Supplementary Information

Flexible Quasi-Solid-State 2.4 V Aqueous Asymmetric Microsupercapacitors with Ultrahigh Energy Density

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Preparation of VN nanosheet arrays (NSAs) on carbon nanotube flims (CNTFs)

The VN NSAs on the CNTFs were synthesized by a two-step method containing a simple solvothermal process and a subsequent annealing treatment. First of all, the CNTFs were treated in O_2 plasma at a power of 150 W for 30 min. In a classic process, 0.3 mL vanadium oxytriisopropoxide (VOT) was dissolved in 45 mL of isopropanol alcohol (IPA) under stirring. Then the solution and the pre-treated CNTFs were transferred into a 50-mL Teflon-lined stainless steel autoclave. After that, the autoclave was sealed and maintained at 200 °C for 10 h. When the autoclave naturally cooled down to room temperature, the resultant CNTFs coated by VOx NSAs were taken out, rinsed in ethanol and then dried at 60 °C overnight under vacuum. At last, the hybrid films were annealed in ammonia at 600 °C for 2 h to obtain the VN NSAs.

Electrochemical Performance Measurements

The electrochemical performance of the prepared samples was evaluated by cyclic voltammetry (CV), galvanostatic charge/discharge (GCD), and electrochemical impedance spectroscopy (EIS) measurements on an electrochemical workstation (CHI 760E, Chenhua) using a three-electrode configuration in 1 M Na_2SO_4 aqueous solution. The fabricated electrode materials were directly used as the working electrode, and Pt wire and Ag/AgCl served as the counter and reference electrodes, respectively. The EIS measurements were taken between 10^5 and 10^{-2} Hz with a voltage amplitude of 5 mV and open-circuit potential. The specific capacitance (C), energy density (E), and power density (P) were calculated according to the following equations:

$$C_A = \frac{I\Delta t}{A\Delta V} \quad , (1)$$

$$E_A = \frac{1}{2} C_A (\Delta V)^2 \ , \quad (2)$$

$$P_A = \frac{E_A}{\Delta t} , \qquad (3)$$

where I is the discharge current, Δt is the discharge time, A represents the total area of the two electrode active materials and ΔV is the potential window. The cycle life tests were conducted with GCD measurements with a constant current density of 2 mA/cm^2 for 5,000 cycles.

Characterizations of materials.

The morphologies of the samples were characterized with a scanning electron microscope (Hitachi S-4800, 5 kV). X-ray diffraction patterns were obtained with a Rigaku D/MAX2500 V with Cu Kα radiation (λ = 1.5418 Å). X-ray photoelectron spectroscopy was recorded on an ESCALab MKII X-ray photoelectron spectrometer with non-monochromatized Mg Kα X-rays as the excitation source. High-resolution TEM images were recorded on an FEI Tecnai G2 20 high-resolution transmission electron microscope at an acceleration voltage of 200 kV.

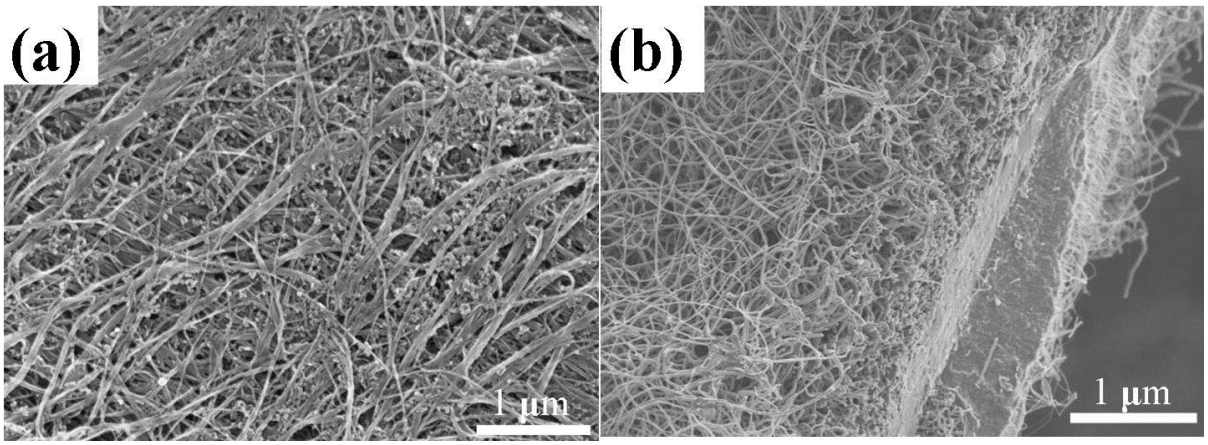

Figure S1 (a) The SEM image of the pristine CNTF. (b) The SEM image for a cross section of the as-prepared NCF@CNTF.

