Catalytic Reactions On Metal Surfaces: Imaging And Chemical Probing With Nanoscale Resolution

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This talk reviews recent progress made in imaging catalytic surface reactions and in providing information on the local chemical composition of reactive layers. Emphasis will be laid to the occurrence of surface restructuring and its influence on the reaction behaviour. The methodical approach is based on video-Field Ion Microscopy (FIM) and atom-probe mass spectrometry (PFDMS). The usefulness of the approach will be demonstrated in several case studies.

We shall start by inspecting shape transformation as found during the field-free interaction of oxygen (O_2) with Rh 3D specimens (given the form of "tips"). FIM reveals that the hemispherical morphology of a Rh tip is transformed into a polyhedral morphology by reaction with O_2 at temperatures between 500 and 550 K. The formation of large slopes and facets with {111} and {001} orientation is characteristic for the shape transformation. (1x2) and (1x3) missing-row reconstruction can be identified in the {113} and {011} planes of the Rh polyhedron.

Similar shape transformation is found while studying kinetic non-linearities in the NO and NO_2 reaction with hydrogen on the surface of a 3D Pt crystal. Both reactions are found to ignite in an explosive manner in the {012} corner planes of a Pt crystal which is reconstructed to resemble a polyhedron. An autocatalytic mechanism is in operation to form the water product. Local chemical probing demonstrates that the Pt surface is not in a purely metallic state.

Finally, CO oxidation on 3D gold crystals is studied at 300 K to demonstrate chemical wave propagation with nanoscale resolution while locally probing the surface for its chemical composition. The reaction is limited to step sites and involves multiple bonded CO (Au-carbonyls). Evidence is obtained that these species may even become mobile.