

Self-sustained reaction oscillations in a new light

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Catalysts are complex material systems accelerating desired chemical reactions in chemical industry, fuel cells and car exhaust treatment. To improve their performance, an atomic-scale understanding of the interplay between catalyst structure, the surrounding gas composition and the catalyst activity under realistic reaction conditions is inevitable. Self-sustained reaction oscillations, in which the catalyst shuts its activity periodically off, have been studied for many years [1, 2] with the aim to avoid reactor instabilities or even reactor explosions. However, no general mechanism, especially for the structure leading to the activity increase and decrease, has been put forward.

Here we combined High Energy Surface X-Ray Diffraction (HESXRD) [3, 4], Planar Laser Induced Fluorescence (PLIF) [5], in-situ Mass Spectrometry (MS) and optical LED reflectance [6] at beamline P07 (DESY) at a photon energy of 77 keV to study self-sustained reaction oscillations during CO oxidation over Pd(001). This allowed, with sub-second time resolution, for correlating the catalyst structure (HESXRD) to the sample's CO₂ production (PLIF, MS) and hence its catalytic activity. The LED light, reflected from the sample surface, provided in addition immediate information on the surface roughness. Our data indicate that the oxidation and reduction of (111)-oriented Pd islands on top of an epitaxial PdO(101) oxide layer, previously reported under reducing conditions close to UHV [7], play a crucial role in the underlying mechanism for the self-sustained reaction oscillations.

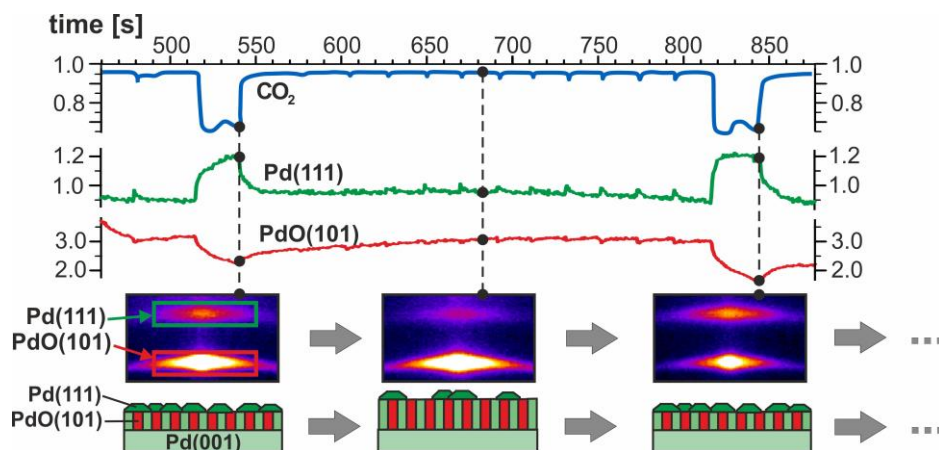


Fig. 1: CO₂ production during self-sustained reaction oscillations measured by in-situ mass spectrometry (blue), integrated peak intensities from reflections of (111)-oriented Pd particles (green) and the PdO(101) bulk oxide (red) deduced from HESXRD 2D maps (second-lowest panel) as function of time. Lowest panel: corresponding sample morphology models.

References:

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