Supporting information

In Situ Growth of 2D BiOI Precursors on Porous Conductive Network for High Performance Bismuth Based Aqueous Battery

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Calculations
1.Single electrode:
The areal capacity was calculated by following equation:

$$C_A = \frac{\int_0^{\Delta t} I \times dt}{S} \tag{1}$$

where C_A (mAh cm^{-2}), Δt (h), I (mA) and S (cm^2) denote the areal capacity, discharging time, constant discharge current and area of the single electrode, respectively. For BiOI/PCF and Ni$_3$S$_2$/PCF, the area are 1.2 cm^2 and 3.6 cm^2, respectively.

2.Ni$_3$S$_2$//Bi full battery:
The charge balance between anode and cathode is required before the assembly of full battery, and calculated from following equations:

$$\frac{A_-}{A_+} = \frac{C_{A+}}{C_{A-}} \frac{\Delta V_+}{\Delta V_-} \tag{2}$$

where the A, C_A and ΔV represent the area, areal capacity and potential window of the electrodes, respectively.
The areal capacity of Ni$_3$S$_2$//Bi full battery ($C_{battery}$) was calculated from the galvanostatic discharging curves, as depicted in equation (1). Noted that the total area of full battery is 2.4 cm^2.
The volumetric energy and power density of the full battery was calculated from following equations:

$$E = \frac{C_{battery} \times \Delta V}{d} \qquad (3)$$

$$P = \frac{E}{1000 \times \Delta t} \qquad (4)$$

where the E (mWh cm^{-3}) is the volumetric energy density, $C_{battery}$ is the areal capacity of full battery, ΔV is the voltage platform of full battery (0.9 V) and d is the thickness of full battery (0.08 cm). P (W cm^{-3}) is the volumetric power density of the full battery, Δt represents the discharging time (h).

3.Computational methods

In this article, all the calculations were performed based on the density functional theory in conjunction with projector augmented wave (PAW) potentials, which is implemented in the Vienna ab initio Simulation Package (VASP)[1,2]. The exchange and correlation potentials are described by the generalized gradient approximation in the Perdew, Burke, and Ernzerhof (GGA-PBE)[3]. The energy cutoff for plane waves was set at 450 eV. Geometry optimizations were terminated when the total energy and atomic force are less than 10^{-5} eV and 0.05 eV Å^{-1}, respectively. A Monkhorst-Pack k-point mesh of 3×3×1 were used.

To simulate the carbon srtuctures of PCF and pristine carbon cloth, three model structures with increasing C atomic densities of 1.77, 2.19, and 2.76 g cm^{-3} were build. Each model structure contains 128 atoms in a periodic cubic cell. The first step of our simulating process is based on an *Ab initio* molecular dynamics (ABMD) simulations. For each model system, the simulation was started from a well-equilibrated melt at 8000 K. Then the system was quenched down to a temperature of 0 K in 15 ps. Finally, the resulting model structures were further relaxed within a first-principles density-functional-theory scheme.

The adsorption energy is calculated by the following equation :

$$\Delta E_{ad} = E_{Sub + Bi} - E_{Sub} - E_{Bi} \qquad (5)$$

where ΔE_{ad} is adsorption energy, $E_{Sub + Bi}$ is the total energy of Bi-adsorbing carbon based substrate, E_{Sub} is the total energy of carbon based substrate, E_{Bi} is the total energy of an isolated Bi atom.

The differential charge density is calculated by:

$$\Delta \rho = \rho_{Sub + Bi} - \rho_{Sub} - \rho_{Bi} \qquad (6)$$

where $\rho_{Sub + Bi}$, ρ_{Sub}, and ρ_{Bi} are the charge density distributions of adsorption system,