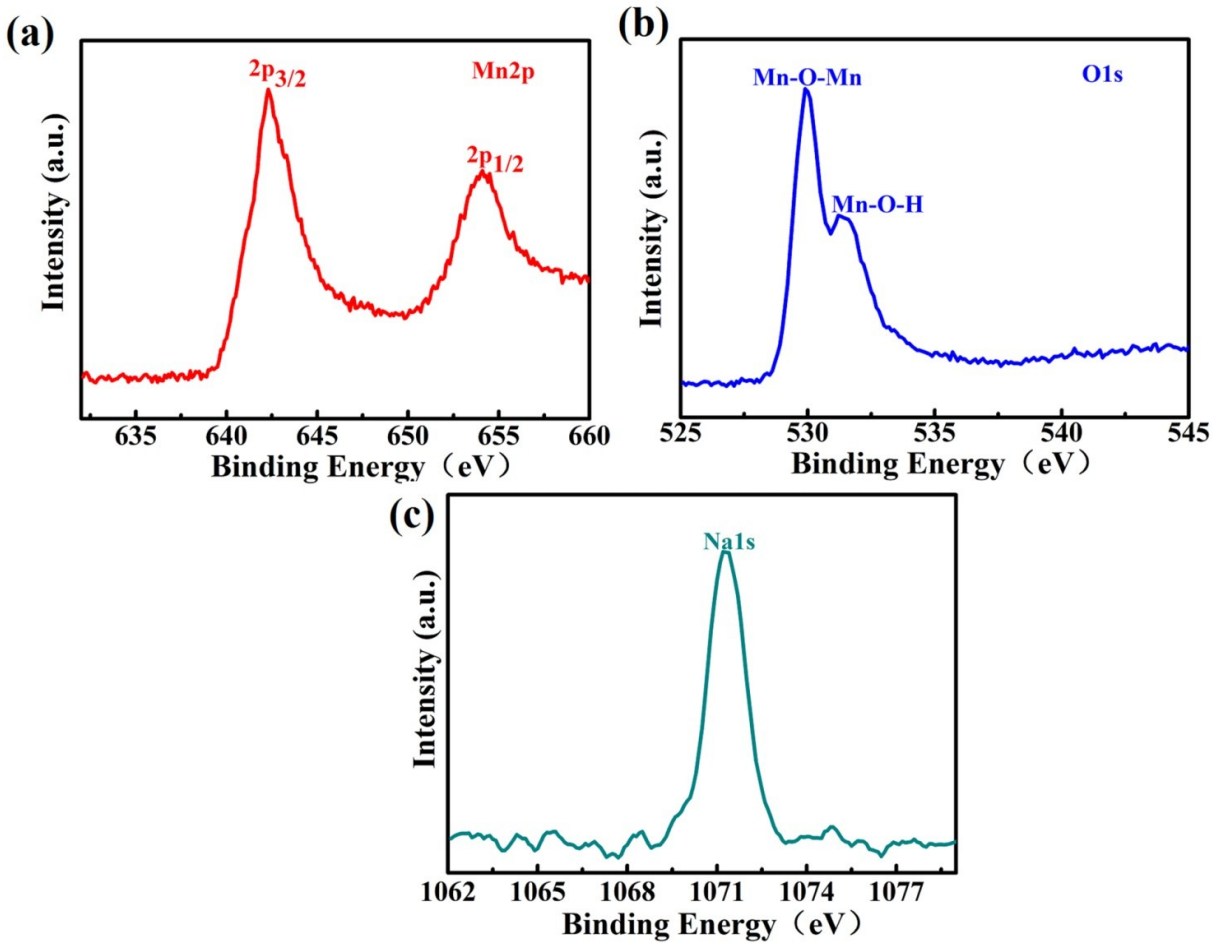

Figure S2 XPS survey of theMn2p (a), O1s (b) and Na1s (a) spectra for the Na-MnO_x.

