Final Technical Report for Grant DE-FG02-96ER14663, entitlted "Understanding and Controlling Metal-Support Interactions in Nanocrystalline Bimetallic Catalysts", Contract Period 9/15/96-10/14/01

James M. Howe[1] and Robert J. Davis[2], Principal Investigators
[1]Department of Materials Science & Engineering and [2]Department of Chemical Engineering
University of Virginia
Charlottesville, VA 22904-4745

Abstract: The objectives of this research were to: 1) determine the catalytic behavior of model Pd and Rh catalysts on unpromoted and ceria-promoted supports, for the reduction of NO and N_2O with CO, 2) determine the microstructures of the catalysts both before and after reaction in order to understand the catalytic behavior, and 3) understand the role of the metal/support interface in the catalytic process. The research examined the influence of Pd particle size and ceria loading on catalytic reaction for the NO+CO reaction. Dihydrogen chemisorption, temperature-programmed desorption (TPD) of NO, and high-resolution transmission electron microscopy (HRTEM) were used to characterize the catalyst samples. It was found that when ceria is used to promote Pd particles, the activity for NO+CO was a maximum for 2-nm-sized Pd particles. The maximum in activity results from a balance between the Pd/ceria interface, which enhances NO dissociation, and the close-packed planes of the Pd particles that facilitate product formation and/or desorption. The variations in apparent reaction orders and results from TPD were consistent with the idea that NO dissociation is promoted on very small particles (1 nm) and by the addition of ceria. Characterization of the catalysts by HRTEM showed that the ceria was typically present in the form of small crystallites from 3-7 nm in diameter, deposited near Pd particles, rather than as a thin film on the alumina support. This occurred whether the ceria was deposited before or after the Pd particles.

Keywords: Catalysis, Heterogeneous, Ceria promotion, Metal-support interface

Publications:
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3. J. H. Holles and R. J. Davis," Structure of $Pd/CeO_x/Al_2O_3$ Catalysts for NO_x Reduction Determined by In-Situ X-ray Absorption Spectroscopy", J. Chem. Phys. B, 104 (2000) 9653.
4. T. M. Murray and J. M. Howe, "Chemical Mixing and Growth Behavior of Ag and Au Nanocrystal Mixtures", in Proc. Intl. Conf. Solid-Solid Phase Transformations Inorganic Mater., TMS, Warrendale (2005) p. 797.

Human Resources: J. H. Holles received his Ph.D. based on this research and is now a professor in the Chemical Engineering Department at Michigan Technological University. T. M. Murray received his Ph.D. based on this research and he now supervises the Electron Microscope Facility in the Materials Science Department at the University of Washington.