Supporting information

Thermochemical transformation in single-step synthesis zeolitic imidazole frameworks under solvent-free

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Content:

1. Material synthesis

1.1. Materials

All the chemical reagents for synthesis materials and substrate of reaction were purchased from Aladdin chemical and used as received without any further purification. The solvents, such as methanol etc., were directly used after purchase from Sinopharm Chemical.

1.2. Thermochemical synthesis

The alumina ceramic boat (30x60x15 mm) containing template materials (250-300 mg of ZIFs) transferred into a quartz tube (OD 60mm, 1000mm), which installed in the muffle furnace (TL 1200, Nanjing Bo Yun Tong Instrument Technology Co.Ltd.). The system purged with Ar underflow rate 50 $cm^3 \cdot min^{-1}$ (mass flow controller, S48-32HMT, Nanjing Bo Yun Tong Instrument Technology Co.Ltd.) for 30 min before ramping temperature. The thermal treatment process was programmable controlling temperature with an external thermocouple detector under the heating rate of $10°C \cdot min^{-1}$. The target temperature was kept for curtain time before natural cooling to room temperature. Additional, the system was continuously flowing with 50 $cm^3 \cdot min^{-1}$ of Ar until the correcting product. The synthesized materials were stored in a desiccator to avoid the moisture adsorption for further used in application or characterization. The synthesized materials were weight based on the weight of template material, which higher weight loss observed in the higher temperature.

