

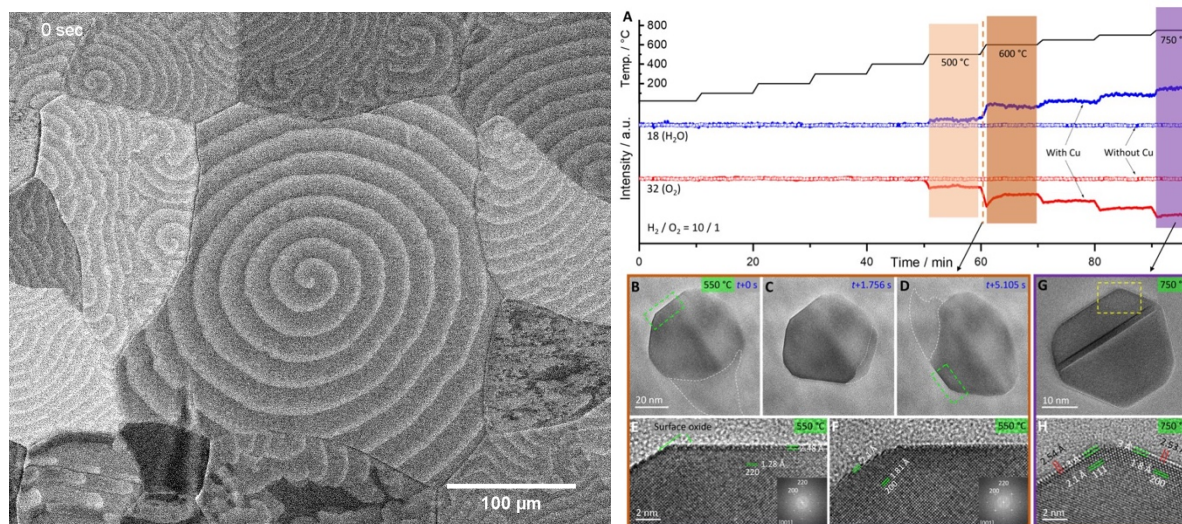
Oscillatory Dynamics at Catalytically Active Interfaces Studied by Multi-Scale Operando Electron Microscopy

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Abstract:

Our aim is to understand processes that lead to the emergence of catalytic function through direct observation using a combination of operando scanning and transmission electron microscopy. Starting with simple model catalysts, such as polycrystalline metal foils, we observe the propagation of chemical waves and reveal how catalytic activity depends on grain orientation, coupling mechanisms and reaction conditions [1]. In the case of redox-reactions on non-noble metals, we find that the active catalyst is operating near a phase-boundary where metallic and oxidized phases co-exist [2]. Real-time imaging reveals fascinating oscillatory redox dynamics that increase in complexity with increasing chemical potential of the gas-phase. When moving from simple model catalysts to industrially relevant metal nanoparticles supported on reducible oxide carriers, we apply in-situ transmission electron microscopy to study effects related to a strong metal-support interaction (SMSI) under reactive conditions [3]. Using the archetypal titania supported platinum nanoparticles as a reference system, and hydrogen oxidation as model redox reaction, it will be shown that the well-described encapsulated state of platinum particles is lost as soon as the system is exposed to a redox-active environment. Structural incoherence at the platinum-titania interface lowers the barrier for redox processes, which give rise to dynamic reconstructions and particle migration. The particle orientation on the support determines the structure of the interface and the resulting particle dynamics, migration, and sintering behaviour. The aim of the presentation is to demonstrate that active catalysts are dynamically adapting to the reaction environment and that catalytic function is related to the catalysts ability to participate in the reaction through reversible changes in its structure and/or (local) composition.



Left: Propagating reaction fronts observed by *in situ* SEM during NO₂ hydrogenation on polycrystalline Pt. **Right:** MS data showing consumption of O₂ and H₂O formation during H₂ oxidation on Cu particles. Simultaneously recorded TEM images reveal oscillatory redox dynamics and co-existence of Cu and Cu₂O. More details will be provided in the talk and are available in [1] and [2].

References:

- [1] Barroo C. et al. Nat Catal 3, 30–39 (2020).
- [2] Huang X. et al. Adv. Mater. 2101772 (2021).
- [3] Beck A. et al. Nat. Catal 4, 488–497 (2021)