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Supporting Information

Hydrogen evolution performance of Co-MOF/H-g-C₃N₄ composite catalysts with different morphologies under visible light

Zhenlu Liu¹, Jing Xu^{*1,3,4}, Tong Xue^{*2}, Xinyu Liu¹, Shengming Xu¹, Zezhong Li¹

1 School of Chemistry and Chemical Engineering, North Minzu University, Yinchuan 750021, PR China

2 School of Materials Science and Engineering, North Minzu University, Yinchuan 750021, PR China

3 Ningxia Key Laboratory of Solar Chemical Conversion Technology, North Minzu University, Yinchuan 750021, PR China

4 Key Laboratory for Chemical Engineering and Technology, State Ethnic Affairs Commission, North Minzu University, Yinchuan 750021, PR China Email: wgyxj2000@163.com

2. Materials and methods

2.1 Chemicals

The chemicals (analytical grade) used in this work were obtained from Aladdin: 2-Methylimidazole ($C_4H_6N_2$), Cobaltous Nitrate Hexahydrate ($Co(NO_3)_2$ 6H₂O), Melamine ($C_3H_6N_6$), Concentrated hydrochloric acid (HCl), deionized water.

2.2 Synthesis of H-g-C₃N₄

Weigh a certain quality of melamine into the tube furnace for calcination. The initial heating temperature was 50 °C, and the temperature was increased to 520 °C at a rate of 5 °C/min, and the temperature was maintained at 520 °C for 150 min. The graphite phase carbon nitride is obtained by grinding the naturally cooled powder.

Add 1g g- C_3N_4 to a beaker of 20 ml concentrated hydrochloric acid. After stirring for 6 h, the proton fossil ink phase carbon nitride was obtained after washing and drying.

2.3 Characterization technique

X-ray diffraction (XRD): Cu Karadiation of 40 kV and 30 mA current, the crystallographic peaks in the range of 10 $^{\circ}$ to 80 $^{\circ}$, SmartlabSE.

Scanning electron microscope (SEM): ZEISS Sigma 500.

X-ray photoelectron spectroscopy (XPS): ESCALAB 250Xi. Brunner-Emmet-Teller (BET): ASAP 2020 M. Photoluminescence (PL): Fluoro Max-4. UV-vis diffuse reflectance spectra (UV–Vis DRS): Lambda 750S. Infrared spectra analysis (IR): Fourier transformation infrared spectrometer.

2.5 Electrochemistry and photocatalysis

The three electrodes of CHI 660E workstation are silver chloride, platinum tablet and sample smear respectively. 0.2 M Na₂SO₄ solution as electrolyte, sample coating dispersion is 10% Nifon solution. A 300 W xenon lamp and a 420 nm cut-off were also used in electrochemical tests.

10 mg catalyst powder and 20 mg sensitizer were evenly dispersed in a flat bottomed quartz bottle containing 30 mL TEOA. The reaction solution was successively processed by ultrasound, agitation and aeration, and then put into the multi-channel photocatalytic reaction system (Beijing PerfectLight, PCX 50B Discover). Each hydrogen-producing cylinder was sampled hourly for five hours. Each sample of 500 μ L was injected into the gas chromatographic system (Ruili SP-2000, 13X chromatographic Column), and the hydrogen production rate was obtained based on the peak area of hydrogen production and the standard curve.



Figure S1(a) SEM of g-C3N4. (b)-(c) SEM of H-g-C₃N₄. (d) SEM of Co-MOF-B. (e) SEM of Co-MOF-D. (f) SEM of Co-MOF-H.



Figure S2(a) HH-125 element scan selection. (b) Element point scanning of Co-MOF-H. (c) Element point scanning of H-g-C₃N₄. (d) Selection of HH-125 element filters. (e) Co element distribution. (f) C element distribution. (g) N element distribution.



Figure S3(a)-(c) Fine analysis of Co, N and O elements for BH-150. (d)-(f) Fine analysis of Co, N and O elements for DH-125.



 $\label{eq:s4} Figure \ S4(a) \ Nitrogen \ adsorption \ desorption \ isotherms \ for \ g-C_3N_4 \ and \ H-g-C_3N_4. \ (b) \ Aperture \ distribution \ maps \ of \ g-C3N4 \ and \ H-g-C_3N_4.$



Figure S5(a)-(c) Regulation of hydrogen production performance of BH-150. (d)-(f) Regulation of hydrogen production performance of DH-125.



Figure S6 EIS impedance diagrams of The BH-150 and DH-125.



Figure S7(a)-(c) Fluorescence decay curves of BH-150, DH-125 and HH-125.

Samples	$S_{BET}(m^2g^{-2})$	Pore volume(cm ³ g ⁻¹)	Average pore size(nm)
g-C3N4	8.27	0.042	17.16
H-g-C3N4	20.86	0.102	18.76
Co-MOF-B	13.60	0.030	7.23
Co-MOF-D	4.34	0.005	39.38
Co-MOF-H	1119.33	0.032	5.15
BH-150	12.91	0.031	11.15
DH-125	9.66	0.031	13.71
HH-125	580.76	0.039	6.81

Table 1.Specific surface areas (S_{BET}), pore volumes, and pore diameter of three samples.