

Supporting Information

High-Crystalline Bismuth superstructure for Ultrastable and High-Performance Flexible Aqueous Nickel-Bismuth Battery

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Experimental Section

Synthesis of Bi and A-Bi sample on carbon cloth. Bi was synthesized on carbon cloth by electrodeposition. The electrodeposition of Bi was conducted in a solution (20 mL) containing 0.05 M Bi(NO₃)₃, 0.2 M ETDA and 0.05 M hexadecyltrimethylammonium bromide (CTMAB) at -0.9 V for 30 min. After electrodeposition, the Bi sample was washed with distilled water. To enhance the crystallinity of the Bi superstructure and adhesion between carbon fibers and these superstructure, the as-obtained Bi sample was further calcined at 200 °C under nitrogen atmosphere. Synthesis of NiO on carbon

cloth: NiO was synthesized on carbon cloth by electrodeposition of Ni and followed by oxidation. The electrodeposition of Ni was conducted in a solution (20 mL) containing 0.1 M nickel nitrate, 0.1 M nickel chloride, 0.5 M boric acid and 34.7 mM sodium lauryl sulfate at -1.4 V for 15 min. After electrodeposition, the Ni sample was washed with distilled water and then oxidized by sonicating in the solution containing 35 mL 4 M HCl and 15 mL ethanol for 10 min. The as-obtained sample was placed under ambient conditions for 30 min to derive NiO electrode.

Structural Characterization: The microstructures and compositions of the electrode materials were analyzed using field-emission SEM (FE-SEM, JSM-6330F), transmission electron microscopy (TEM, FEI Tecnai G2 F30), Raman spectroscopy (Renishaw inVia), XPS (XPS, ESCALab250, Thermo VG) and X-ray diffractometry (XRD, D8 ADVANCE). The XRD measurement was conducted using a Bruker D8 Advance X-ray diffractometer with Cu K α radiation ($\lambda = 1.5418 \text{ \AA}$) at room temperature. The scan was recorded at 40 kV and 26 mA from 10° to 80° at scan speed of 5°/min. For XRD measurement was directly carried out on the Bi samples without removing the carbon cloth substrates. Thermogravimetric analysis (TGA) curves were conducted by using a Netzsch TG209F1 analyzer.

Electrochemical Measurement: Cyclic voltammetry (CV), galvanostatic charge/discharge measurements and electrochemical impedance spectroscopy were conducted employing an electrochemical workstation (CHI 760E). The electrochemical studies of the individual electrode were performed in a three-electrode cell, with a carbon rod counter electrode and a saturated calomel electrode (SCE) reference

electrode. All the electrochemical characterizations for electrodes and aqueous Ni//Bi battery were performed in 1 M KOH aqueous solution. The upper and lower cut-off voltage of the single Bi anode during the GCD test is 0 and -1.4 V, respectively. While for the NiO//Bi battery, the upper and lower cut-off voltage is 1.8 and 0 V, respectively.

Calculations:

1. Single Electrode :

The areal capacities of the Bi and NiO electrodes were measured by galvanostatic charge/discharge method based on the following equation:

$$C_s = \frac{\int_0^{\Delta t} I \times dt}{S} \quad (1)$$

where C_a (mA h cm⁻²) is the areal capacity, I (mA) is the constant discharging current, Δt (h) is the discharging time and S (cm²) is the area of Bi electrode.

2. NiO//Bi battery:

The areal capacities of the Ni//Bi battery (C_{cell-a}) were measured by galvanostatic charge/discharge method based on the following equation:

$$C_{cell-a} = \frac{\int_0^{\Delta t} I \times dt}{S} \quad (2)$$

where C_{cell-s} (mA h cm⁻²) is the areal capacity of the of the Ni//Bi battery, I (mA) is the constant discharging current, Δt (h) is the discharging time and S (cm²) is the area of the NiO//Bi battery.

