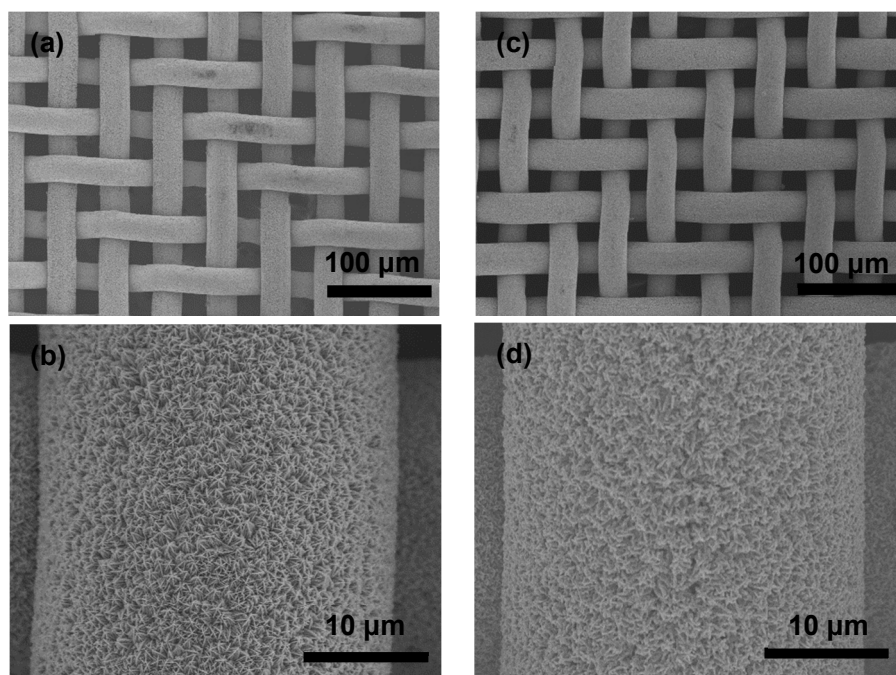


Fig. S2 SEM images of (a,b) the precursor VO_2 and (c,d) V_2O_5 .

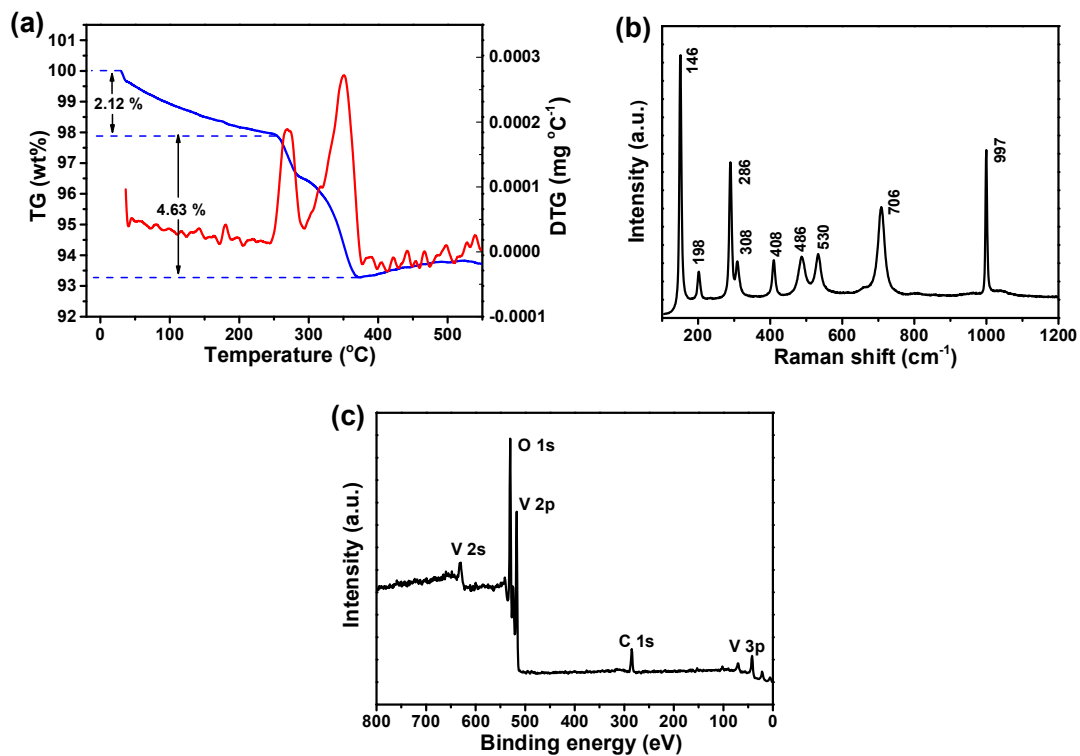


Fig. S3 (a) TGA curves, (b) Raman result and (c) XPS spectrum of the V_2O_5 film electrode.