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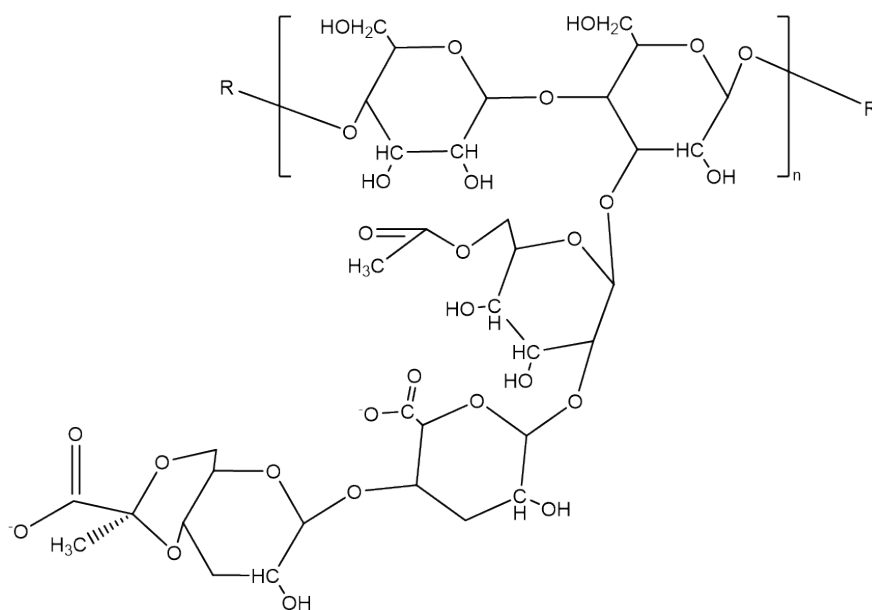carbon based substrate, and isolated Bi atom, respectively.
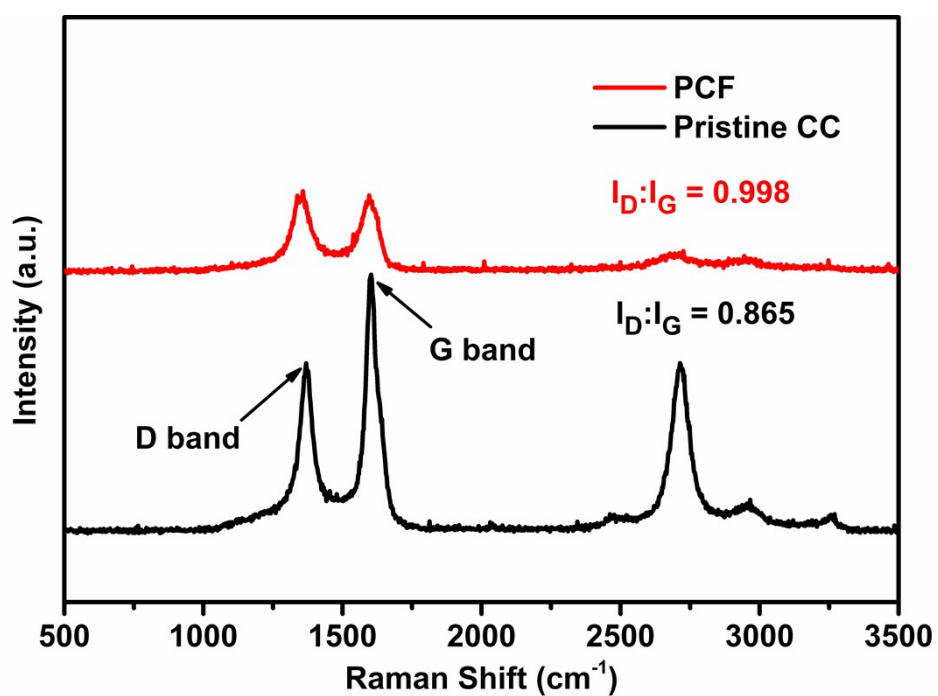

Figure S1. The molecular structure of xanthan gum

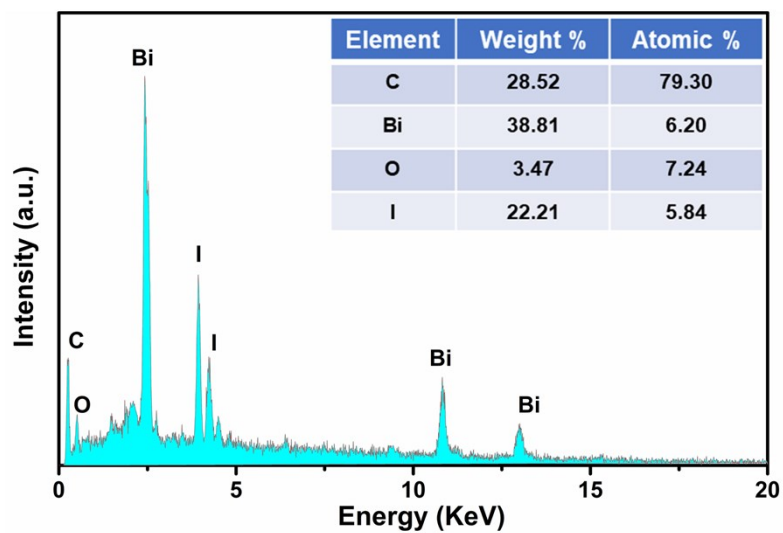
Figure S2. Raman spectra of the pristine CC and PCF samples.

