Studies for the development and understanding of heterogeneous photocatalysts

Prof. Dr. Jennifer Strunk

Leibniz Institute for Catalysis (LIKAT) at the University of Rostock, Germany

Photocatalytic processes that use only (sun)light as energy source are nowadays in development for a number of applications, ranging from water treatment[1] over selective oxidations to the generation of industrial resources from CO_2 and H_2O. In spite of decades of research, however, little systematic insight into reaction mechanisms or the nature of catalytic active sites has been obtained. This hinders reaction engineering and material science approaches to optimize the process and the photocatalysts, respectively. While such studies are already challenging in classical catalysis, an additional level of complexity is added in photocatalysis due to the dependence of reaction rates on the light intensity and the challenge to establish reproducible reaction conditions.[2] In this contribution, two successful examples for such systematic studies under controlled reaction conditions will be highlighted.

In studies of photocatalytic CO_2 reduction on TiO_2 under controlled reaction conditions CH_4, CO and H_2 are detected as main products. A variation of the reaction conditions in a continuous-flow reactor revealed that CH_4 yield is maximized under low CO_2 and H_2O concentrations below ~1vol%. Physisorbed H_2O is sufficient to carry out the reaction (Figure 1). The CH_4 yield shows a square-root dependence on the light intensity, indicating a major influence of charge carrier recombination. Elevated temperature (60°C) increases the CH_4 yield.[3,4]

Supported vanadium oxides with structures ranging from isolated sites to nanoparticles were used for the selective oxidation of CH_3OH to H_2CO. While all vanadium oxide species can carry out the reaction thermally catalyzed, only isolated sites and oligomers can carry out the photoreaction, i.e. nanoparticles are inactive (Fig. 2). Isolated vanadium oxide sites on silica display an exclusive H_2CO selectivity in the photoreaction at room temperature, but H_2CO desorption is rate-limiting. Changes in photoreactivity are attributed to the quantum size effect.[5]

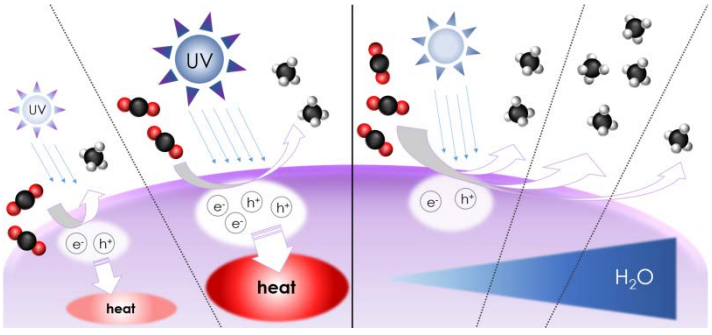

Figure 1: Influence of reaction conditions in CO_2 reduction.

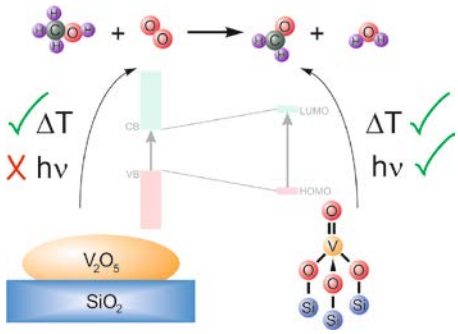

Figure 2: V/SiO_2 in CH_3OH oxidation to H_2CO.

References

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Curriculum Vitae - Prof. Jennifer Strunk

Academic Education

2008	PhD in Industrial Chemistry, Ruhr University Bochum (with M. Muhler)
2004	Diploma in Chemistry, Major: Industrial Chemistry, Ruhr-University Bochum, Germany

Profession

since 2017	Head of the department Photocatalysis, Leibniz Institut für Katalyse e.V., Rostock, Germany
2014 - 2016	Research Group Leader at the MPI for Chemical Energy Conversion, Mülheim/Ruhr, Germany
2010 - 2014	BMBF Junior Research Group Leader, Industrial Chemistry, Ruhr-University Bochum
2008 - 2010	Postdoctoral research at University of California, Berkeley / Lawrence Berkeley National Laboratory, CA, USA (with A.T. Bell)

Awards and other activities

2015	Two months research stay on “In situ Raman Spectroscopy” with Israel E. Wachs, Lehigh University, Bethlehem, PA, USA
2014	Jochen-Block-Preis of the German Catalysis Society e.V. (GeCatS)
2013	Selected as participant for the 63rd Lindau Nobel Laureate Meeting
since 2013	Member of the “Deutsche Bunsengesellschaft”
2011 - 2012	Awarded to be Member of the “Global Young Faculty”, Mercator Research Center Ruhr
since 2011	Member of GeCatS, DECHEMA, GDCh
since 2008	Member of the American Chemical Society (ACS)
2006 - 2007	PhD fellowship from the Heinrich-Böll-Stiftung