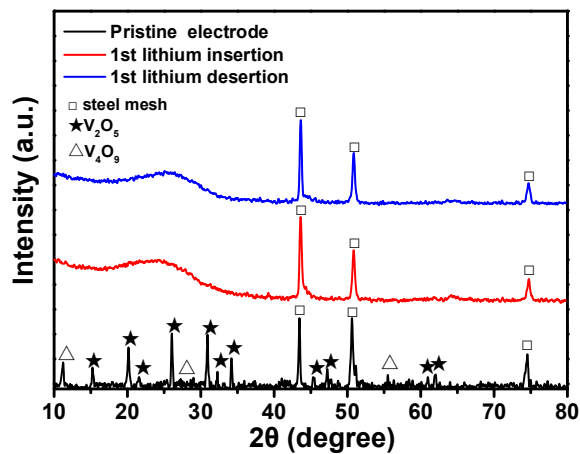


Fig. S4 XRD results of V_2O_5 cathode after the first lithiation/delithiation. It is evident that the crystallinity greatly decreases upon lithium insertion.

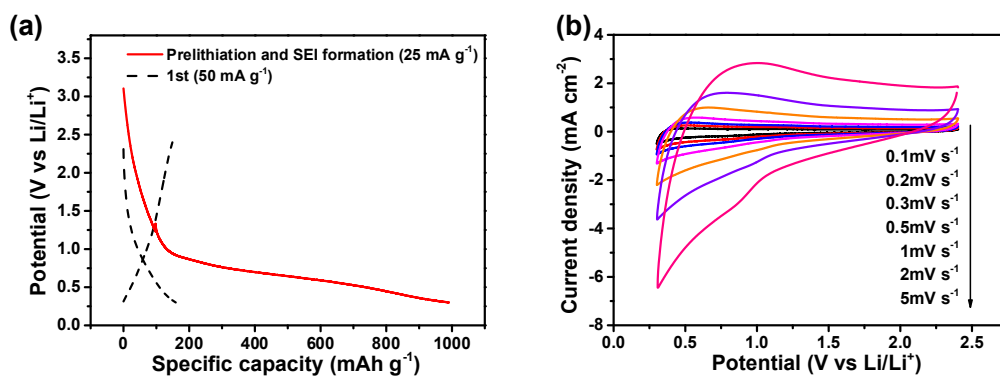


Fig. S5 (a) Prelithiation/SEI formation and subsequent 1st charge/discharge profiles of the anode. (b) CV curves of LiAC anode at different scan rates.

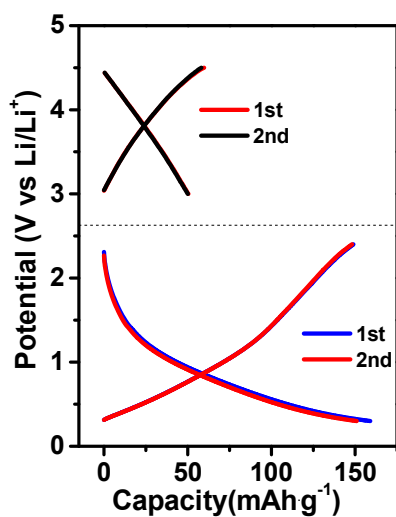


Fig. S6 Comparison of charge/discharge curves of AC as cathode and LiAC as anode. LiAC anode stores charges not merely based on electric-double-layer capacitance (EDLC), it also based on capacitive Li insertion/desertion.

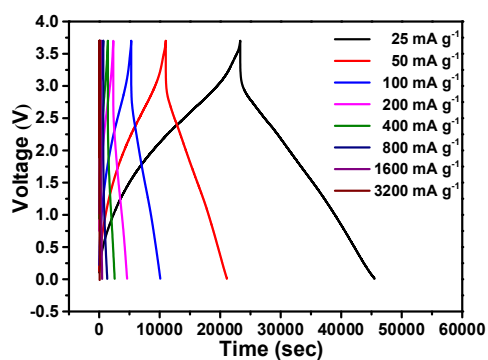


Fig. S7 Typical charge/discharge profiles of the LIHC device at various current densities