Supporting Information

A Versatile Sea Anemone-Inspired Strategy toward 2D Hybrid Porous Carbons from

Functional Molecular Brushes

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Experimental

Materials. 4-vinyl pyridine (4VP, Aladdin) was purified by basic alumina column chromatography to remove the inhibitor. 2,2-Azobis-(2-methylpropionitrile) (AIBN, Aladdin) was employed after recrystallization. Single-layer graphene oxide dispersion in N, N-dimethylformamide (DMF) was purchased from Hangzhou Gaoxi Technology Co., Ltd. Other chemicals and solvents were purchased from commercial sources and used without purification. *Synthesis of GO-g-P4VP*. Typically, 60 mg of GO dispersed in 48 mL of DMF was added to Schlenk and sonicated for 1 h, followed by introducing 6.4 mL of 4VP monomer. Under N₂ flow protection and vigorous stirring, 0.05 g of AIBN was added and stirred for 30 h at 65 °C. The product was collected by centrifugation and washed with DMF several times, giving GO-*g*-P4VP. The as-obtained GO-*g*-P4VP was then dispersed in DMF with a concentration of ~10 mg mL⁻¹ for further use.

For gel permeation chromatography (GPC) measurement, the free P4VP chains were also obtained through precipitating the supernatant in excess methanol, followed by filtering and drying in vacuum at 40 °C.

Synthesis of 2DHPCs. 10 mL of GO-g-P4VP dispersion was first mixed with cobalt nitrate (0.5 g), boric acid (0.8 g), or phytic acid (2 mL), respectively, and stirred overnight. The products were then centrifuged and washed with DMF several times, followed by freeze-drying for 48 h. Afterward, the 2DHPC-Co was obtained via carbonization at 800 °C for 3 h at a heating rate of 5 °C min⁻¹ in flowing N₂ in a tube furnace. The 2DHPC-B was obtained via carbonization at 800 °C for 2 h at a heating rate of 5 °C min⁻¹, followed by washing with hot water to remove the residual boric oxide. The 2DHPC-P was obtained via carbonization at 1000 °C for 2 h at a heating rate of 5 °C min⁻¹.

For comparison, G-Co was prepared by mixing GO with cobalt nitrate directly and carbonization under the same condition. GO was heat-treated at 800 °C for 2 h, giving rGO.

Material characterization. The nanomorphologies were visualized with a scanning electron microscopy (SEM, S-4800) and a transmission electron microscopy (TEM, Tecnai G2 Spirit). Powder X-ray diffraction (XRD) was performed on a D-MAX 2200 VPC diffractometer with Cu-Ka radiation. The surface characteristics of the samples were investigated using a Thermo ESCALAB 250 X-ray photoelectron spectrometer (XPS). The pore structures of the samples were measured by Micrometrics ASAP 2020. The BET surface area (S_{BET}) was analyzed by Brunauer-Emmett-Teller theory. Pore size distribution was calculated based on the original Density Function Theory (DFT). The total pore volume was measured according to the amount adsorbed at a relative pressure P/P_0 of 0.9973. The thermogravimetric analysis (TGA) was performed by using Perkinelmer PE Pyris1. The Fourier-transform infrared (FTIR) measurements of the samples were performed with IR spectroscopy (Bruker TENSOR 27), using the KBr disk method. Atomic force microscopy (AFM) characterization was carried out on Bruker Multimode 8 under tapping mode. Elemental analysis was performed on an Elementar Analysensysteme GmbH Vario EL analyzer. Macromolecular weight was measured with a Waters Breeze GPC.

Oxygen evolution reaction (OER) test. The electrochemical tests were carried out in a threeelectrode system on an electrochemical workstation (CH1660C). A glassy carbon (GC) electrode (Pine Instrument Company) was used as a working electrode, while platinum foil was utilized as the counter electrode and an Ag/AgCl worked as the reference electrode. Typically, 2 mg catalysts mixed with 50 μ L Nafion solution were dispersed in water (200 μ L) and ethanol (250 μ L) solution by sonicating for at least 30 min to form a homogeneous ink. And then, 10 μ L of the mixed solution was dropped onto a GC electrode. Pure oxygen gas was used to purge the 1 M KOH solution for 30 min to keep the solution to oxygen saturation before testing. The working electrodes were firstly activated several times until the signals were stabilized and the polarization curves were recorded in 1 M KOH. All the measurements were carried out at the rotating speeds of 1600 rpm. *Lithium-sulfur (Li-S) battery tests.* Sulfur and 2DHPC-B (7:3 by weight) were thoroughly mixed and sealed in the autoclave at 155 °C for 20 h, leading to the formation of S@2DHPC-B composite. The sulfur content of S@2DHPC-B was detected to be 67.43 wt% based on element analysis. The working electrodes were fabricated by mixing the S@2DHPC-B composite with carbon black (Super P) and polyvinylidene difluoride (PVDF) binder with a weight ratio of 7:2:1 in N-methyl-2-pyrrolidinone (NMP) and then cast onto carbon-coated Al foil substrate, followed by drying at 55 °C for 12 h. The electrolyte was 1 M lithium bis(trifluoromethylsulfonyl)imide (LiTFSI) in a mixture of 1,3-dioxolane (DOL) and dimethyl ether (DME) (1:1 by volume) with 1 wt% LiNO₃ additive. Cell assembly was carried out in an Ar-filled glovebox. The galvanostatic charge/discharge tests were performed by using a LAND CT2001A battery tester at different current densities within a cut-off voltage window of 1.7-2.8 V. The current density was 0.2 C for the first three cycles before long term cycling tests. The cyclic voltammetry (CV) study was conducted by using a CHI660C electrochemical workstation between 1.7 and 2.8 V at a scan rate of 0.2 mV s⁻¹.

