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Two-dimensional Conjugated Metal-orgnaic Framework Electrocatalyst

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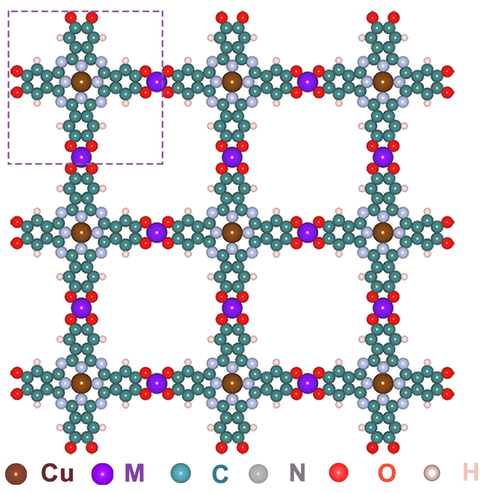
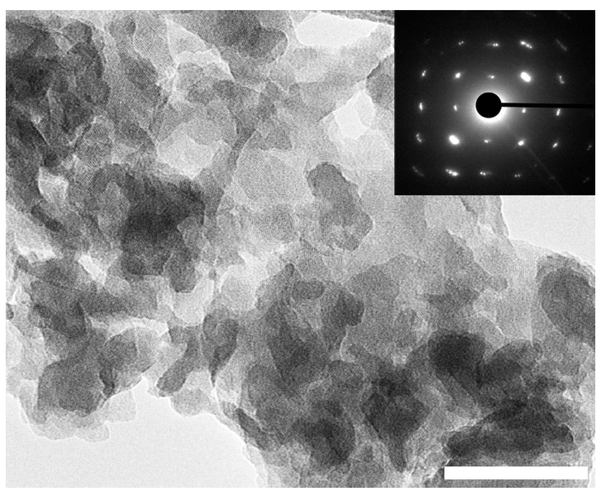
Two-dimensional conjugated metal-organic framework (2D *c*-MOF), with highly in-plane *π*-conjugation and weak out-plane *π*-*π* stacking, has emerged as one novel class of promising electrocatalysts due to the intrinsic electrical conductivity, high surface area, dense active sites and structural diversity. Herein, we developed a copper-phthalocyanine-based 2D *c*-MOF (PcCu-O_8-Co/Zn) with square planar CoO_4/ZnO_4 complexes as linkages toward electrocatalysis oxygen/carbon dioxide reduction reaction (ORR/CO_2RR). PcCu-O_8-Co mixed with carbon nanotubes exhibits excellent electrocatalytic ORR activity ($E_{1/2}$=0.83 V vs. RHE and j_L=5.3 mA cm^{-2}) in alkaline media owing to the synergistical contribution of 2D conjugated porous structure and dense CoO_4 sites with unique electric structure. The PcCu-O_8-Zn with carbon nanotube harvests high CO_2RR performance with CO selectivity of 88% and tunable molar H_2/CO ratio (1:7~ 4:1) toward syngas synthesis. The contrast results unveil a synergistic catalytic mechanism; ZnO_4 complexes act as catalytic sites for CO_2 conversion while CuN_4 centers promote the protonation of adsorbed CO_2 during the CO_2RR. Our works highlight the 2D conjugated MOFs with optimized composition/architecture and electronic structure as effective electrocatalysts toward ORR and CO_2RR

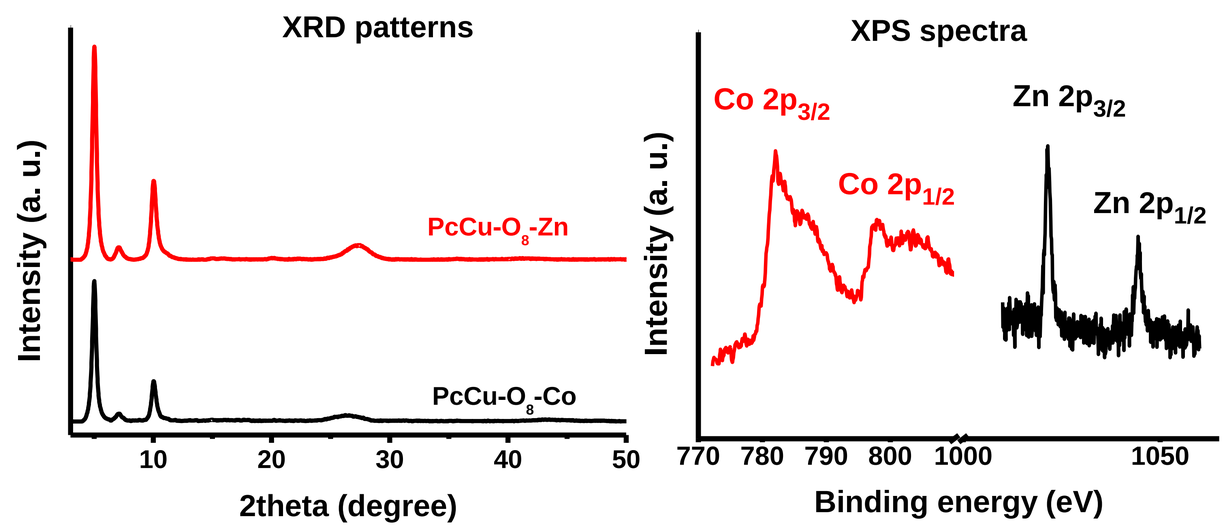
Morphology and Structure of PcCu-O_8-Co/Zn



