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Thermodynamic Properties of Oxidation Catalysts

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Many important heterogeneous catalysts function through and oxidation and reduction cycles. For example, the selective oxidation reactions that are carried out over vanadia-based catalysts proceed via Mars-Van Krevelen mechanism, in which the reactant is oxidized by the vanadia and the vanadia is then reoxidized in a separate step. Similarly, ceria plays a critical role in automotive-exhaust catalysis, where it is used to store and release oxygen via reduction and oxidation reactions. Therefore, a detailed understanding of reactions on these materials requires knowledge of their thermodynamic redox properties. While such thermodynamic information is available for bulk compounds, typical catalytic materials are in the form of nano-particles, supported on high-surface-area oxides, and often in the form of a mixed oxide or solid solution. The catalytic properties of these types of materials are known to be different but there is very little information about even the most basic thermodynamic information, such as oxidation enthalpies.

In this talk, I will discuss an electrochemical method for measuring redox thermodynamics of solids, coulometric titration. This technique has been applied to both ceria and vanadia catalysts in order to understand how redox properties are affected by mixing with a second oxide and by the presence of a support. The effect of changes in the oxygen binding on reaction rates will be discussed.