2. Characteristic properties of materials

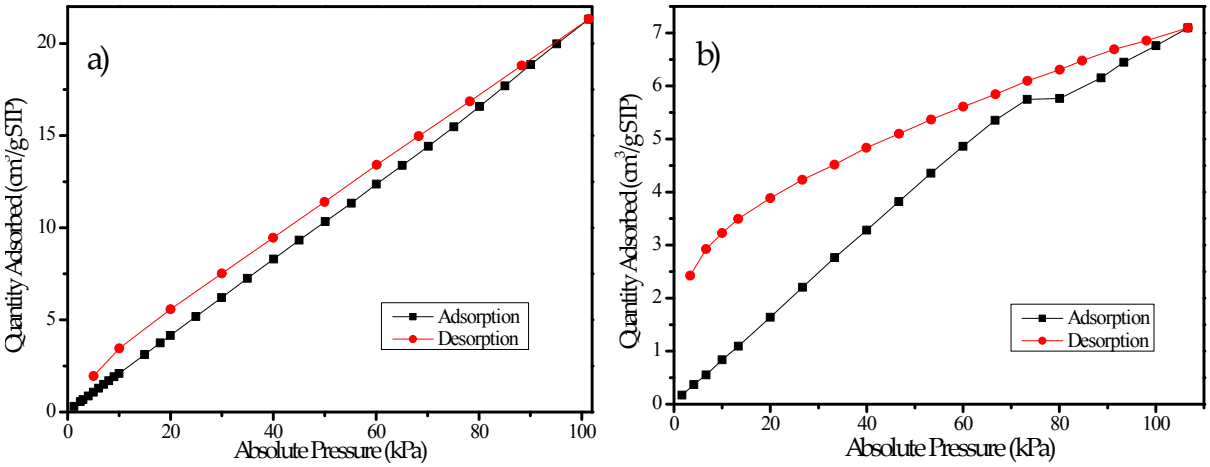


Figure S1. The CO_2 (a) and CH_4 (b) adsorption at 273K on T-ZIF-67

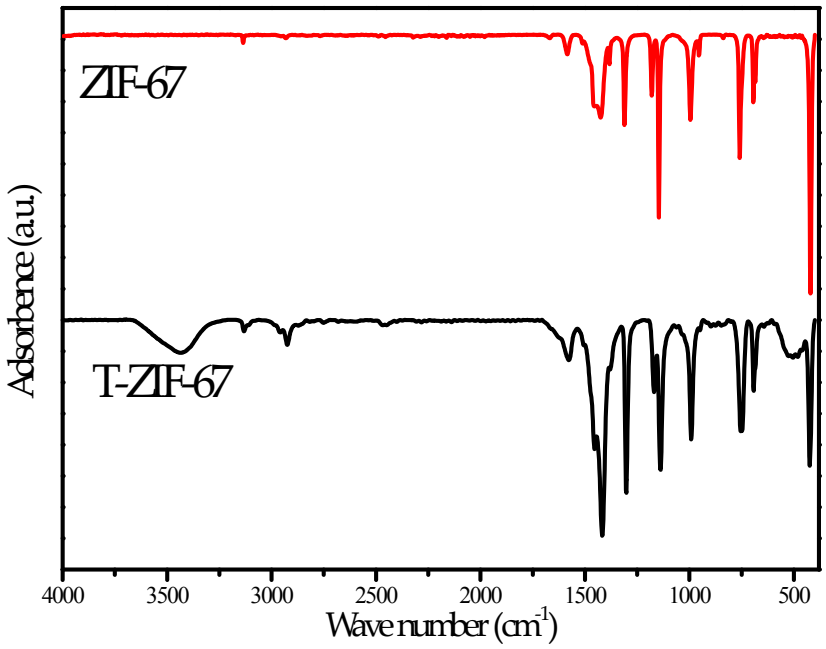
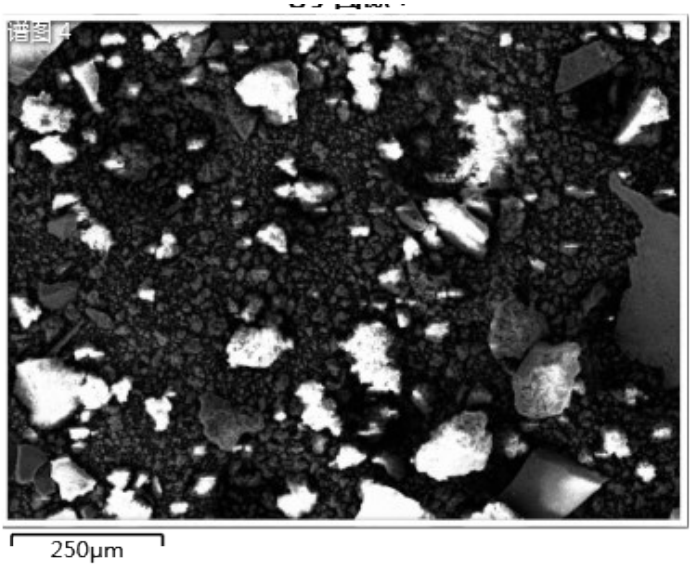


Figure S2. The FT-IR spectra of T-ZIF-67 comparable with reported ZIF-67 (CrystEngComm, 2018, 20, 3601, DOI: 10.1039/c8ce00392k)



Elements	wt%
C	41.68
N	19.39
O	6.00
Co	32.92

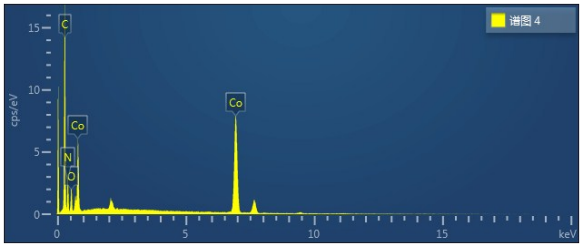


Figure S3. The element content in T-ZIF-67 via scanning electron microscopy energy dispersive spectrometry (SEM-EDS)

