From molecular to polymeric porphyrin-based material for the activation and reduction of CO2

Artificial photosynthesis aims at the production of solar fuels as a replacement of fossil energy carriers. For these processes catalysts are developed in our group for the indirect (electrolysis coupled to renewable electricity) or direct (photocatalytic) reduction of CO2 into energy-rich chemicals.

We have recently developed porphyrin-based catalysts with modified second coordination sphere for the efficient two-electron reduction of CO2 to CO. Lessons from these studies have comforted our strategy that manipulating the second coordination spheres may greatly help to enhance the catalytic performance of molecular catalysts. In this thesis project we will address the development of catalytic materials for reduction of CO2 to more reduced forms than CO eg. methanol or ethanol. We will develop a solid-state carbonaceous electrodes modified with polymers of metallo-porphyrins. Such polymers are already known for the excellent catalytic performance at the molecular level for the reduction of CO2 to methanol or ethanol. Their fixation at the surface of electrodes is expected to form stable and cost effective catalytic material for CO2 reduction. In a different strategy to elaborate new polymeric materials for the heterogenous electrocatalytic reduction of CO2, we will design new porphyrin dimers connected by a diacetylene function. Polymerization under irradiation of diacetylene function will be used to generate polydiacetylenes (PDAs), which are conjugated polymers of general formula (=CG-C=C-CG’=) where G and G’ stand for embarked functional groups which are designed to enhancing the catalytic activities. Different metal centers such Fe, Co, and Cu inside the porphyrin core will be tested and their respective electrocatalytic response investigated.

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