Element	Weight %	Atomic %
C	28.52	79.30
Bi	38.81	6.20
O	3.47	7.24
I	22.21	5.84

Figure S3. The EDS result of BiOI NSs.
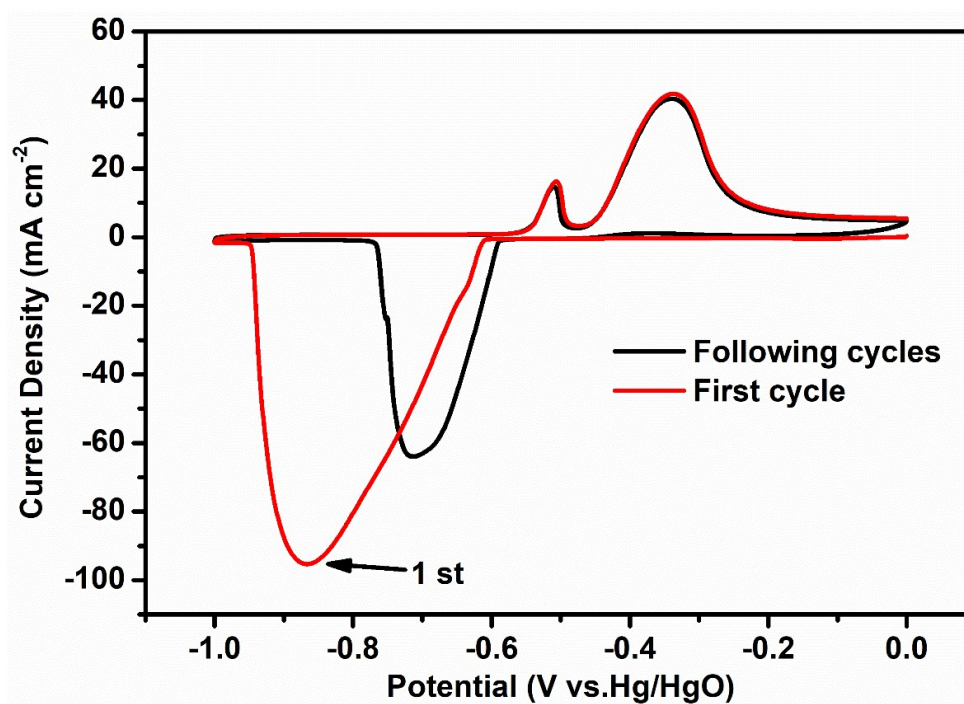
Figure S4. CV curves of BiOI/PCF sample at a scan rate of 5 mV s^{-1}
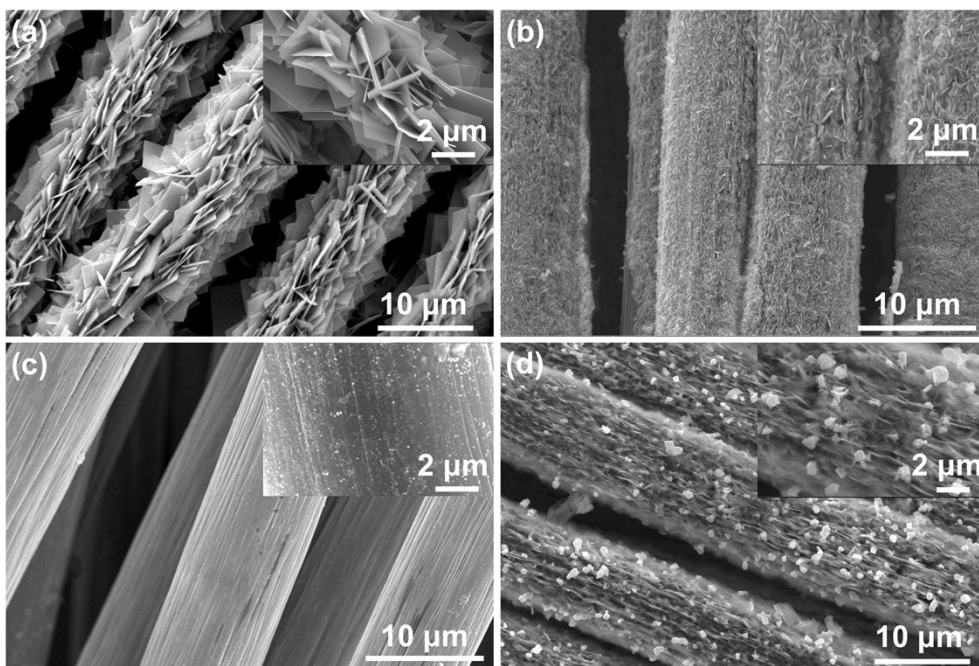
Figure S5. The SEM images of BiOI/CC and BiOI/PCF electrode before and after cycling test. (a-b) SEM images of BiOI/CC and BiOI/PCF before cycling test, (c-d) SEM images of BiOI/CC and BiOI/PCF after cycling test.
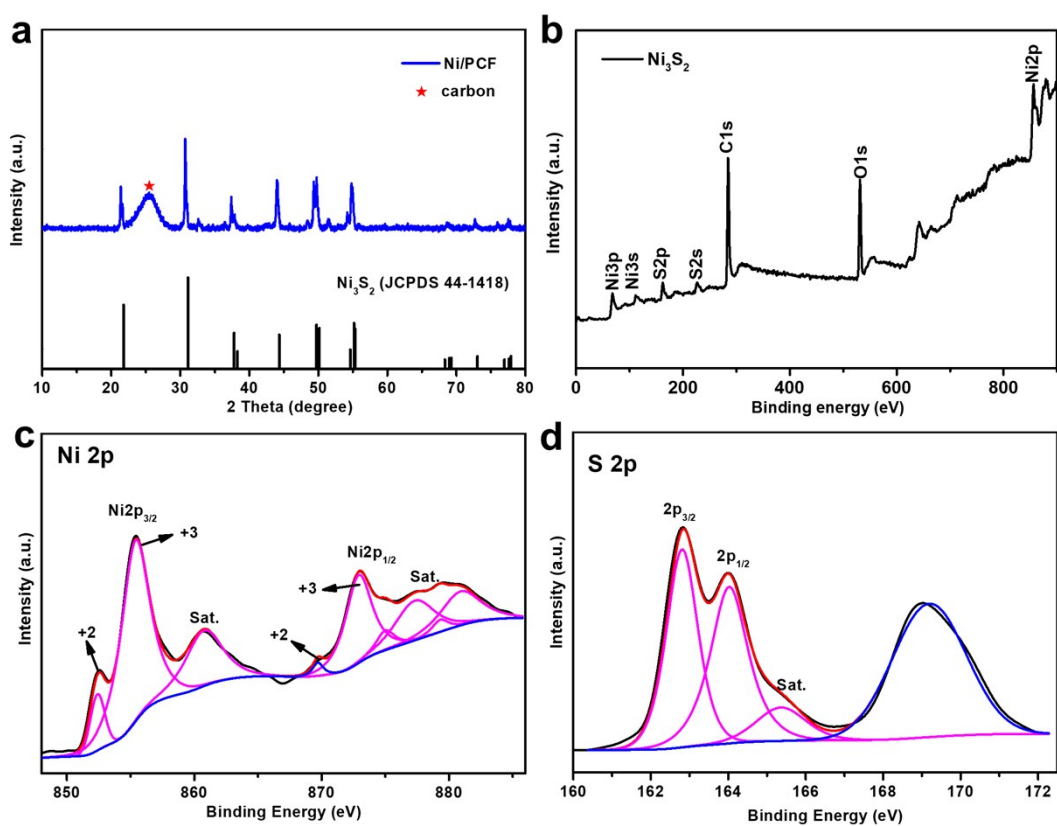

