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Recent developments of in-situ surface analysis of selective oxidation catalysis.

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In situ analysis is intended to link quantitatively the catalytic performance of a system with physico-chemical properties of its geometric and electronic structure. This requires the experiments to be conducted such that on-line observation of catalytic performance is coupled to spectroscopic observations without transfer or breaking of reaction conditions.

The first example concerns the function of transition metal promoters in “superacidic” zirconia catalysts for butane isomerisation. Mn was found to be an active promoter inducing the idea that a sort of redox-reaction may be triggered by the multivalent Mn centres at the surface. Initial ex-situ EXAFS studies with frozen systems indeed suggested a correlation of the Mn valence with catalytic activity. A thorough real in-situ study, however, showed that while monitoring the catalytic activity no change of the Mn valence was observed classifying the apparent positive ex-situ correlation as transfer artefact.

VPO is a still enigmatic catalyst system, as we understand only very rudimentary the atomic-scale structure function relationship. The hypothesis about structure sensitivity opposes the notion about amorphous overlayers and little is experimentally known about the crystal structure in bulk and surface of the working catalysts. Using in-situ XAS at the V-Ledge and ex-situ HRTEM observations we arrive at the conclusion that a defective surface structure contains a small amount of centres that were observed to undergo reversible valence changes with their catalytic function. The high-resolution data available from cutting edge synchrotron radiation sources allow to predict the bond lengths of the active V-O interactions and to draw conclusions about realistic surface configurations of the working catalyst.