Supporting Information

Enhancing hydrogen adsorption capacity of metal organic frameworks M(BDC)(TED)_{0.5} through constructing bimetallic structure

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Synthesis Information

Synthesis of Cu_{0.5}Zn_{0.5}(BDC)TED_{0.5}

A mixture of copper nitrate trihydrate (0.585 g), zinc nitrate hexahydrate (0.722 g), terephthalic acid (0.536 g), and triethylenediamine (0.906 g) was dissolved in 50ml of N, N-dimethylformamide (DMF), sonicate for 15 minutes and stir well for 15 minutes. Then the solution was transferred to a 100ml polytetrafluoroethylene reactor and kept at a crystallization temperature of 120 °C for 48 h. After cooling down to room temperature. The reaction solution was centrifuged at 11000 rpm for 5 minutes, and the supernatant was decanted. Subsequently, soak the solid precipitate in methanol for 15 minutes and centrifuge again, and this process was repeated 3 times. After centrifugation, the cleaned crystals were vacuum dried at 100 °C for 12 hours.

Synthesis of Cu_{0.5}Co_{0.5}(BDC)TED_{0.5}

A mixture of copper nitrate trihydrate (0.585 g), cobalt nitrate hexahydrate (0.705 g), terephthalic acid (0.536 g), and triethylenediamine (0.906 g) was dissolved in 50ml of N, N-dimethylformamide (DMF), sonicate for 15 minutes, and stir well for 15 minutes. Then the solution was transferred to a 100ml polytetrafluoroethylene reactor and kept at a crystallization temperature of 120 °C for 48 h. After cooling down to room temperature. The reaction solution was centrifuged at 11000 rpm for 5 minutes, and the supernatant was decanted. Subsequently, soak the solid precipitate in methanol for 15 minutes and centrifuge again, and this process was repeated 3 times. After centrifugation, the cleaned crystals were vacuum dried at 100 °C for 12 hours.

Synthesis of Cu_{0.5}Ni_{0.5}(BDC)TED_{0.5}

A mixture of copper nitrate trihydrate (0.585 g), nickel nitrate hexahydrate (0.705 g), terephthalic acid (0.536 g), and triethylenediamine (0.906 g) was dissolved in 50ml of N, N-
dimethylformamide (DMF), sonicate for 15 minutes, and stir well for 15 minutes. Then the solution was transferred to a 100ml polytetrafluoroethylene reactor and kept at a crystallization temperature of 120 °C for 48 h. After cooling down to room temperature. The reaction solution was centrifuged at 11000 rpm for 5 minutes, and the supernatant was decanted. Subsequently, soak the solid precipitate in methanol for 15 minutes and centrifuge again, and this process was repeated 3 times. After centrifugation, the cleaned crystals were vacuum dried at 100 °C for 12 hours.

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precipitate in methanol for 15 minutes and centrifuge again, and this process was repeated 3 times.

After centrifugation, the cleaned crystals were vacuum dried at 100 °C for 12 hours.

**Synthesis of Co_{0.5}Ni_{0.5}(BDC)TED_{0.5}**

A mixture of cobalt nitrate hexahydrate (0.705 g), nickel nitrate hexahydrate(0.705 g), terephthalic acid (0.536 g), and triethylenediamine (0.906 g) was dissolved in 50ml of N, N- dimethylformamide (DMF), sonicate for 15 minutes, and stir well for 15 minutes. Then the solution was transferred to a 100ml polytetrafluoroethylene reactor and kept at a crystallization temperature of 120 °C for 48 h. After cooling down to room temperature. The reaction solution was centrifuged at 11000 rpm for 5 minutes, and the supernatant was decanted. Subsequently, soak the solid precipitate in methanol for 15 minutes and centrifuge again, and this process was repeated 3 times. After centrifugation, the cleaned crystals were vacuum dried at 100 °C for 12 hours.
Supporting Figures

Figure S1 Schematic diagram of Zn(BDC)TED$_{0.5}$ unit cell structure: (light blue-nitrogen; red-oxygen; blue-zinc; gray ball-carbon; little white ball-hydrogen; yellow-tunnel)

**Note:** There are open microporous channels in the Zn(BDC)TED$_{0.5}$ unit cell structure. The channels are connected to 4 connected BDC-TED cages, sharing the pore wall, and the channel is located in the center of the unit cell structure. The structure of Zn(BDC)TED$_{0.5}$ is mainly composed of BDC-TED pillars to form the subject frame, and the metal cluster ZnO$_4$ is located in the center of the unit cell, forming the main pore structure.
Figure S2 EDS results of $M_{0.5}M_{0.5}$(BDC)$_{TED_{0.5}}$, (a): $Cu_{0.5}Co_{0.5}$(BDC)$_{TED_{0.5}}$, (b): $Cu_{0.5}Ni_{0.5}$(BDC)$_{TED_{0.5}}$, (c): $Zn_{0.5}Ni_{0.5}$(BDC)$_{TED_{0.5}}$, (d): $Cu_{0.5}Zn_{0.5}$(BDC)$_{TED_{0.5}}$ (e): $Zn_{0.5}Co_{0.5}$(BDC)$_{TED_{0.5}}$, (f): $Co_{0.5}Ni_{0.5}$(BDC)$_{TED_{0.5}}$
Figure S3 IR curve of M(BDC)TED$_{0.5}$

**Note:** According to the infrared absorption spectrum of M(BDC)TED$_{0.5}$ crystal, its infrared absorption peaks mainly appear in two regions:

1. The absorption peak near 748 cm$^{-1}$ is the M-O stretching vibration of the metal MO cluster in the M(BDC)TED$_{0.5}$;

2. The two infrared absorption peaks at 1350-1650 cm$^{-1}$ are the C-O vibration peak of the carboxyl group in the organic ligand, which is mainly caused by the symmetric vibration peak of O=C-O at 1386 cm$^{-1}$ and the asymmetric vibration at 1623 cm$^{-1}$.
Figure S4 Thermogravimetric curve of the samples

Figure S5 Hydrogen adsorption isotherms of M(BDC)TED$_{0.5}$

Figure S6 Hydrogen adsorption isotherms of M$_x$M$_{1-x}$(BDC)TED$_{0.5}$
Figure S7 Hydrogen adsorption isotherm of Cu$_{0.625}$Ni$_{0.375}$(BDC)TED$_{0.5}$ at 273 K

Figure S8 XRD patterns of Cu$_{0.625}$Ni$_{0.375}$(BDC)TED$_{0.5}$ before and after adsorption