PhD in Chemistry
Dissertation Defense

Presents a Seminar Titled:

"A Study of the CO Oxidation Reaction Mechanism Over Pt/SnO2/SiO2 Catalysts Using Diffuse Reflectance Fourier Transform Spectroscopy"

Presented By

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The study of the complex CO oxidation reaction mechanism over noble metal reducible oxide (NMRO) catalysts has proven to be integrally important to the currently relevant topics of atmospheric chemistry and automobile catalysis, as well as to a deeper understanding of heterogeneous catalytic reaction/ deactivation mechanisms. Heterogeneous catalysis is on the frontier of modern physical science, especially with the availability of powerful in-situ surface characterization techniques that can be employed to study catalytic reactions as they proceed. We utilized diffuse reflectance vibrational spectroscopy (DRIFTS) to study the complex CO oxidation reaction mechanism at 125°C over a series of Pt/SnO_2 catalysts coated onto silica gel. We observed that surface reconstruction to islands/aggregates/arrays of isolated OH, linearly bound PtCO and molecular water was a necessary prerequisite for CO oxidation over the $Pt/SnO_2/SiO_2$ catalysts. We also identified a low frequency Si-O-Si TO rocking mode that was created upon the formation of the Pt/Sn bimetallic catalyst and appeared to be necessary for CO oxidation activity. The low frequency Si-O-Si TO mode was observed in conjunction with islands of both isolated OH and linear PtCO on catalyst samples that were active for CO oxidation. To further characterize the CO oxidation reaction mechanism we measured the normalized peak height over time of all molecular species relevant to the reaction. Plots of the normalized peak height over time revealed the possibility of oscillations, subdiffusion, caging and memory, period adding, synchronization and unidirectional interaction between pairs of molecular species. When pairwise plane trajectories for *xy*, *xz* and *yz* phase planes for two (*x*,*y*,z) systems were plotted, the 2D plot of the normalized peak height of isolated OH and linear PtCO molecular species revealed diffusional jumps that preceded autocatalytic oscillations around two steady state solutions. From this analysis we predicted bistable kinetics between linear PtCO and isolated OH molecular species over $Pt/SnO_2/SiO_2$ catalysts. In addition we believe that we have possibly recorded two coupled Lorenz butterfly attractors with a common central axis. To test for deterministic chaos and underlying structure we carried out next amplitude analyses and attractor reconstructions, respectively, using the normalized peak height data. The reconstructed isolated OH and linear PtCO attractors both showed evidence for underlying structure that could suggest fractal dynamics. We also present the unexpected results of an additional experiment where we observed a possible phase transition during reductive CO pretreatment that involved both surface molecular species and the lattice modes of the glass substrate. Upon introduction of testgas, we observed generalized synchronization in the normalized peak height behavior of most of the molecular species participating in the CO oxidation reaction mechanism. We hope that the insights reported in this study will make a contribution to the exhaustively studied CO oxidation reaction mechanism as well as to the general field of heterogeneous catalytic reaction mechanisms.

Wednesday, April 23, 2014 at 11:00 in the CAVE