The Mn 2p XPS spectrum shows two peaks centered at 642.6 and 654.4 eV, which can be assigned to the binding energy of Mn 2p3/2 and Mn 2p1/2, respectively, whereas the peaks in the O 1s band at 530.1 and 531.5 eV correspond to Mn-O-Mn and Mn-O-H, respectively. After insertion Na^+ into MnO_x, it is very clearly seen that an apparent peak of Na1s is located at 1071.3 eV.

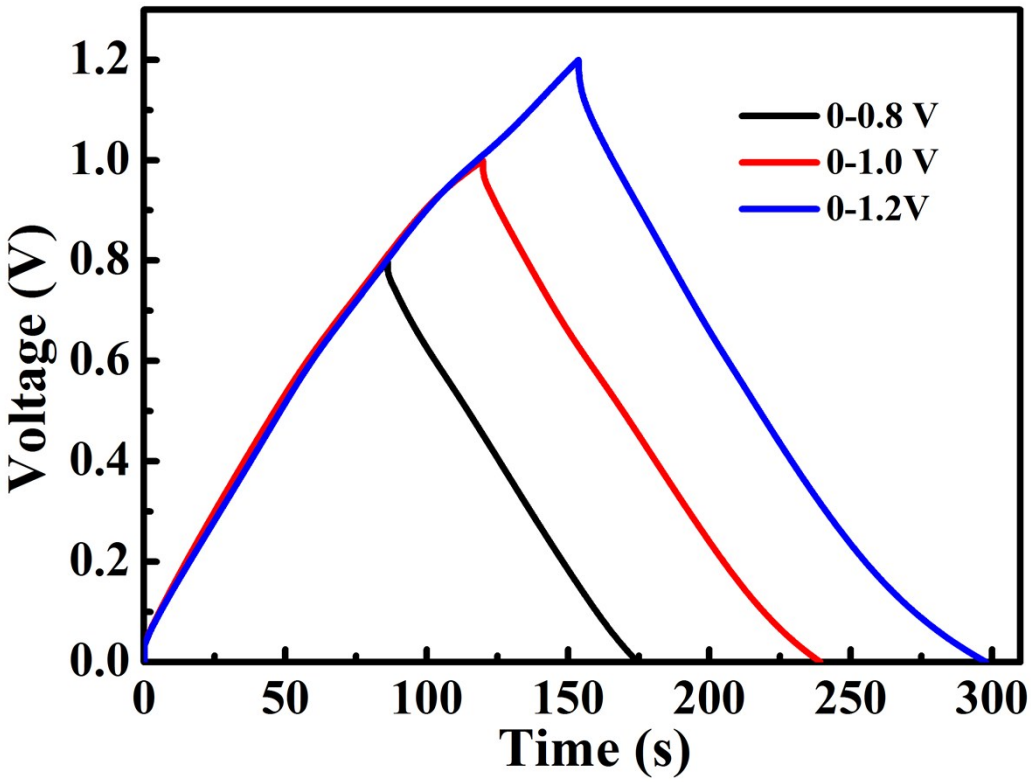

Figure S3 GCD curves of the Na-MnOx@NCF/CNTF collected over different voltages from 0.8 to 1.2 V at a current density of 4 mA/cm^2.

