Structure and Reactivity in Catalytic Systems Involving Metal Oxides and Electrode Surfaces

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Structure and Reactivity in Catalytic Systems Involving Metal Oxides and Electrode Surfaces

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RESEARCH ACCOMPLISHMENTS

During the course of this contract, we have achieved the following:

1. We have developed secondary ion mass spectrometry as a kinetic tool and have used it to characterize a number of interesting metal-metal oxides and metal-adsorbate systems. The most significant of these include the behavior of rhodium and platinum on titanium dioxide thin films. We showed conclusively that the rhodium and platinum overlayers become encapsulated with TiO$_2$ when these metals on metal oxides are heated. This is the most direct evidence of longstanding interest in strong metal support interactions for the mechanism by which the interaction occurs. We have also characterized carefully the role of small amounts of impurities in noble metals and their influence on catalysis. These levels are established by secondary ion mass spectrometry and lie below the detectability limits of Auger electron spectroscopy. Finally, we have established the kinetics of surface decomposition of species like methoxide on platinum using SIMS to monitor the decomposition channels directly.

2. We have established a convenient way for measuring thermal desorption spectra of powders and in the same system taking Auger electron spectroscopy. This system has been used to characterize the migration of hydrogen atoms from transition metal particles onto the oxides which support them. The metals include platinum and ruthenium, while the oxides include silica and titania. This represents the first time that kinetic measurements (activation energies and preexponential factors) have been measured for this spillover process on well-characterized systems.

3. We have established, using copper-based systems, an inverse strong metal support interaction effect. This work establishes that charge transfer interactions are of some significance in these systems. Because copper has a filled d-electron shell, it cannot accept charge from an oxide in the same way that platinum does, for example. What we showed is that copper catalysts can be activated for hydrogen and CO chemisorption by placing the copper on an oxide support designed to withdraw electron density from the copper.
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