9th Annual Ambient Pressure X-ray Photoelectron Spectroscopy Workhop



Contribution ID: 33

Type: Oral contribution

Operando APXPS/SFG/XANES and in situ SPEM/PEEM of catalytic surface reactions

Wednesday 7 December 2022 16:40 (20 minutes)

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 interfaces [1]. Three examples of model and technological catalysts illustrate what can be learnt from synchrotron based spectroscopic and microscopic studies.

i) Operando APXPS/SFG/MS:

CO oxidation on Pt/ZrO2 prepared by atomic layer deposition (ALD) was examined by sum frequency generation (SFG) spectroscopy and ambient pressure X-ray photoelectron spectroscopy (AP-XPS @MAX IV), combined with mass spectrometry (MS) [2]. Complemented by Density Functional Theory (DFT), we show that the reaction onset is determined by a delicate balance between CO disproportionation and oxidation. ii) In situ SPEM/PEEM:

H2 oxidation on polycrystalline Rh was studied by scanning photoelectron microscopy (SPEM @ELETTRA) and photoemission electron microscopy (PEEM), which allow local surface analysis and visualising the heterogeneity of ongoing reactions on a µm-scale [3]. This revealed an anisotropy of surface oxidation, yielding an oxidation map. In situ PEEM imaging of ongoing H2 oxidation directly compares the local reactivity of metallic and oxidised Rh, revealing a high transient activity of Rh surface oxide, providing a direct imaging of a structure-activity relation for plenty surface structures. In a follow-up SPEM study [4], an unknown coexistence of four different states was observed: an active steady state, an inactive steady state and multifrequential oscillating states.

iii) Operando APXPS/XANES/MS:

Turning from model systems to applied catalysis, AP-XPS and X-ray absorption near edge structure (XANES @SLS/PSI) were employed to characterize Ni/ZrO2 and Ni/MgO-ZrO2 upon H2 pretreatment and during Partial Oxidation of Methane (POM) to Syngas at 750 °C (activity monitored by inline MS). During POM (partial) Ni re-oxidation occurred, although Ni° is often suggested as active phase, but the Ni oxidation state was sensitive to feed gas changes.

The insights by monitoring ongoing reactions may stimulate new ways of catalyst design.

References

- 1. G. Rupprechter, Small (2021) 2004289
- 2. V. Pramhaas et al., ACS Catalysis 11 (2021) 208–214
- 3. P. Winkler et al., Nature Communications 12 (2021) 69
- 4. P. Winkler et al., Nature Communications 12 (2021) 6517
- 5. J. Asencios et al., in preparation

Acknowledgements: Work supported by the Austrian Science Fund (FWF) and the Austrian Academy of Sciences (ÖAW).

if "Other", please specify:

I apply for a travel grant

No

Author: RUPPRECHTER, Günther (TU Wien)

Presenter: RUPPRECHTER, Günther (TU Wien)

Track Classification: Catalysis