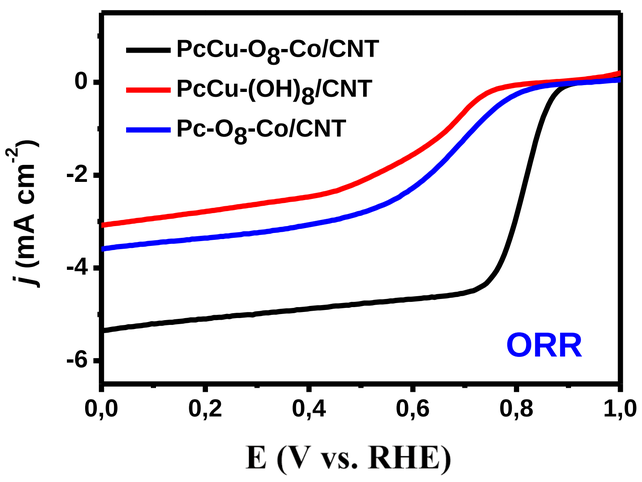
Schematic structure



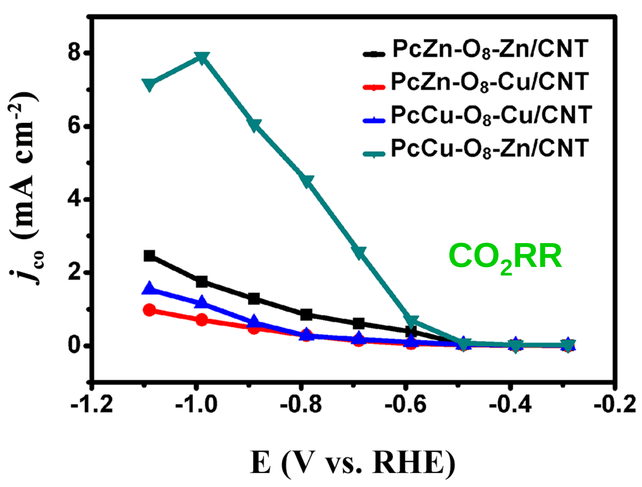
TEM image



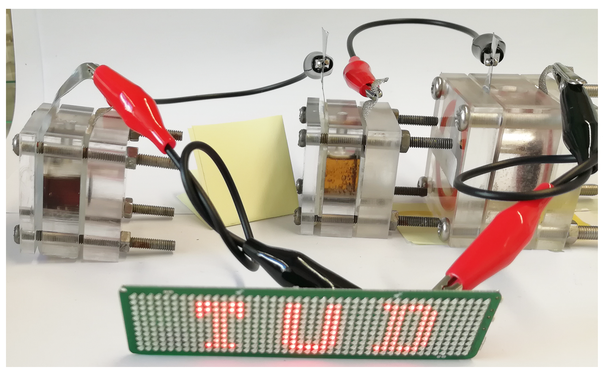
ORR and CO_2RR Performance of PcCu-O_8-Co/Zn