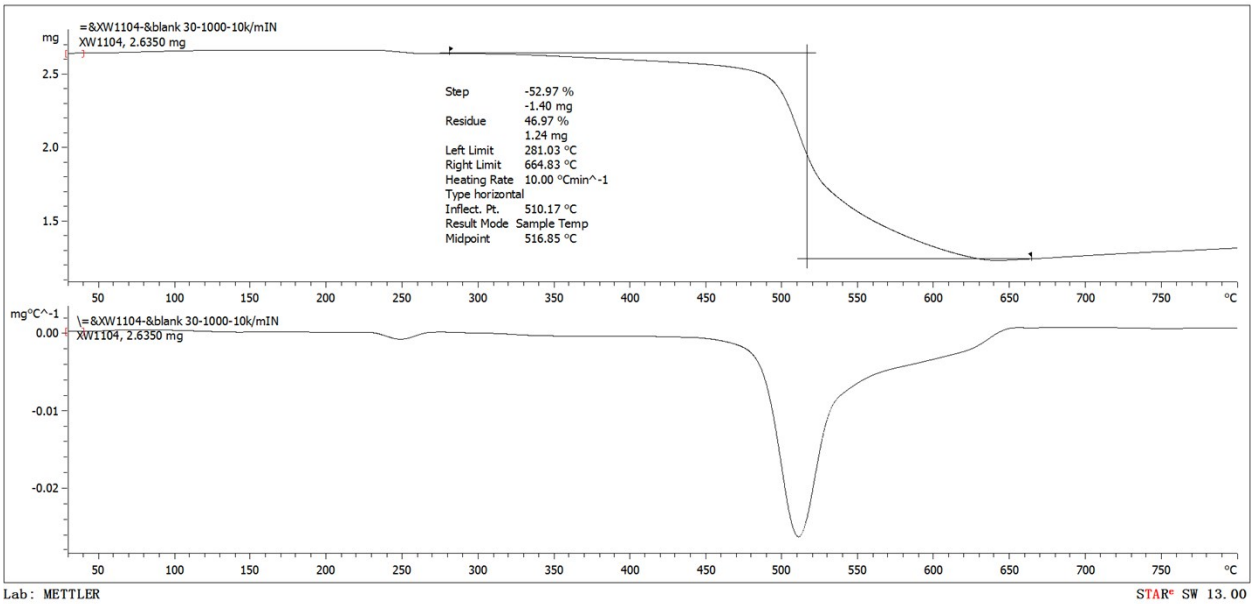


Figure S4. The thermal gravimetric analysis (TGA) of T-ZIF-67

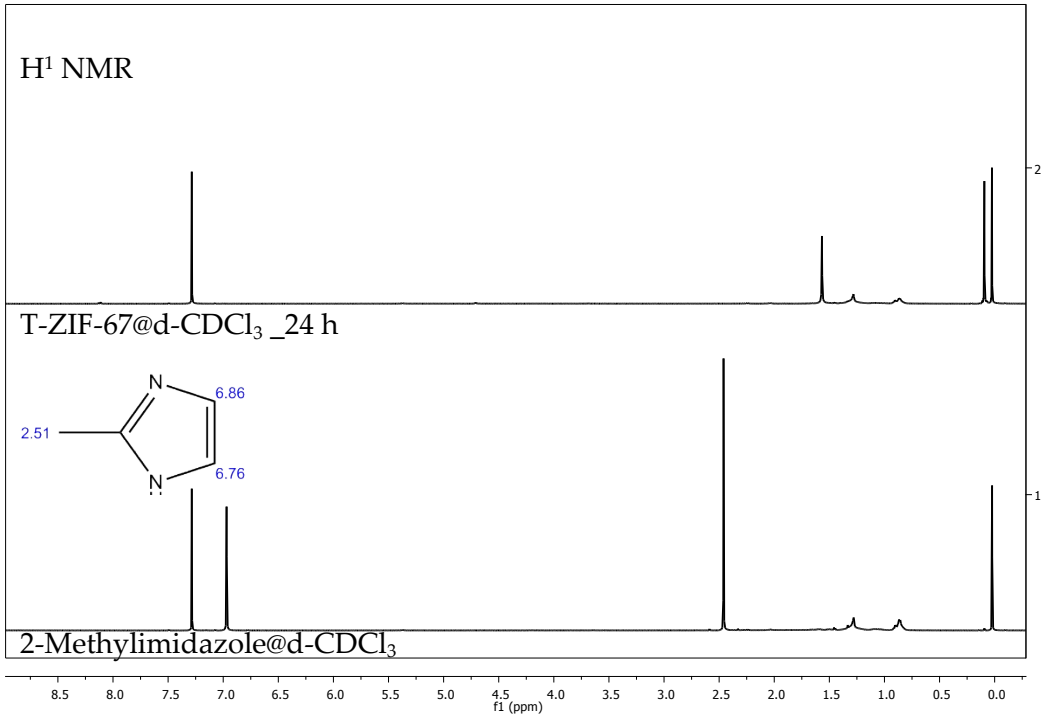


Figure S5. The H^1 NMR of soak solution T-ZIF-67 for 24 (top) and 2-methylimidazole (down) in d-$CDCl_3$ solvent.

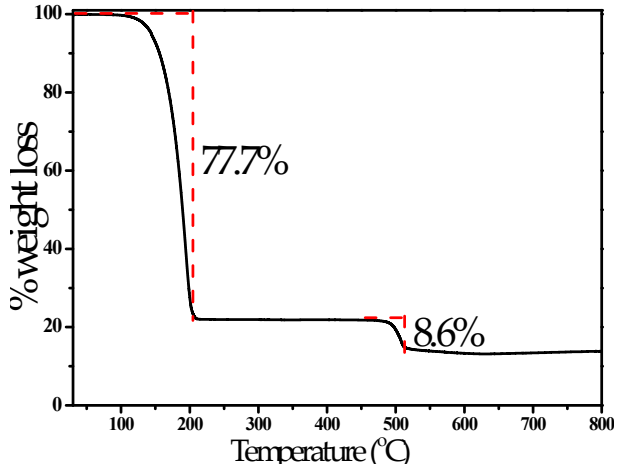


