Supplementary Information for

Ultrathin V₂O₅ Nanosheet Cathodes: Realizing Ultrafast Reversible Lithium Storage

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Experimental

Liquid-exfoliation of bulk V₂O₅

All the starting materials were of analytically pure grade and used as received. In a typical procedure, bulk V_2O_5 powder (50 mg, Alfa Aesar) was added into a 100 mL glass bottle containing a formamide solution (50 mL, Sigma-Aldrich) and the whole mixture was shaken to suspend the powder and kept overnight. The resulting suspension was then sonicated at room temperature for 3 days. After ultrasonic treatment, the exfoliated V_2O_5 nanosheets were isolated from the upper solution of glass bottle via centrifugation and washed with ethanol for several times to remove residual formamide, and then dried in oven (70 °C) overnight for further characterization. The yield of V_2O_5 nanosheets is around 60%, which is determined by the weight percentage of exfoliated V_2O_5 nanosheets to the bulk V_2O_5 reactants.

Materials Characterization

X-ray powder diffraction (XRD) patterns were recorded on a Bruker AXS D8 advance X-ray diffractometer at the 2 θ range of 10 to 60° using Cu K α radiation. The morphology was investigated by using a field-emission scanning electron microscopy (FESEM) system (JEOL, Model JSM-7600F), and the nanostructure was characterized by using a transmission electron microscopy (TEM) instrument (JEOL, Model JEM-2010) operating at 200 kV. Nitrogen adsorption/desorption isotherms were conducted at 77 K (ASAP 2020). Atomic force microscopy (AFM) (Digital Instruments) was used to determine the thickness of the V₂O₅ nanosheets.

Electrochemical Measurements

The coin-type cells were assembled in an argon-filled glove-box, where both moisture and oxygen levels were less than 1 ppm. The cathodes were fabricated by mixing V₂O₅ nanosheets or bulk V₂O₅, carbon black and poly(vinyldifluoride) (PVDF) at a weight ratio of 80:10:10 in *n*-methyl-2-pyrrolidone (NMP) solvent. The resulting mixture was then pasted onto the aluminum foil and punched into small disks (\emptyset =14 mm). The working electrodes had a thickness of around 30 µm with a mass loading of ~1 mg. Lithium foils were used as anodes and the electrolyte solution was made of 1M LiPF₆ in ethylene carbonate (EC)/dimethyl carbonate (DMC) (1/1, w/w). The cells were tested on a NEWARE multi-channel battery test system with galvanostatic charge and discharge in the voltage range of 4.0-2.0 V. Electrochemical impedance spectra (frequency range: 0.001 ~ 10⁵ Hz) of V₂O₅ electrodes in coin-type cells were performed with an electrochemical workstation (CHI 660C) using lithium foils as reference and counter electrodes, and 1M LiPF₆ in EC/DMC (1/1, w/w) as the electrolyte.



Figure S1. XRD patterns of bulk V_2O_5 before (a) and after (b) immersing in the formamide solution for overnight. It can be clearly seen that the bulk V_2O_5 after immersing in the formamide solution for overnight shows a new peak at a 20 value of around 6.5° (*d*-spacing = 1.4 nm), indicating the intercalation of formamide molecules into the interlayer space of crystal V_2O_5 .



Figure S2. FESEM image of bulk V_2O_5 .



Figure S3. Nitrogen adsorption/desorption isotherms of bulk V_2O_5 and ultrathin V_2O_5 nanosheets.



Figure S4. (a) Electrochemical impedance spectra of bulk V_2O_5 and ultrathin V_2O_5 nanosheet electrodes measured at the 4th fully discharged state. The high-middle frequency semicircle represents the charge-transfer process and a straight slopping line at low frequencies corresponds to Li⁺ diffusion in solid electrode known as Warburg impedance.