Modeling copper-containing monooxygenases active sites for dioxygen or water activation

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Copper-containing enzymes have recently gained attention due to the discovery of new oxygenases with high potential for the production of fuels: lytic polysaccharide monooxygenases (LPMO) and particulate methane monooxygenase (pMMO).[1,2] LPMOs boost recalcitrant polysaccharides breakdown via oxidative cleavage and pMMO is a membrane-bound protein that oxidizes methane into methanol. Both enzymes are able to perform hydroxylation of non-activated and highly energetic C-H bonds and display a common and unusual bidentate binding mode of the N-terminal histidine on the copper ion (Figure 1).

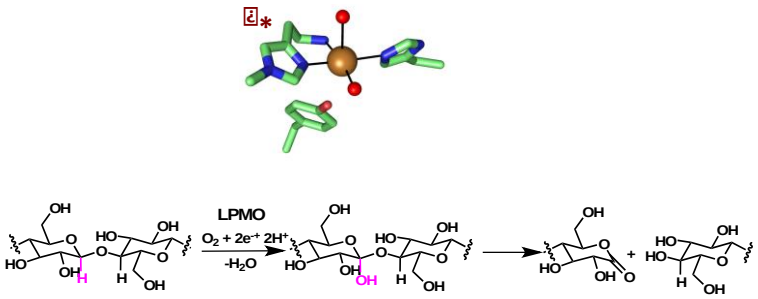

Figure 1 Active site structure (histidine-brace motif highlighted) and reaction catalyzed by LPMO

The preparation of relevant low molecular-weight models of metalloenzymes provides very important tools for mechanistic studies and for the development of metal-containing catalysts. Our group is interested in the study of metalloenzymes (as LPMO) associated with the development of bioinspired copper-containing complexes.[3] Based on knowledge acquired on dioxygen activation, copper complexes to achieve water activation for O-atom transfer reaction are also investigated.[4]

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