The energy density E and specific power density P of the Ni//Bi battery were obtained from the following equations:

$$E = \frac{\int_{V_1}^{V_2} C_{cell} \times dV}{d} \quad (5)$$

$$P = \frac{E}{1000 \times \Delta t} \quad (6)$$

where E (mWh cm⁻³) is the energy density of the NiO//Bi battery, C_{cell} is the specific areal capacity obtained from Equation (4), V (V) is the voltage window and d is the thickness of the Ni//Bi battery (0.08 cm). P (W cm⁻³) is the specific power density of the Ni//Bi battery and Δt (h) is the discharging time.

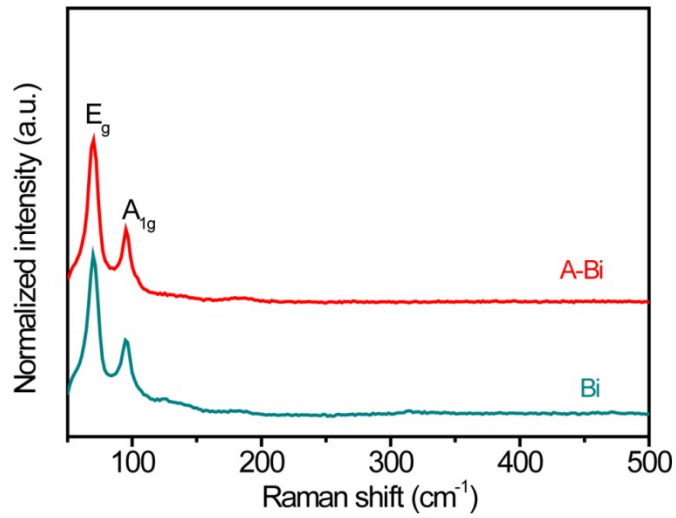


Figure S1. Raman spectr of the pristine Bi and A-Bi samples.

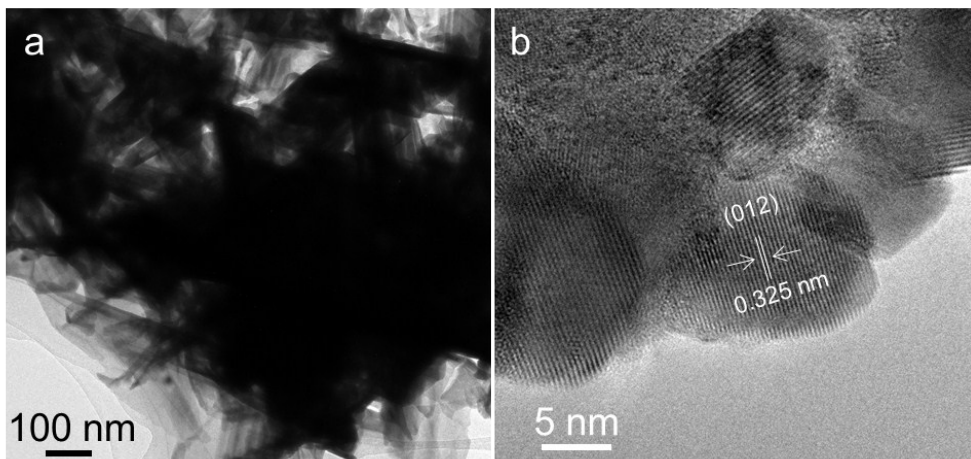


Figure S2. TEM and HRTEM images of the pristine Bi superstructure.

