

UniCat Colloquium

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Photocatalytic H_2 formation with Cobalt Complexes of Poly-pyridine Ligands

Photocatalytic water splitting is an approach to support the world's future energy demand. The reductive half reaction of this process generates H2 and is the subject of extensive, ongoing studies. The water reduction catalysts (WRC) play thereby a central role. Their stabilities and efficiencies are of particular importance. The understanding of mechanisms and the preparation of new catalysts are crucial for progressing towards useful systems. We have synthesized a series of new Coll-based WRCs with tetra- and penta-pyridine type ligands. In the presence of sacrificial electron donors such as ascorbic acid and photosensitizers, these WRCs proved to be highly efficient and turnover numbers of up to 10,000 could be achieved.



Kinetic investigations with different methods provided an insight into rate constants and limiting steps. We found in particular, that the oxidized form of the sacrificial electron donor, dehydroascorbic acid, short cuts catalysis by eback transfer which limits performance. Adding a second, irreversible e- donor to the system enhances performance and allows catalysis to run to completion. In the presentation, syntheses and characterization of the new WRCs, mechanistic results from catalytic cycles and first attempts to switch from homo- to heterogeneous architectures will be presented.

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