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

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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^{o} 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
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if "Other", please specify:

I apply for a travel grant

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