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Transition metal complexes of pyridine-containing macrocycles as catalysts for selective oxidations and CO₂ valorisation reactions

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Iron, the most abundant transition metal on earth, and its complexes are knowing an increasing interest in organic synthesis. The field of iron-catalysed oxidation reactions is of great importance not only in synthetic organic chemistry, but also in biochemistry and industrial applications. In order to design catalysts capable of performing high regio- and/or stereo-selective C-OH, C-H or C=C bond oxidations, it is important the choice of critical components of iron coordination sphere, namely the donor atoms and their geometry. In this lecture, I will provide a perspective on the catalytic applications of iron(III) and zinc(II) metal complexes of tetraaza 12-membered pyridine containing macrocyclic ligands. I will focus on the selective iron(III) catalysed epoxidation or dihydroxylation of alkenes by using hydrogen peroxide as terminal oxidant.² Depending on the anion of the iron(III) metal complex employed as catalyst, a completely reversed selectivity was observed (Figure). Our approach towards the selective oxidation of alcohols by using the same catalytic system will also be covered. As for iron, catalytic applications of zinc complexes fall in the scientific community's effort to develop more eco-friendly chemical processes. CO2 is the principal greenhouse gas, largely recognized as responsible for global warming, but it is also an abundant C1 source. Limiting CO2 emissions can only stem the problem but to solve it a circular economy based on carbon dioxide should be pursued and in this respect, research in the last decade has focussed on the design of systems able to promote the functionalisation of CO₂. I will outline our approach towards the synthesis of cyclic carbonates by cycloaddition of CO2 to epoxides by using zinc(II) complexes.

Fig. Controllable iron(III)-catalysed alkene epoxidation or dihydroxylation reactions are performed by judicious choice of anion in [Fe(III)(Pc-L)] catalysts (Pc-L = pyridine-based 12-membered tetraaza-macrocyclic ligands).

Key words: macrocyclic ligands, homogeneous catalysis, iron, zinc, oxidation, CO_2 functionalization.

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