Watching catalysts at work: time-resolved spectroscopy under dynamic conditions

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Physico-chemical analytical methods are invaluable tools for materials characterization. A number of these methods have become indispensable to obtain key structural information on heterogeneous catalytic materials under operando conditions. However, spectroscopic experiments are typically performed at steady state under static reaction conditions thus in principle precluding our access to information on relevant species, e.g. intermediates. Structural changes associated to near surface variations involved in reaction mechanisms are often subtle or obscured by bulk structural changes or contributions, e.g. a significant portion of the support but also of a metal particle that are extraneous to the reaction. In pulsed experiments, the catalyst is stimulated to perform a specific event and to exhibit the structural changes associated with it. Because of the nature of such experiments, second to sub-second time resolution is required, while pulse repetition (modulation) improves the signal-to-noise ratio of any spectroscopic/diffraction method. Further enhancement of the structural change can be achieved by filtering the static signals, e.g. the unresponsive bulk or dominant adsorbed species, by phase sensitive analysis. From a catalytic view point, pulse experiments simulate well the operation of automotive catalysts. Hence, the potential of pulse experiments is demonstrated in selected cases using infrared, UV-vis spectroscopy and X-ray based methods to characterize catalysts involved in NOx selective catalytic reduction and three-way catalysis. It is demonstrated how subtle structural changes can be captured and their temporal response assessed precisely by this approach. This provides access to a detailed, often unprecedented, structural dynamics of catalytic systems.

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Born in 1972 in Bergamo, Italy, Davide Ferri obtained his PhD from the Swiss Federal Institute of Technology Zurich (ETHZ) in 2002 on in situ IR spectroscopy of catalytic solid-liquid interfaces. After a stay at Bruker Optics as sales and application manager, he moved to ETHZ as Oberassistent of Prof. A. Baiker. In 2007 he became group leader at Empa with a major focus on exhaust catalysis, perovskite-type oxides and operando spectroscopy. He is currently senior scientist at PSI with interests in catalysts for the emission control of natural gas and diesel vehicles, regenerative catalysts, catalysts for liquid phase reactions and the combination of time-resolved spectroscopy and diffraction methods to study catalysts at work.