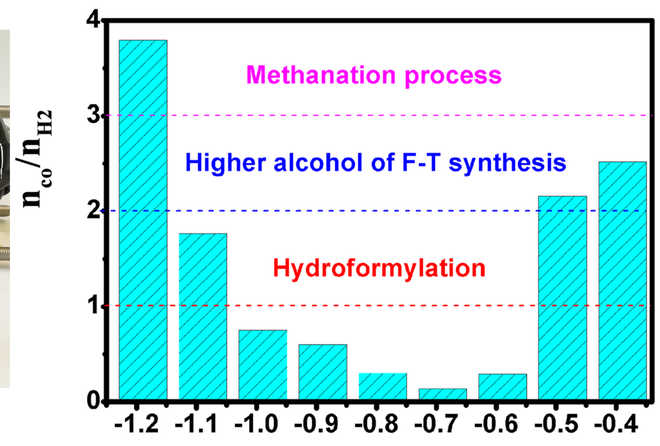
ORR polarization curves



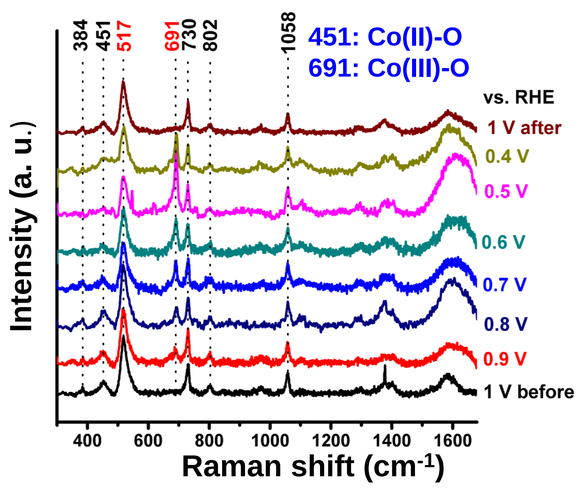
CO current density



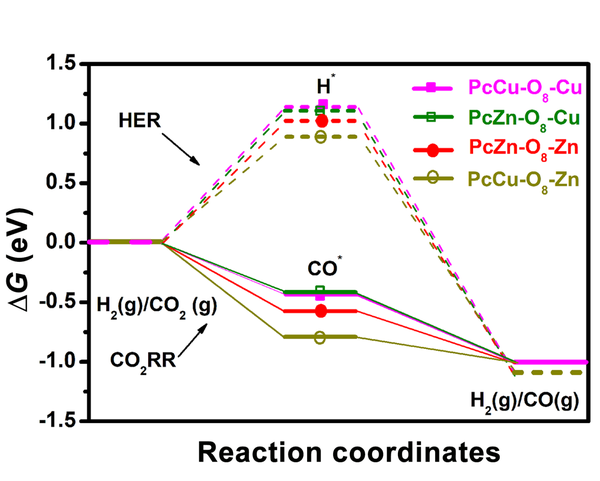
Zn-air battery



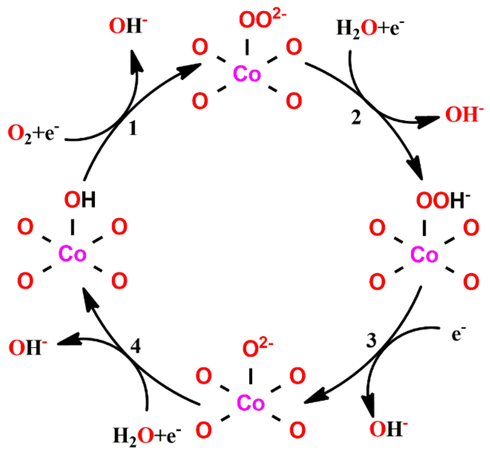
Catalytic Reaction Mechanism



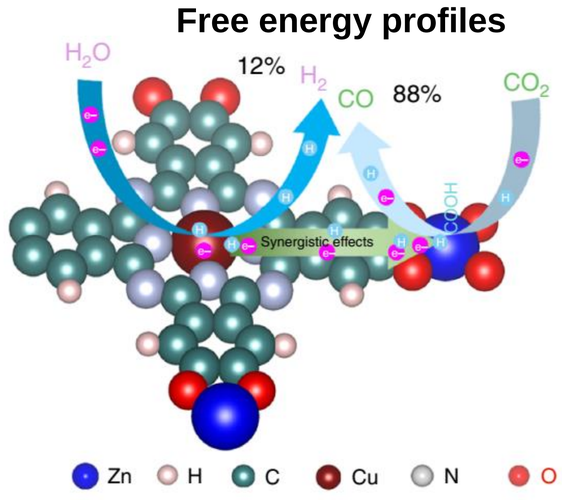
In-situ Raman spectra



Free energy profiles



ORR mechanism



CO_2RR process

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REFERENCES
[1] H. Zhong, K. H. Ly, M. Wang, Y. Krupskaya, X. Han, J. Zhang, J. Zhang, V. Kataev, B. Büchner, I. M. Weidinger, S. Kaskel, P. Liu, M. Chen, R. Dong, X. Feng, Angew. Chem. Int. Ed. 31 (2019) 10787.
[2] H. Zhong, M. Ghorbani-Asl, K. H. Ly, J. Zhang, J. Ge, M. Wang, Z. Liao, D. Makarov, E. Zschech, E. Brunner, I. M. Weidinger, J. Zhang, A. V. Krasheninnikov, S. Kaskel, R. Dong, X. Feng, Nat. Commun. 1(2020) 1409.

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