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The advancement of polyoxometalate (POM) science requires rational routes to targeted structures and compositions for systematic studies of reactivity and properties, and our new synthetic methodologies and associated ^{17}O -enrichment techniques are enabling detailed solution studies of reactive POMs. [1, 2] The resulting access to homologous series of POMs is revealing the consequences of atom substitution within structural frameworks and provides the basis for theoretical DFT studies and a deeper understanding of subtle electronic effects and bonding variations. [3] A detailed understanding of proton and electron transfer processes is emerging from studies of protolytic reactions in POM-catalysed reactions, *e.g.* alkene epoxidation with H_2O_2 , [4] and investigations of non-aqueous reduction are providing electron-rich and super-reduced POMs. [5] We are also exploring the self-assembly and templating properties of amphiphilic POMs, multifunctional catalysts that exploit the synergy created by incorporation of POMs at interfaces, and the design and synthesis of highly active POMs for small molecule activation. [6]

References.

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