Conversion of Carbon Dioxide to Versatile Green Chemicals With Isoreticular Metal-Organic Frameworks

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Recent advances in CO2 capture and fixation allowed the transformation of atmospheric CO2 into useful organic molecules, but they are limited by extreme reaction conditions or the use of dangerous reagents like phosgene due to the absence of optimal catalysts. Herein, we synthesized two series (zinc and zirconium) of isoreticular metal-organic frameworks (IRMOFs), functionalized porous materials with abundant Lewis-acid sites, in varying pore sizes, and nitrogen sites as heterogeneous catalysts for the cycloaddition of CO2 and epoxides to cyclic carbonates under mild conditions. Notably, the epoxide conversion rate and selectivity of Zr-IRMOFs, with high structural stability and retention property, far exceeded those of Zn-IRMOFs. In addition, comparing the BPDC and BDC ligands demonstrates that the optimal pore size for catalysis was 12-16 Å. Besides, nitrogen-functionalized ligands (BPYDC, BPDC-NH2) played a crucial role in providing additional binding sites for CO2 adsorption and abundant Lewis base sites for CO2 activation. We proposed a catalytic mechanism based on a literature kinetic study, reaction mixture analysis, and density functional theory calculations. It is notable that the Zr-BPYDC catalyst demonstrated a >99% conversion rate with >99% selectivity with high retention at mild conditions (1atm, 80°C, 0.5 mol% loading). Our research adds important insight to the systematic understanding of different physical and chemical properties of MOFs that influence CO2 conversion.

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