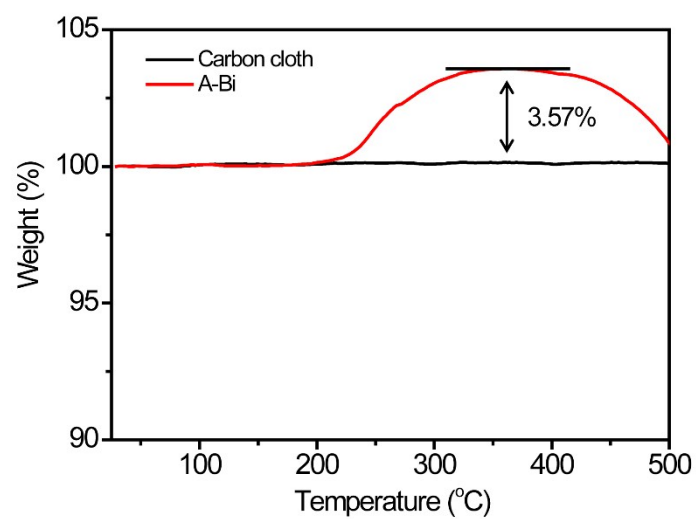


Figure S3. TGA curves for the blank carbon cloth and A-Bi sample on carbon cloth.

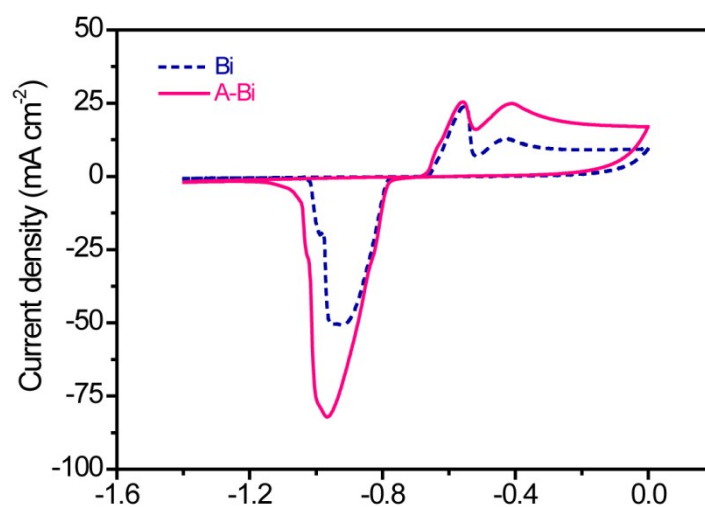


Figure S4. CV curves of the pristine Bi and A-Bi electrodes at scan rate of 10 mV s⁻¹.

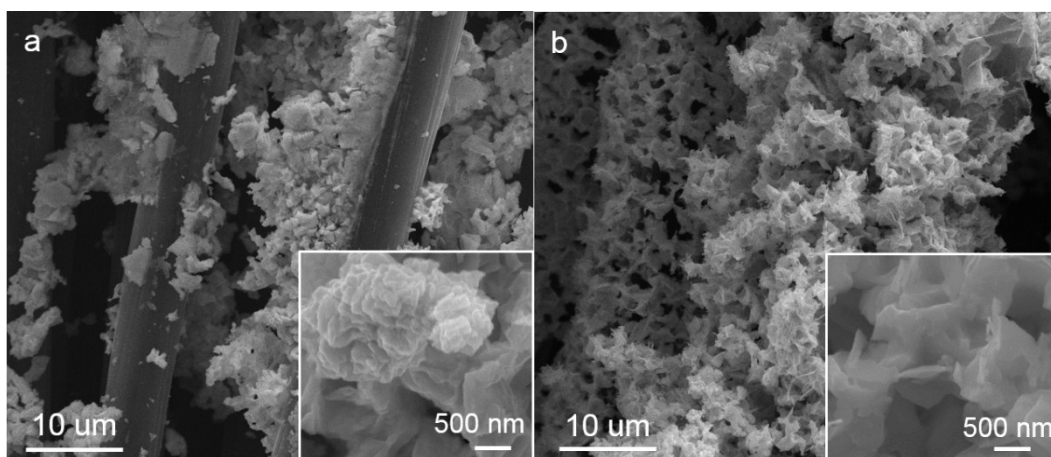


Figure S5. SEM images of (a) Bi sample and (b) A-Bi sample after cycling test.

