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Selective Oxidation of Alkanes over Nanostructured Oxide Catalysts

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Oxides of vanadium and molybdenum are essential components of heterogeneous catalysts applied in selective oxidation of C₂-C₄ alkanes. Novel materials that effectively activate the highly stable C-H bonds in light alkanes and at the same time suppress further oxidation of desired products may contribute to radical changes in the raw material basis and the product streams in chemical industry. In this regard, selectivity is the key issue for sustainable utilization of resources and reduction of CO₂ emissions. Our efforts are focussed on understanding the correlations between nature and dynamics of molecular structures on the surface of Mo and V based catalysts and their selectivity in the oxidation of ethane, propane, and butane with focus on propane oxidation. The formation of oxygenates is studied over crystalline phases such as vanadium phosphates and molybdenum oxide based bronze structures, like the M1 phase of MoVTeNb mixed oxides. The catalytic performance of such systems strongly depends on the method of catalyst preparation. Morphology, microstructure and termination of the catalysts have been studied by electron microscopy, X-ray diffraction, microcalorimetry, photoelectron and FTIR spectroscopy. It is proposed that surface restructuring under conditions of propane oxidation may provoke spatial isolation of vanadium containing active sites. Highly dispersed V and Mo oxide species supported on surface-modified mesoporous silica (SBA-15) are used as model systems to investigate the effect of the degree of aggregation of metal oxo-species on the reactivity in oxidative dehydrogenation (ODH) of propane that generally represents the first reaction step in selective oxidation of propane to oxygenates. The results are discussed in terms of similarities and specifics of ODH and selective oxidation addressing the effects of (i) molecular structure of active ensembles on the catalyst surface, (ii) structural motives in the catalyst framework, (iii) chemical and structural complexity, (iv) oxidation state of the elements under reaction conditions, (v) the collective electronic properties of the solid, and (vi) the dynamics of the catalyst surface under varying operation conditions.