Well-defined water oxidation and light-driven reduction catalysts

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Well-defined water oxidation and light-driven reduction catalysts

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One of the most appealing research areas is the mechanism understanding of multi-electron multi-proton processes, being central in the activation of small molecules such as the water oxidation to O_2 (WO) or reduction to H_2 (WR) processes.[1] In this line, we have discovered that readily available iron coordination complexes based on aminopyridine ligands are highly efficient homogeneous WO but also WR catalysts.[2-5] We present here one of the few examples of homogeneous and well-defined WO catalysts based on 1st row transition metals, which allows for a mechanistic studies. To gain insight into the mechanism of the Fe-catalyzed WO catalysis, we carried out a detailed study through kinetics, spectroscopic monitoring of intermediates, isotopic effects, isotopic labeling, electronic effects and DFT calculations.[3-5] The roles of the high oxidation state oxo-iron (IV) and (V) and new Fe-O-Ce species in the O-O forming event as well as possible intermediates in the oxidation of organic substrates[5] will be discussed.

Likewise, cobalt complexes based on aminopyridine ligands form robust homogeneous catalytic systems for light-driven reductions, in which their electronic and structural properties can be easily tuned.[6] By a combination of experimental and computational studies we have obtained key information about intermediate species and the mechanism for the photochemical reduction of water to H_2 but also ketones, aldehydes and olefins. The mechanism aspects will be discussed on this basis.

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Julio **Lloret-Fillol** graduated in Chemistry from the Universidad de Valencia in 2001 where he also obtained his PhD in 2006, working under the supervision of Prof. Lahuerta and Prof. J. Pérez-Prieto. After his PhD he moved to the University of Heidelberg where he stayed two years as a postdoctoral MEyC fellow and two years as a postdoctoral Marie Curie fellow. Since 2010 he has been working as independent research leader at Universitat de Girona (Ramón y Cajal programme). In 2014 he obtained a position as Young Research Group Leader at the Institut de Química Computational i Catàlisi (UdG). In November 2014 he started his independent research career at the Institute of Chemical Research of Catalonia (ICIQ).

At ICIQ, Dr. Lloret-Fillol develops a project aimed to merge concepts at the edge of the fields of solar fuels production and catalytic transformation facilitated by coordination complexes to perform new light-driven catalytic transformations of organic substrates. This project may open newer and greener avenues toward the application of artificial photosynthetic schemes in catalytic transformations of organic molecules of added value.