

UniCat Colloquium

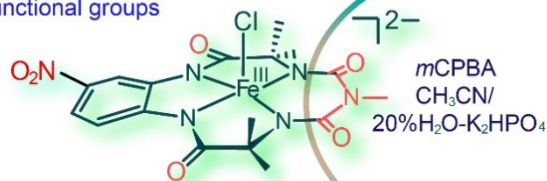
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Developing Fe-complexes as Catalysts for Biomimetic Oxidation of Unactivated Alkyl C-H bonds

Chemoselective oxidation of alkyl C–H bonds in complex natural products under mild conditions have the power to reorient biomimetic natural product synthesis. Non-heme iron complexes bearing tetradentate N-donor ligands with cis labile sites, mimicking the active site of Rieske Dioxygenase enzymes, show great promise for chemoselective aliphatic C–H hydroxylation. However formidable hurdle that limits their widespread application include obtaining high levels of positional selectivity and expanding the substrate scope to include complex structures that contain more than a single polar functional group and/or arene/heteroaromatic ring(s). In this lecture, the mechanism-guided development of a peroxidase mimicking Fe-complex based on the bTAML macrocyclic ligand framework (Fe-bTAML), that perform selective oxidation of unactivated 3° bonds with unprecedented regioselectivity (3°:2° of 110:1 for adamantane oxidation), high stereoretention (99%), and high turnover numbers (TONs up to 300(using mCPBA/NaOCl as the oxidant, will be discussed. Mechanistic studies, which include correlation of product formed with FeV(O) reactive intermediates generated during the reaction, indicate that the major pathway involves the cleavage of C-H bonds by FeV(O). This methodology was then used to perform selective oxidation of several substrates including natural products with predictable selectivity, high stereoretention (>98%), and very low catalyst loadings (1-2 mol %). Finally, I will also discuss our recent efforts on using the Fe-bTAML complex to selectively catalyse the photocatalytic hydroxylation and epoxidation reactions of alkanes and alkenes respectively using water as the oxygen-atom source

- Unprecedented regioselectivity for 3° over 2° C-H bond R-H
- 18 substrates including arenes, N-heterocycles and polar functional groups



- Selective oxidation of complex natural products
- Low catalyst loading (1-2 mol %)

Wednesday, June 20, 2018 at 5:15 PM

TU Berlin, Institute of Chemistry
Straße des 17. Juni 115, 10623 Berlin

Building C, Lecture Hall C 264

Prof. Dr. Driess (TUB)

Organizer

Coffee and cake will be served 30 minutes before the lecture. Guests are cordially invited to attend!
Prof. Dr. Matthias Driess - Chair of the Cluster of Excellence UniCat - www.unicat.tu-berlin.de