Title:	Engineering a Nanoscale Primary Amide-Functionalized 2D Coordination Polymer as an Efficient and Recyclable Heterogeneous Catalyst for the Knoevenagel Condensation Reaction
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Abstract:	This work reports the design, structural characterization, and catalytic behavior of the first example of primary amide-functionalized coordination polymers (CPs), namely, {[Cd2(2-bpbg)(fum)2(H2O)2]·8.5H2O}n (1) (where 2-bpbg = N,N′-bis(2-pyridylmethyl)-1,4-diaminobutane-N,N′-diacetamide and fum = fumarate). CP 1 is synthesized from a one-pot self-assembly of starting materials in methanol under ambient conditions in excellent yield and purity, allowing an easy access to multigram quantities of it within few hours. As an example, CP 1 was used as a highly efficient heterogeneous catalyst in the carbon–carbon bond-forming Knoevenagel condensation reaction for the conversion of benzaldehyde to benzylidene malononitrile. CP 1 possesses both Lewis acidic and Brønsted basic character for the presence of unsaturated metal sites and primary amide groups, respectively, making it a highly competent bifunctional catalyst for such type of reactions. Surprisingly, on the one hand, 100% conversion was observed using only 2 mol % catalyst within 1 h at 27 °C in methanol. On the other hand, 2 mol % and 3 mol % catalyst loadings but without a solvent gives 93% and 100% conversions, respectively, in 1 h at 27 °C. CP 1 is far better than those reported in the literature. To prove the uniqueness and efficiency of the primary amide-based ligand, a similar compound with a pyridyl-based ligand was also synthesized, {[Cd2(tpbn)(fum)2]·6H2O}n (2) (where tpbn = N′,N′,N″,N″-tetrakis(pyridin-2-ylmethyl)butane-1,4-diamine). With CP 2 under the same catalyst loading and conditions (2 mol %, 27 °C, 1 h), only 28% conversion was observed. This demonstrates that selective heterogeneous catalytic properties of 1 over 2 are due to the presence of the primary amide moieties and open metal sites. Moreover, CP 1 can easily be separated from the reaction mixture and reused for five consecutive cycles without significant loss of its activity. Both 1 and 2 were fully characterized by elemental analysis, infrared spectroscopy, thermogravimetric analysis, and single-crystal and powder X-ray diffraction. These crystallize in the triclinic $P\bar{1}$ space group, showing their isostructural nature and three-connected, uninodal {63} honeycomb net topology.
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