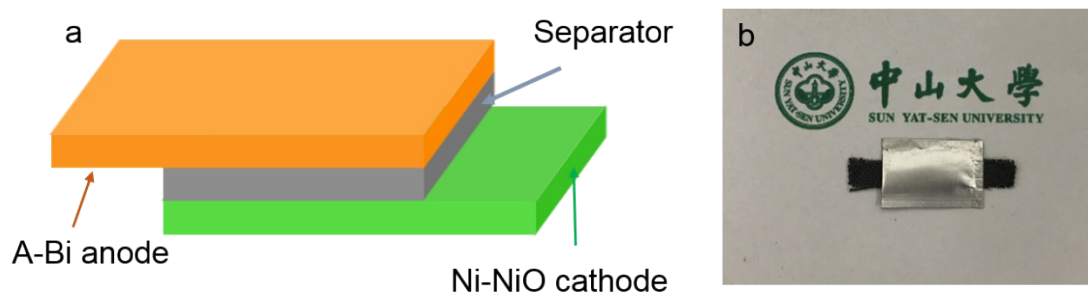


Figure S6. (a) The schematic diagram of our NiO/Bi battery. (b) The photograph of the NiO/Bi battery.

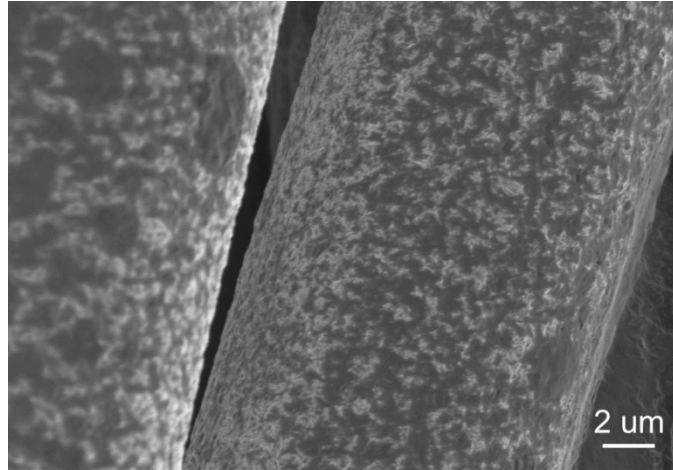


Figure S7. SEM image of the as-prepared NiO electrode on carbon cloth.

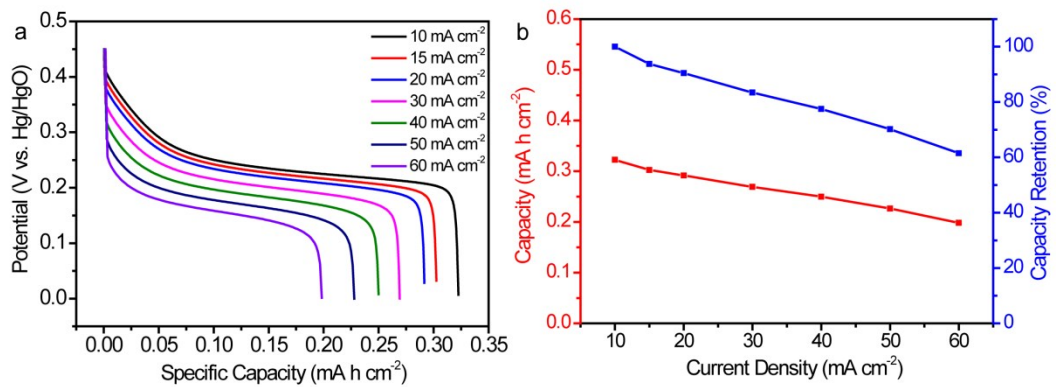


Figure S8. (a) Galvanostatic discharge curves of the NiO electrode at various current densities. (b) Areal capacity and capacity retention of the NiO electrode as a function of current density.