Figure S6. The thermal gravimetric analysis of feed solid-mixture precursor (CoO:2-MIM, 1:4)

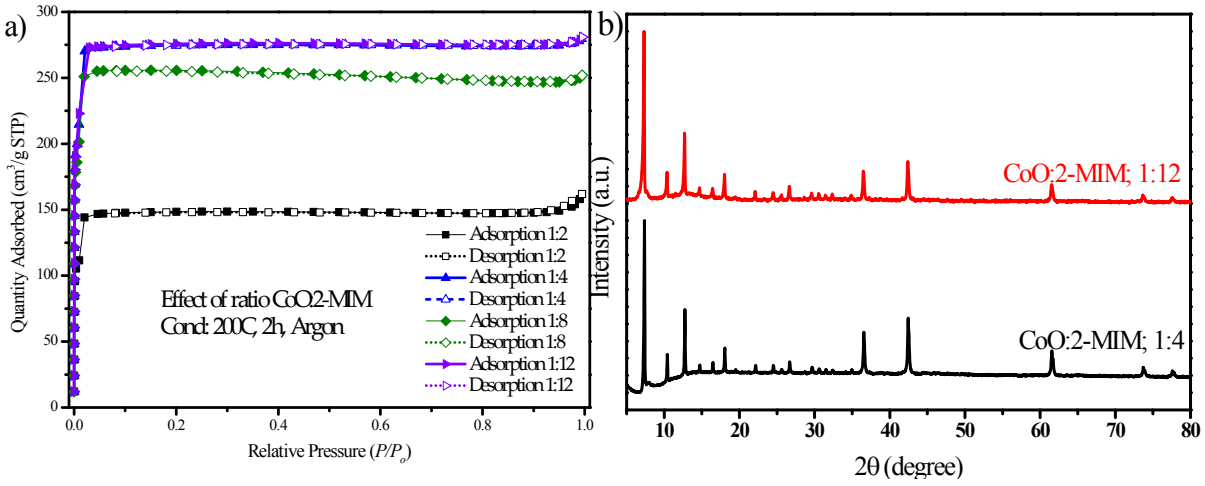


Figure S7. a) The isotherm of nitrogen adsorption at 77K and b) the XRD pattern on synthesized material with variance ratio of CoO:2-MIM. Synthesis condition at 200°C for 2h under argon flow ($50 cm^3 \cdot g^{-1}$).

