

CHEMISTRY COLLOQUIUM

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11:25 AM, Room 111, Life Sciences

“X-Ray Spectroscopy in Catalysis Research: Application to Au Catalysts”

Jeff Miller
Chemical Sciences and Engineering Division
Argonne National Laboratory

The Advanced Photon Source (APS) at Argonne National Laboratory is one of the premier synchrotrons in the world and is just a few minutes from IIT. Experiments can be done at the APS that were not possible just a few years ago. This is especially true for characterization of heterogeneous catalysts. Most often X-ray absorption spectroscopy (EXAFS) has been used for structural determination although information about the electronic properties of metal particles is also available from the near edge spectra (XANES). This seminar will give a few examples for the application of EXAFS and XANES for understanding the steps in catalyst preparation, structural and electronic changes in small nanoparticles as well as determination of the active site during the catalytic cycle.

Catalyst Preparation: Hydrogen tetrachloroaurate (III), HAuCl_4 is the most frequently used compound for the preparation of supported gold catalysts. Deposition on supports, however, is only effective when the preparation solution is maintained at a pH of about 7. At these conditions, extensive hydrolysis of HAuCl_4 is observed. In addition, upon adsorption of HAuCl_4 solutions onto supports there are additional changes in the ligands bonded to Au^{+3} . Pre-treatment of the adsorbed Au in air or hydrogen lead to additional changes also determined by XAFS that alter the catalytic properties.

Effect of Au particle size: Au catalysts with metallic particles less than about 30 Å display enhanced catalytic activity. For particles in this size range, as the size of the particles decrease, there is a large decrease in the Au-Au bond distance up to about 0.15 Å. In addition, there are systematic changes in the intensity of the XANES spectra suggesting that small particles are electron rich compared to Au foil. These small Au particles also react with air at RT leading to oxidation of about 10% of the metallic atoms to Au^{+3} . These results suggest that catalytic activity is due to the changes in the electronic properties in small Au particles. A combination of the EXAFS and XANES spectra have helped identify the active site and time-dependent XANES studies have been used to identify the rate determining step in the catalytic cycle for CO oxidation.