UPPSALA UNIVERSITET

Operando and Theoretical Surface Science Studies of Heterogeneous Catalysts

Thursday 25 March 10.15—12.00

Join Open Zoom Symposium at Uppsala University:
https://uu-se.zoom.us/j/62301982844

Symposium in connection to David Langhammer's thesis 26 March

Chair: Prof. Lars Österlund, Div. Solid State Physics, Dept. Materials Science and Engineering: lars.osterlund@angstrom.uu.se

10.15 – 11.00 Prof. Günther Rupprechter, Institute of Materials Chemistry, TU Wien, Austria:
"Operando Surface Spectroscopy and Microscopy During Catalytic Reactions"

Abstract: *Operando* characterization of working catalysts, requiring the simultaneous measurement of catalytic performance, is crucial to identify the relevant catalyst structure/composition and how molecules interact with surfaces/interfaces. Two examples, covering nanoparticles, thin films and meso-scale aggregates, bridge the "materials and pressure gaps".
i) *Area-averaging operando spectroscopy*: CO oxidation on Pt/ZrO_2 prepared by atomic layer deposition (ALD) was examined by sum frequency generation (SFG) spectroscopy and near ambient pressure X-ray photoelectron spectroscopy (NAP-XPS), combined with mass spectrometry (MS) [1]. Combining experimental data with ab initio Density Functional Theory (DFT), we show that the reaction onset is determined by a delicate balance between CO disproportionation (Boudouard reaction) and oxidation. Disproportionation occurs on low-coordinated Pt sites at high CO coverages, when the remaining C-atom is stabilized by a favorable coordination. At variance with the general expectation, rough Pt nanoparticles are seemingly less active than smoother Pt films.
ii) *Spatially-resolved operando microscopy*: H_2 oxidation on polycrystalline Rh was studied by scanning photoelectron microscopy (SPEM) and photoemission electron microscopy (PEEM), which allow local surface analysis and visualising the heterogeneity of ongoing reactions on a µm-scale [2]. This revealed an anisotropy of surface oxidation (depending on the local step/edge density, yielding an *oxidation map*), as well as its effect on catalytic activity. In situ PEEM imaging of ongoing H_2 oxidation directly compares the local reactivity of metallic and oxidised Rh, demonstrating the effect of surface oxides. Employing the velocity of propagating reaction fronts as indicator of reactivity, a high transient activity of Rh surface oxide was observed. The corresponding *velocity map* reveals the structure-dependence of such activity, representing a direct imaging of a structure-activity relation for plenty well-defined surface structures within one sample.

[1] V. Pramhaas et. al, ACS Catalysis, 11 (2021) 208–214
[2] P. Winkler et al, Nature Communications, 12 (2021) 69

11.00 – 11.30 Prof. Per-Anders Carlsson, Dept. Chemistry and Chemical Engineering, Chalmers University of Technology, Göteborg:
"In Situ Time-Resolved FTIR Spectroscopy in Heterogeneous Catalysis"

11:30 – 12:00 Dr. Jolla Kullgren, Dept. Chemistry - Ångström, Uppsala University:
"Characterization of Defects at The Ceria Surface Using Simulations"