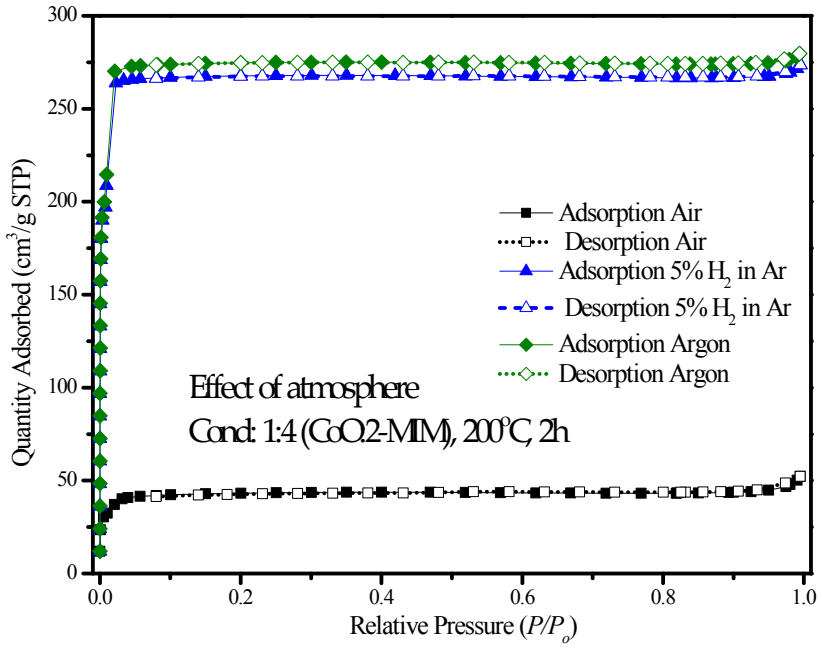


Figure S8. The isotherm of nitrogen adsorption at 77K on synthesized material with variance atmosphere. Synthesis condition at CoO:2-MIM (1:4), 200°C for 2h.

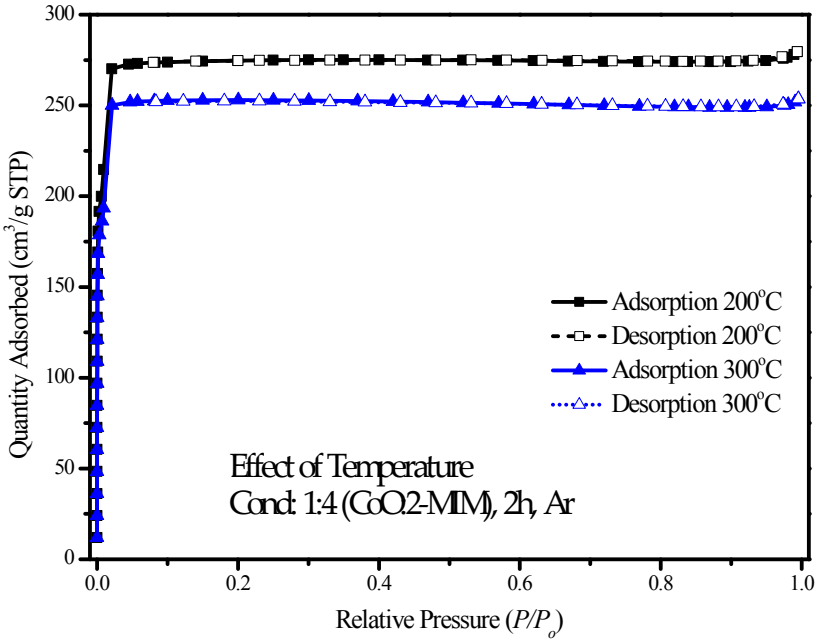


Figure S9. The isotherm of nitrogen adsorption at 77K on synthesized material with variance temperature treatment. Synthesis condition at CoO:2-MIM (1:4) for 2h under argon flow ($50 cm^3 \cdot g^{-1}$).