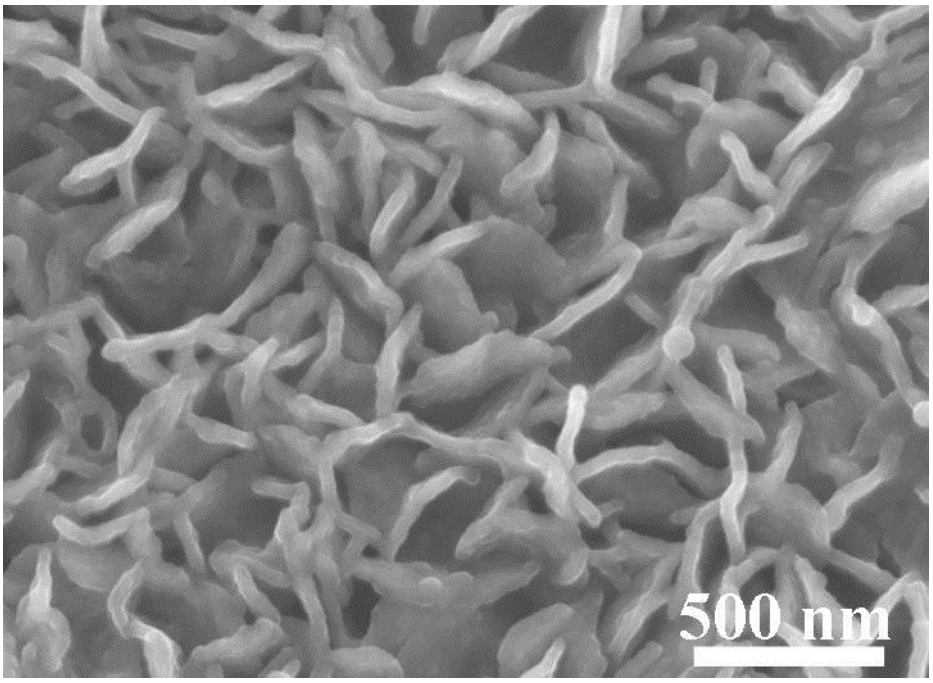

Figure S4 Scanning electron microscopy (SEM) image of VN NSAs on the CNTF.

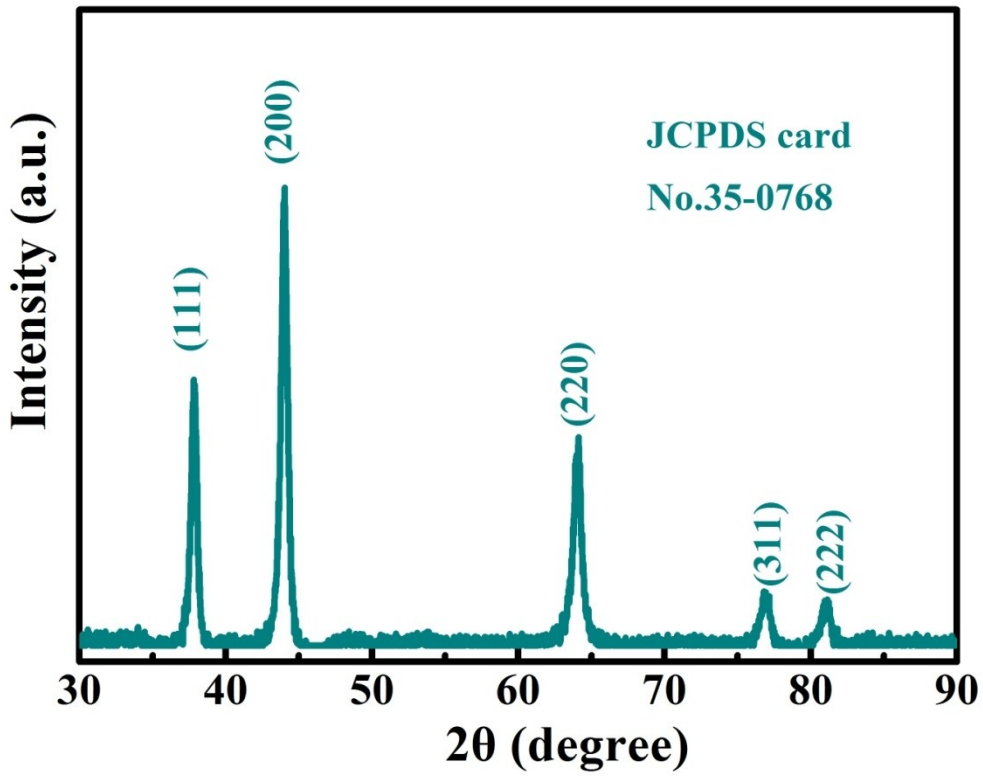

Figure S5 X-ray diffraction pattern of the VN NSAs.

