Seminar

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Molecular Catalysis of Electrochemical Reactions: Principles and Application to Mechanistic Studies of N_2O and CO_2 reduction

Date & Time: July 3rd (Wed), 17:15-18:30

Location: 本館 3 階, 第 2 会議室

<Abstract>

Molecular electrochemistry is a field studying molecular processes triggered by an electron transfer that is, most often, associated with the formation or breaking of bonds. In this presentation, we will discuss molecular catalysis of electrochemical reactions and recall the basic principles of such processes focusing on the use of cyclic voltammetry to decipher mechanisms and then illustrate these principles with examples.

Homogeneous electrochemical catalysis of CO_2 reduction to CO with iron tetraphenylporphyrins as catalysts will first be briefly described focusing on the role of proton donors as co-substrate as well as on modification of the catalyst structure. Then we will discuss the catalysis of N_2O to N_2 with a series of organic catalysts and transition metal complexes (rhenium bipyridyl carbonyl complexes) allowing to contrast outer- and inner-sphere mechanisms. In the latter case, the generation of a strong coordinating ligand as co-product of the reaction might be detrimental for an efficient catalysis because it can bind the metal center and block or slow down the catalytic process. This self-modulation phenomenon is revealed, formally described and illustrated. The mechanism of N_2O reduction with iron tetraphenylporphyrin as catalyst will also be presented and it will be shown that a switch from Fe(0) to Fe(I) as active form of the catalyst is observed and that a self-modulation process is also at play as opposed to the case of CO_2 reduction. Finally, the role of acids as co-substrate will be presented again contrasting the behavior of CO_2 and N_2O, two isoelectronic molecules.