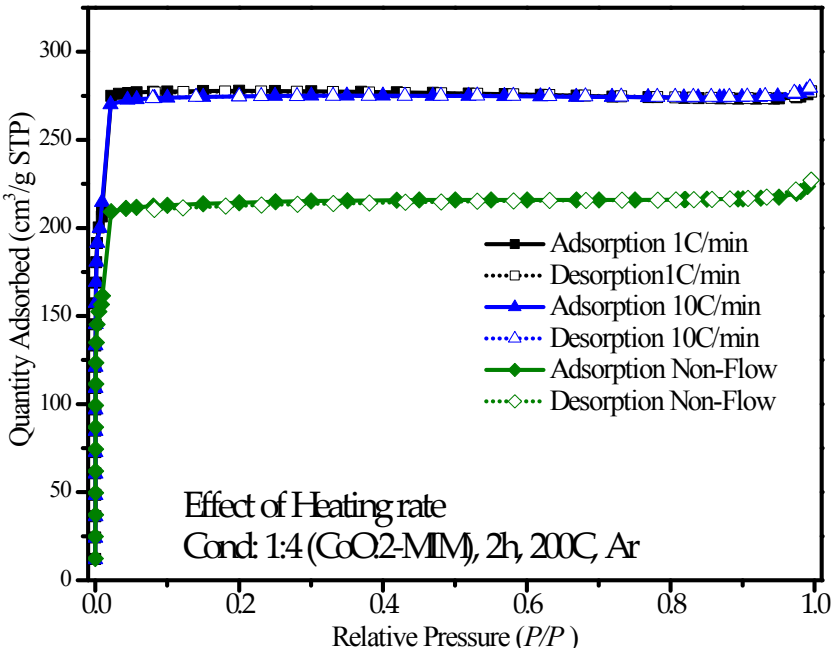


Figure S10. The isotherm of nitrogen adsorption at 77K on synthesized material with variance heating rate. Synthesis condition at CoO:2-MIM (1:4), 200°C, 2h, under argon flow ($50 cm^3 \cdot g^{-1}$). Non-flow: the stagnant under Ar atmosphere (no flow during thermal treatment), at heating rate 10°C/min.

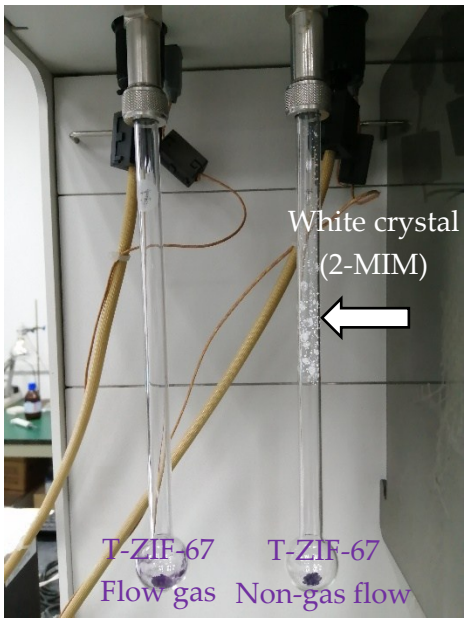


Figure S11. The crystal observation during degasing sample before porosity analysis.

