

Structure and Catalytic Function of Oxide Nanostructures

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The redox and acid-base surface properties of supported oxide nanostructures change markedly as their local structure and electronic properties vary with domain size. This evolution in domain size provides a broad range of catalytic behavior and site structures and also reaction chemistries often unavailable on the surfaces of bulk oxides. Turnover rates and selectivity for redox and acid catalysis depend on the size of these oxide domains, as they evolve from isolated cations into disordered clusters and finally into larger crystals with bulk-like properties. These changes in turnover rates reflect the ability of oxide domains to delocalize electron density, which is required to stabilize cationic reactive intermediates and transition states in acid and oxidation catalysis, respectively. Isomerization of alkyl-arenes, dehydration of alkanols, and oxidative dehydrogenation of alkanes on oxides of V, Mo, Nb, and W provide striking examples of the remarkable catalytic diversity introduced by surface structures and electronic properties that become available as catalytic structures change in size and dimensionality. The reactive and disordered nature of these small catalytic domains introduces structural and mechanistic challenges, which require that their local structure be probed during reaction and that these structures be related to their catalytic function at the level of elementary reaction events.