

SPRING 2015 CHEMISTRY COLLOQUIA—Cosponsored by NCMN



Professor Gabor A. Somorjai University of California at Berkeley

March 20, 2015

3:00 Reception 548 Hamilton Hall 3:30 Seminar 112 Hamilton Hall



E. ROGER WASHBURN MEMORIAL LECTURE

Molecular catalysis science. Nanoparticle synthesis, and instrument development for characterization under reaction conditions. Conquering catalytic complexity.

Most heterogeneous, homogeneous and enzyme catalysts are nanoparticles. Colloid synthetic techniques of monodispersed metal nanoparticles in the 0.8 - 10 nm range with precise size distribution indicate that most catalytic reactions to be structure sensitive. Monometallic and bimetallic nanoparticles as well as core-shell structures with oxide shell and metal core are used to study multipath catalytic reactions with high product selectivity. It was found that the size and shape of metal nanoparticles control catalytic reaction rates and selectivities. In order to explore reasons of size and shape control high pressure in-situ surface techniques that include sum frequency generation vibrational spectroscopy, high pressure scanning tunneling microscopy and wellcontrolled oxide metal interfaces along with synchrotron techniques of X-ray spectroscopies are utilized under reaction conditions. Using these techniques, reaction intermediates, changes of surface composition of bimetallic catalysts under reducing and oxidizing conditions and mobility of reactants and surface structure using high pressure scanning tunneling microscopy were explored and determined. Small nanoparticles, below 2 nm range, were found to have changes of electronic structure that give rise to high oxidation state clusters under reaction conditions. These were utilized to heterogenize homogeneous catalysis using metal nanoclusters in the range of 40 atoms or less to carry out homogeneous catalytic reactions on heterogenized surfaces. Oxide metal interfaces were found to be Lewis acid to selectively control chemistry at very high turnover rates. Hydrogen and carbon monoxide catalytic oxidation generates a steady electron flow across platinum-titanium Schottky diodes. The thin metal film (about 4 nm) provides hot electrons that render the oxide metal interfaces active reaction sites. Nanocrystal bilayers carry out tandem catalysis using cerium oxide, platinum, silica sandwiches to produce combined methanol decomposition and hydroformylation chemistry instead of hydrogenation. In the future construction of catalyst architecture to increase selectivity of multipath reactions should be explored to carry out primary and secondary product reactions similar to enzyme catalytic processes.



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