Figure S6. (a) The XRD pattern of Ni_3S_2 grown on PCF substrate.(b) XPS full spectra

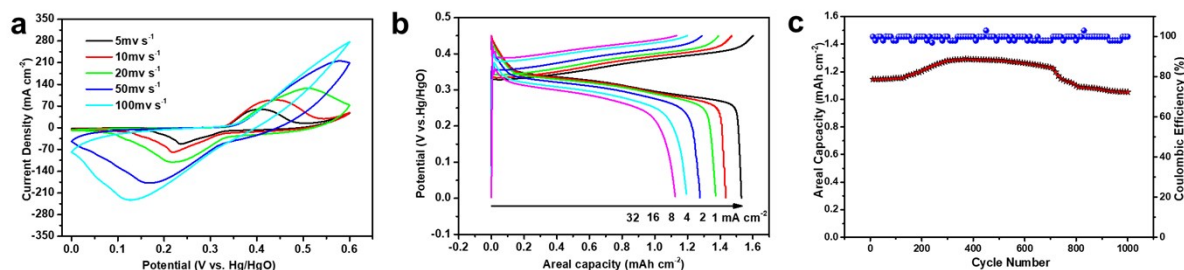and (c) Ni 2p; (d) S 2p high resolution spectra of the as-prepared Ni_3S_2.

Figure S7. The electrochemical preformances of the Ni_3S_2 cathode: (a) CV curves at different scan rates, (b) galvanostatic charge/discharge curves of the Ni_3S_2 cathode at different current densities. (c) cycling test of the Ni_3S_2 cathode at a current density of 32 mA cm^{-2}.

Reference

1. A, G. Kresse , and J. Furthmüller b. "Efficiency of ab-initio total energy calculations for metals and semiconductors using a plane-wave basis set - ScienceDirect." Computational Materials Science 6. 1(1996):15-50.
2. Kresse, G. G., and J.J. Furthmüller. "Efficient Iterative Schemes for Ab Initio Total-Energy Calculations Using a Plane-Wave Basis Set." Physical review. B, Condensed matter 54(1996):11169.
3. Perdew, J. P., K. Burke, and M. Ernzerhof. "Generalized Gradient Approximation Made Simple." Physical Review Letters 77.18(1998):3865-3868.