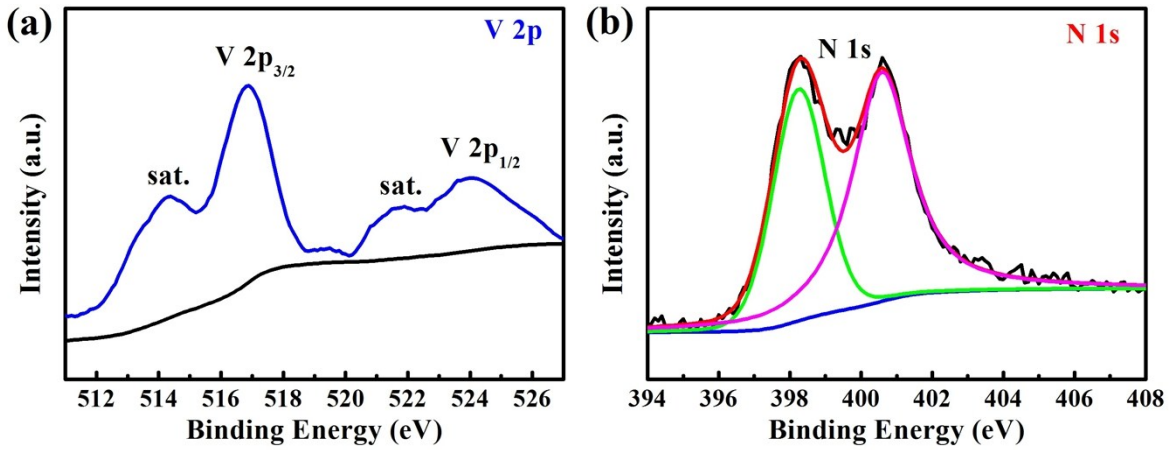

Figure S6 High-resolution XPS survey spectra of the as-prepared VN electrode material. (a) XPS spectrum of the V 2p with two peaks centered at 517 and 524 eV matching with the V $2p_{3/2}$ and V $2p_{1/2}$ spin-orbits. (b) XPS spectrum of the N 1s fitted to two peaks centered at 398 and 400 eV after deconvolution.

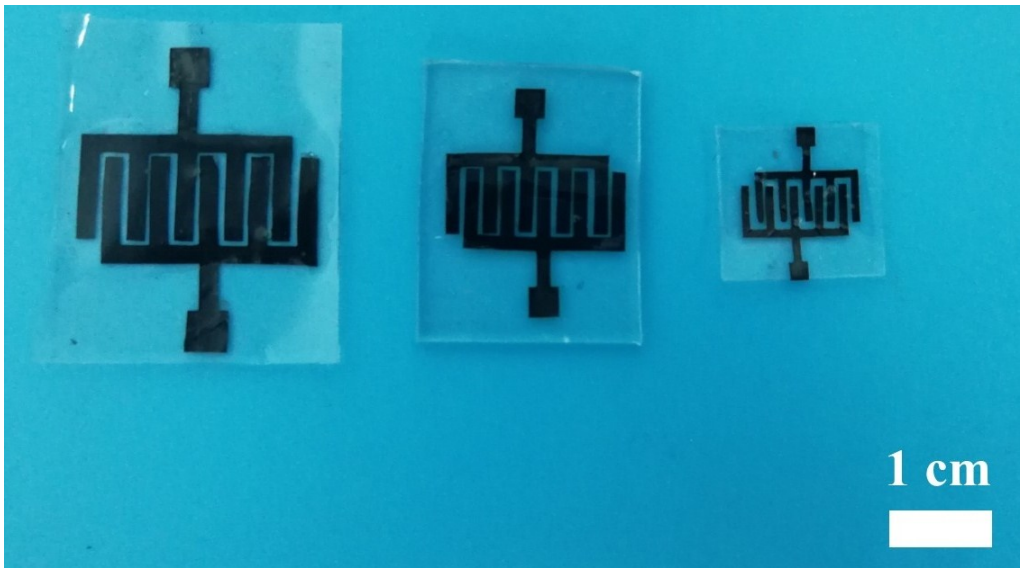

Figure S7 The digital image of the different sizes AMSCs.

