Title:	Effect of Unsaturated Metal Site Modulation in Highly Stable Microporous Materials on CO2 Capture and Fixation
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Abstract:	We have designed and synthesized two unprecedented microporous three-dimensional metal–organic frameworks, {[Cd6(TPOM)3(L)6]·12DMF·3H2O}n (1) and {[Zn2(TPOM)(L)2]·2DMF·H2O}n (2), based on a flexible quadritopic ligand, tetrakis(4-pyridyloxymethylene)methane (TPOM), and a bent dicarboxylic acid, 4,4′-(dimethylsilanediyl)bis-benzoic acid (H2L). The networks of 1 and 2 share a 4-c uninodal net NbO topology but exhibit different metal environments due to coordination preferences of Cd(II) and Zn(II). The Cd(II) center in 1 is six-coordinated, whereas the Zn(II) center in 2 is only four-coordinated, making the latter an unsaturated metal center. Such modulation of coordination atmosphere of metal centers in MOFs with the same topology is possible due to diverse binding of the carboxylate groups of L2–. Both 1 and 2 have relatively high thermal stability and exhibit permanent porosity after the removal of guest solvent molecules based on variable temperature powder X-ray diffraction and gas adsorption analysis. These materials exhibit similar gas adsorption properties, especially highly selective CO2 uptake/capture over other gases (N2 and CH4). However, because of the presence of an unsaturated Lewis acidic metal site, 2 acts as a very efficient heterogeneous catalyst toward the chemical conversion of CO2 to cyclic carbonates under mild conditions, whereas 1 shows very less activity. This work provides experimental evidence for the postulate that an unsaturated metal site in MOFs enhances adsoprtion of CO2 and promotes its conversion via the Lewis-acid catalysis.
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