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Prof. Dr. Felix Tuczek

Institut für Anorganische Chemie, Christian-Albrechts-Universität zu Kiel

Doing Chemistry with Air: Activation of N_2 and O_2 in Biological Systems

Dinitrogen (N_2) and dioxygen (O_2) are the main constituents of the earth´s atmosphere. The bonding and activation of these molecules as well as their metabolic conversions are fundamental to all forms of life. Many biologically important oxidation and oxygenation reactions are mediated by iron and copper enzymes. Prominent examples of the latter are particulate methane monooxygenase (pMMO) and tyrosinase (TY). Whereas pMMO mediates the conversion of methane to methanol, TY catalyzes the hydroxylation and two-electron oxidation of L-tyrosine to L-DOPAquinone, which constitutes the first step of melanine biosynthesis. In the talk, small-molecule models of pMMO and TY are presented, and their electronic structures and reactivities are described.[1,2]

Nitrogen fixation involves the bonding of N_2 at the iron-molybdenum cofactor of the enzyme nitrogenase and its conversion into a bioavailable form; i.e., ammonia. In search of a single-site molybdenum phosphine complex catalyzing dinitrogen protonation and reduction in analogy to the biological process we have developed the pentadentate tetrapodal phosphine (pentaPod) ligand $P5^{Me}$.[3] The derived molybdenum dinitrogen complex $[Mo(N_2)(P5^{Me})]$ generates 26 equivalents of ammonia from N_2, employing SmI_2/H_2O as PCET reagent. Recently we replaced the dimethylphosphine groups of $P5^{Me}$ by phospholanes, leading to the ligand $P5^{Pln}$. The derived tungsten complex $[W(N_2)(P5^{Pln})]$ is the first W complex chemocatalyzing the conversion of N_2 to NH_3.[4] The implications of the results on the biomimetic reduction of N_2 are discussed.

[1] R. Jurgeleit, B. Grimm-Lebsanft, B. M. Flöser, M. Teubner, S. Buchenau, L. Senft, J. Hoffmann, M. Naumova, C. Näther, I. Ivanović-Burmazović, M. Rübhausen, F. Tuczek *Angew. Chem. Int. Ed.* **2021**, *60*, 14154-14162.
[2] R. Schneider, T. A. Engesser, C. Näther, I. Krossing, F. Tuczek, *Angew. Chem. Int. Ed*. **2022**, *61*, e202202562
[3] T. A. Engesser, A. Kindjajev, J. Junge, J. Krahmer, F. Tuczek, *Chem. Eur. J.* **2020**, *26*, 14807
[4] A.-M. Vogt, T. A. Engesser, J. Krahmer, N. Michaelis, M. Pfeil, J. Junge, C. Näther, N. Le Poul, F.Tuczek, *Angew. Chem. Int. Ed.* **2025**, *64*, e202420220

Donnerstag, **3. Juli 2025, 17.15 pm**
Hörsaal N05, Auf der Morgenstelle 16

gez. Ivana Fleischer