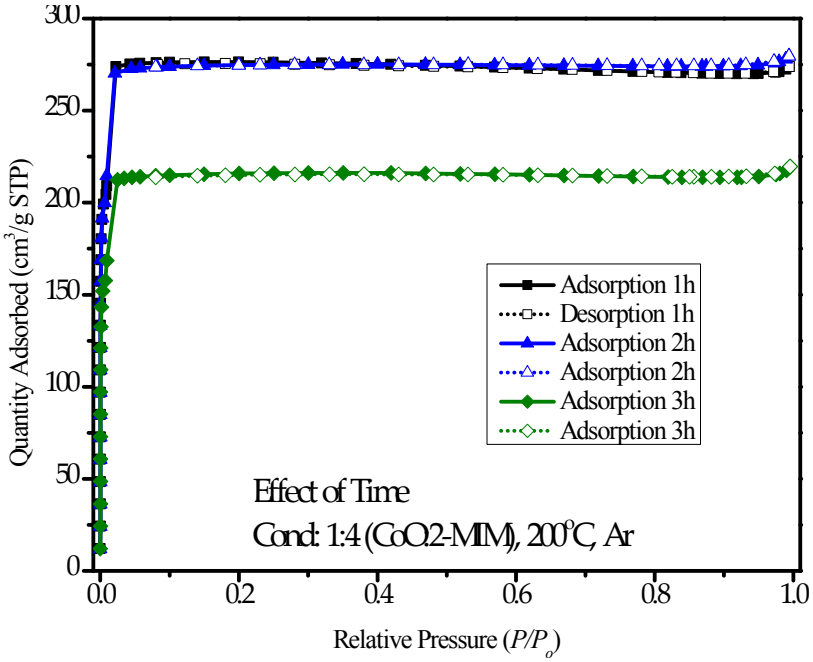


Figure S12. The isotherm of nitrogen adsorption at 77K on synthesized material with variance treatment time. Synthesis condition at CoO:2-MIM (1:4), 200°C under argon flow (50cm^3·g^{-1}).

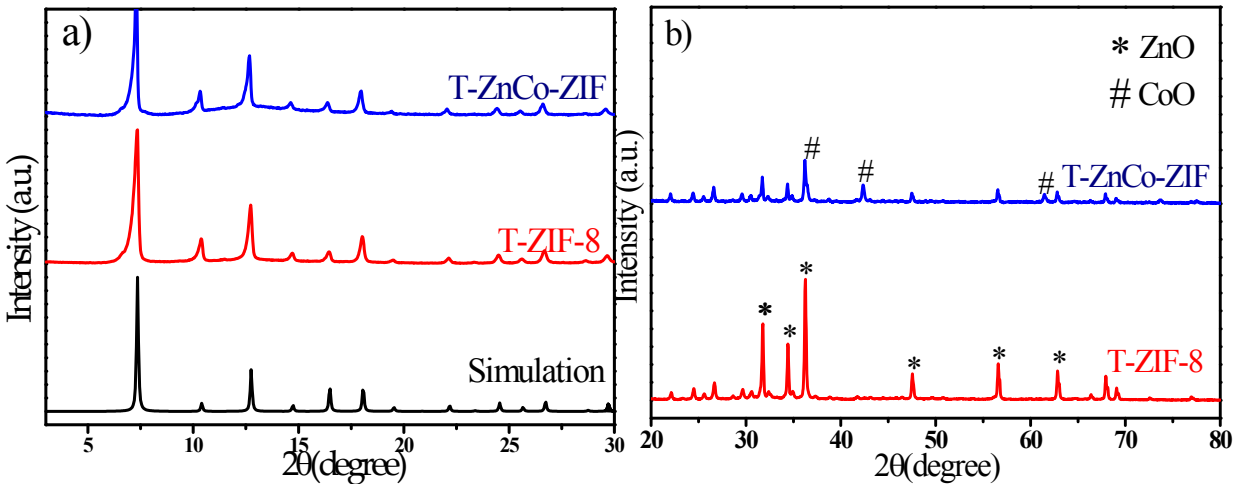


Figure S13. The XRD patterns of T-ZIFs in low angle (a) and high angle (b).

