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Structure-activity Relationships of Heterogeneous Catalysts from Time-resolved X-ray Absorption Spectroscopy

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Knowing the composition and the evolution of the bulk structure of a heterogeneous catalyst under working conditions (in situ) is a prerequisite for understanding structure-activity relationships and, eventually, for a rational catalyst design. X-ray absorption spectroscopy (XAS) can be employed to study a catalytically active material in situ. In addition to steady-state investigations, the technique permits experiments with a time-resolution in the sub-second range to monitor the structural evolution of bulk phases and from that to elucidate the solid-state kinetics of the reactions involved. Combined with mass spectrometry, the evolution of the short-range order structure of a heterogeneous catalyst, the average valence of the constituent metals, and the phase composition can be obtained. Molybdenum oxide based catalysts are extensively employed for the partial oxidation of alkenes. The reduction and the re-oxidation of MoO_{3-x} are the two crucial steps in the redox mechanism of partial oxidation reactions. Here we present results obtained from time-resolved studies on the reduction of MoO_3 in hydrogen and in propene, and on the oxidation of MoO_2 in oxygen. A comprehensive mechanism for the reduction and the re-oxidation of MoO_3 is proposed and the consequences of this mechanism for the partial oxidation of propene on MoO_3 are discussed.