Defining Catalytically Active Sites and the Mechanism of Catalyzed Reactions: Challenges for the Experimentalist and the Theoretician

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The intrinsic activity and selectivity of catalysts are determined by the composition and structure of the catalyst. Advances in methods of catalyst characterization have led to a unprecedented level of detail concerning the structure of catalysts at the atomic scale. Studies carried using these techniques have led to the recognition that even when a uniform catalyst particle size can be achieved, it is still difficult to define uniquely what is meant by a catalytically active site. The situation is somewhat better for isolated site catalysts, for which the local composition and structure are virtually the same for all sites. This talk will illustrate the challenges of defining what is meant by an active site. The use of multiple experimental methods in combination with theoretical methods will be used to illustrate how one can ultimately achieve confidence in the definition of what is meant by an active site. We will also discuss how one can develop a picture for the mechanism of reactions catalyzed at isolated sites, and use this information to gain knowledge about the influence of local composition and structure on the kinetics of catalyzed reactions. As illustrations, we will look at the decomposition of N₂O, the direct oxidation of CH₄ to acetic acid, and the oxidation of CH₄ to CH₂O. In all cases, it will be shown that a complete picture of what is a catalytically active site and an understanding of the sequence of elementary steps involved in transforming reactants to products can only be achieved through the use of experimental and theoretical techniques in combination.