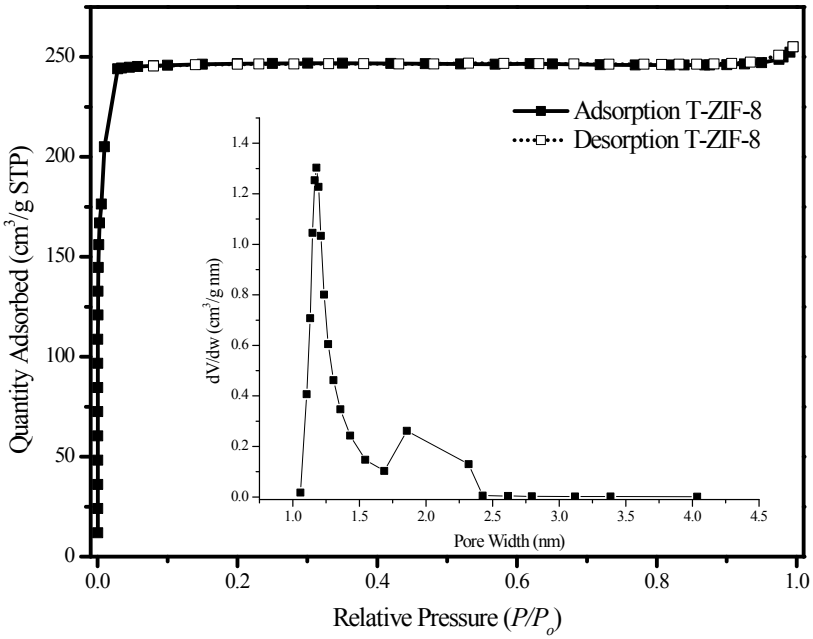


Figure S14. The isotherm of N_2 adsorption at 77K of T-ZIF-8 and pore size distribution (insert figure).

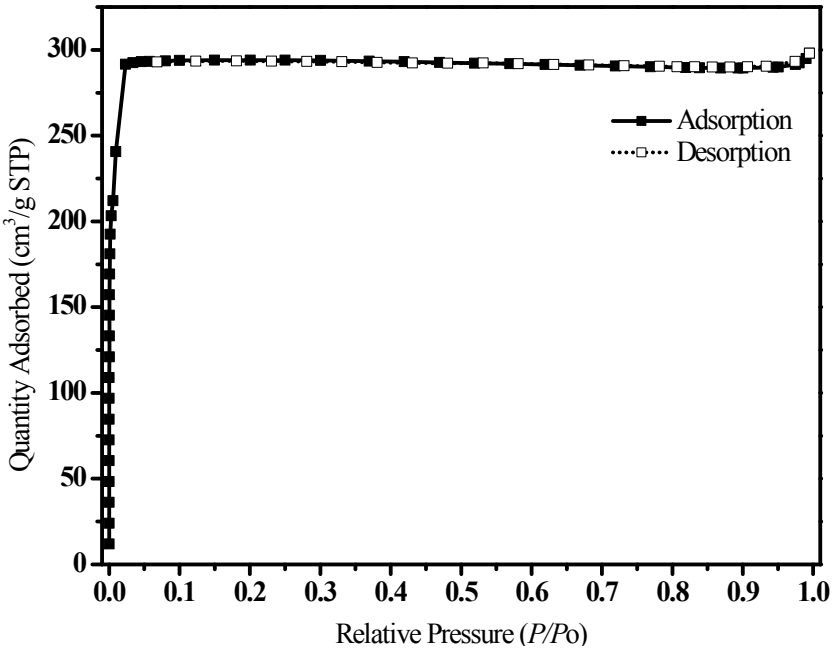


Figure S15. The isotherm of N_2 adsorption at 77K of bimetallic T-Zn/Co-ZIF-8 and pore size distribution (insert figure).