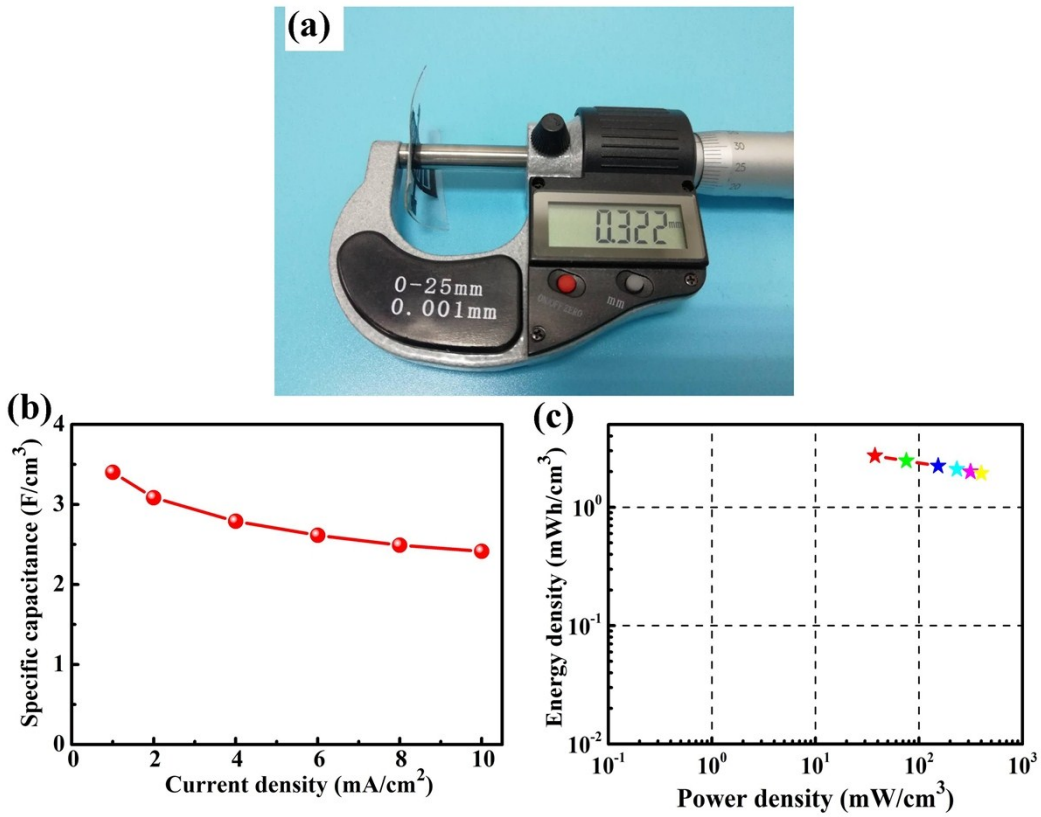

Figure S8 (a) The Photographs show the assembled AMSC, whose thickness is about 0.322 mm. (b) Volume specific capacitances calculated from the charge-discharge curves as a function of the current density. (c) Volume energy and power densities measured for our AMSC.

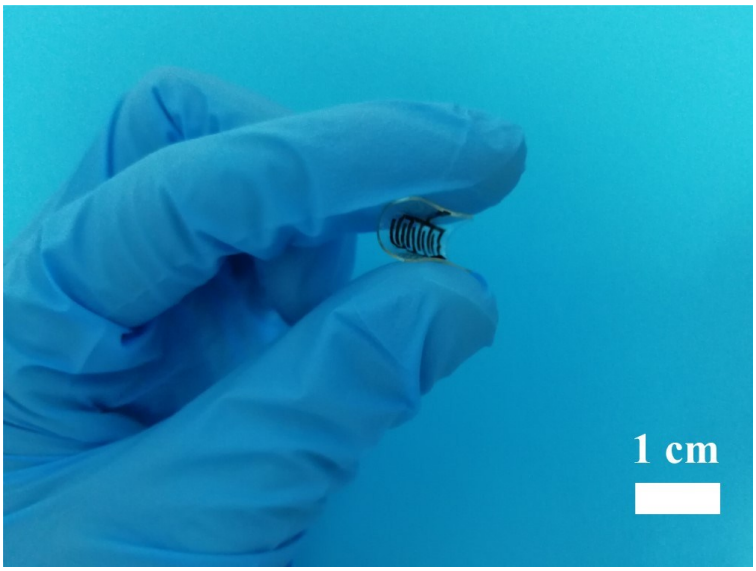

Figure S9 The optical photographs of the bended AMSC.