

Copper(II) coordination polymers self-assembled from aminoalcohols and pyromellitic acid: highly active precatalysts for the mild water-promoted oxidation of alkanes.

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Three novel water-soluble 2D copper(II) coordination polymers— $[\{\text{Cu}_2(\mu_2\text{-dmea})_2(\text{H}_2\text{O})\}_2(\mu_4\text{-pma})]_n \cdot 4n\text{H}_2\text{O}$ (**1**), $[\{\text{Cu}_2(\mu_2\text{-Hedea})_2\}_2(\mu_4\text{-pma})]_n \cdot 4n\text{H}_2\text{O}$ (**2**), and $[\{\text{Cu}(\text{bea})(\text{Hbea})\}_4(\mu_4\text{-pma})]_n \cdot 2n\text{H}_2\text{O}$ (**3**)—were generated by an aqueous medium self-assembly method from copper(II) nitrate, pyromellitic acid (H_4pma), and different aminoalcohols [*N,N*-dimethylethanolamine (Hdmea), *N*-ethyldiethanolamine (H_2edea), and *N*-benzylethanolamine (Hbea)]. Compounds **2** and **3** represent the first coordination polymers derived from H_2edea and Hbea . All the products were characterized by infrared (IR), electron paramagnetic resonance (EPR), and ultraviolet–visible light (UV-vis) spectroscopy, electrospray ionization–mass spectroscopy (ESI-MS($\pm$)), thermogravimetric and elemental analysis, and single-crystal X-ray diffraction (XRD), which revealed that their two-dimensional (2D) metal–organic networks are composed of distinct dicopper(II) or monocopper(II) aminoalcoholate units and μ_4 -pyromellitate spacers. From the topological viewpoint, the underlying 2D nets of **1–3** can be classified as uninodal 4-connected layers with the **sql** topology. The structures of **1** and **2** are further extended by multiple intermolecular hydrogen bonds, resulting in three-dimensional (3D) hydrogen-bonded networks with rare or unique topologies. The obtained compounds also act as highly efficient precatalysts for the mild homogeneous oxidation, by aqueous H_2O_2 in acidic $\text{MeCN}/\text{H}_2\text{O}$ medium, of various cycloalkanes to the corresponding alcohols and ketones. Overall product yields up to 45% (based on cycloalkane) were attained and the effects of various reaction parameters were investigated, including the type of precatalyst and acid promoter, influence of water, and substrate scope. Although water usually strongly inhibits the alkane oxidations, a very pronounced promoting behavior of H_2O was detected when using the precatalyst **1**, resulting in a 15-fold growth of an initial reaction rate in the cyclohexane oxidation on increasing the amount of H_2O from ~ 4 M to 17 M in the reaction mixture, followed by a 2-fold product yield growth.

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