chem2Dmat
August 31 - September 03, 2021 ● Bologna, Italy
European Conference on Chemistry of Two-Dimensional Materials

Two-Dimensional Conjugated Framework Electrocatalyst

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Two-dimensional conjugated metal-organic frameworks (2D *c*-MOFs), with highly in-plane π-conjugation and weak out-plane π-π stacking, have emerged as novel generation of promising electrocatalysts, due to the intrinsic electrical conductivity, permanent pores, high surface area, and structural diversity. Herein, we developed a copper-phthalocyanine-based 2D *c*-MOF (PcCu-O_8-Co/PcCu-O_8-Zn) with square planar cobalt/zinc-bis(dihydroxy) complexes as linkage toward electrocatalysis oxygen/carbon reduction reaction (ORR/CO_2RR). PcCu-O_8-Co 2D *c*-MOF 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, which is the record value among the reported intrinsic MOF electrocatalysts. The PcCu-O_8-Zn with carbon nanotube harvests high CO_2RR performance with high CO selectivity of 88%, long-term durability, and tunable molar H_2/CO ratio (1:7~ 4:1) toward syngas synthesis. The contrast results unveil a synergistic catalytic mechanism; the ZnO_4 complexes act as catalytic sites for CO_2 conversion while the CuN_4 centers promote the protonation of adsorbed CO_2 during the CO_2RR. Our works highlight the 2D conjugated MOFs with optimized the composition/architecture and electronic structure as effective electrocatalysts toward ORR and CO_2RR.

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.

Figures

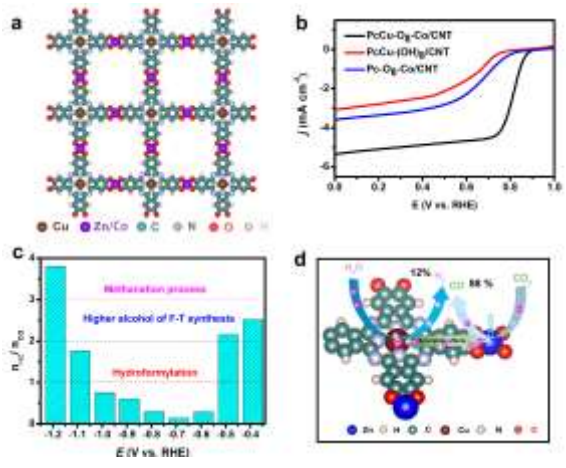

Figure 1: a. Schematic structure of 2D *c*-MOF. b. ORR polarization curves of 2D *c*-MOF/CNT. c. Molar H_2/CO ratio of 2D *c*-MOF/CNT from CO_2RR. d. Proposed CO_2RR process of 2D *c*-MOF.