Lithium polysulfide adsorption tests. Lithium polysulfide (Li₂S₆) solution was prepared by dissolving the stoichiometric amounts of sulfur and lithium sulfide (Li₂S) with a molar ratio of 5:1 in a mixed solvent of DME/DOL (1:1 by volume). The concentration of Li₂S₆ solution was set as 2 mmol L⁻¹. Typically, 5 mg of samples were placed in 2.5 mL of Li₂S₆ solution for adsorption tests.



Fig. S1 TEM image of GO-g-P4VP.



Fig. S2 FTIR spectra of GO and GO-g-P4VP.



Fig. S3 Elemental analyses of GO and GO-*g*-P4VP.



Fig. S4 SEM image of GO-g-P4VP after adsorping Co²⁺.



Fig. S5 (a) XRD pattern, (b) XPS full scan, and (c and d) high-resolution XPS spectra for 2DHPC-Co. (e) N_2 adsorption-desorption isotherms and (f) DFT pore size distribution curves of 2DHPC-Co and G-Co.



Fig. S6 (a) TEM image, (b) HRTEM image, and (c) corresponding elemental mappings of C, N, and B for 2DHPC-B.



Fig. S7 (a) SEM image, (b) HRTEM image, and (c) corresponding elemental mappings of C, N, and P for 2DHPC-P.



Fig. S8 (a) XPS full scan of 2DHPC-B. (b) The high-resolution N 1s spectrum showing the existence of graphitic quaternary N (N-Q), pyrrole N (N-5), C-N-B bonds, and pyridine N (N-6). (c) The high-resolution B 1s spectrum showing the existence of boron species, *i.e.*, B₂O₃, B-CO₂, and B-C₂O species.



Fig. S9 (a) XPS full scan of 2DHPC-P. (b) The high-resolution N 1s spectrum showing the existence of graphitic quaternary N (N-Q), pyrrole N (N-5), and pyridine N (N-6). (c) The high-resolution P 2p spectrum showing the existence of P-O and P-C species.



Fig. S10 (a) N_2 adsorption-desorption isotherm ($S_{BET} = 460 \text{ m}^2 \text{ g}^{-1}$) and (b) pore size distribution curve for 2DHPC-B.



Fig. S11 (a) N_2 adsorption-desorption isotherm (S_{BET} = 308 m² g⁻¹) and (b) pore size distribution curve for 2DHPC-P.



Fig. S12 The OER electrocatalytic activities of the recently reported catalysts.^[1-7]



Fig. S13 (a) The digital photographs of the adsorption toward Li_2S_6 on rGO and 2DHPC-B. (b) Cyclic voltammograms and (c) discharge-charge curves of S@2DHPC-B at 0.2 C for the initial 3 cycles (1 C = 1675 mA g⁻¹). (d) Rate performances of S@2DHPC-B and S@rGO at various current densities. Long-term cycle stability and Coulombic efficiency of S@2DHPC-B at (e) 1 C and (f) 3 C.

Table S1. Comparison of electrochemical properties between S@2DHPC-B and recentlyreported 2D carbon cathodes.

Sample	Electrochemical properties	Reference
S@2DHPC-B	1155-700 mAh g ⁻¹ at 0.5-3 C, 768 mAh g ⁻¹ for 100 cycles at 1 C, 373 mAh g ⁻¹ for 500 cycles at 3 C	This work
S@UHCS-900	900-~180 mAh g ⁻¹ at 0.2-5 C, ~580 mAh g ⁻¹ for 100 cycles at 1 C	[8]
Pt/graphene-Li ₂ S ₈	789 mAh g ⁻¹ for 100 cycles at 0.1 C, ~350 mAh g ⁻¹ for 300 cycles at 1 C	[9]
G-NBCL/S	1050-480 mAh g ⁻¹ at 0.2-3 C, 588 mAh g ⁻¹ for 250 cycles at 1 C	[10]
G/S	~800-~350 mAh g ⁻¹ at 0.5-5 C, 700 mAh g ⁻¹ for 70 cycles at 0.5 C	[11]
EFG-S	660-480 mAh g ⁻¹ at 1-4 C, 650 mAh g ⁻¹ for 350 cycles at 0.5 C	[12]
L-GPCS	1000-583 mAh g ⁻¹ at 0.25-5 C, ~620 mAh g ⁻¹ for 100 cycles at 0.5 C	[13]
S-NPC/G	932-786 mAh g ⁻¹ at 0.2-1 C, 608 mAh g ⁻¹ for 300 cycles at 1 C	[14]
S@Co/N-PCNSs	871-520 mAh g ⁻¹ at 0.5-5 C, 633 mAh g ⁻¹ for 200 cycles at 1 C 619 mAh g ⁻¹ for 200 cycles at 2 C	[15]

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