The Department of Chemistry and Biochemistry

Seminar Series

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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Diffuse reflectance vibrational spectroscopy (DRIFTS) was utilized to study low temperature CO oxidation (125 °C) over a series of $Pt/SnO_2/SiO_2$ samples. Molecular species relevant to the CO oxidation reaction mechanism were identified, and it was determined that surface reconstruction to islands of isolated OH, islands of linear PtCO and molecular water were a necessary prerequisite for CO oxidation over Pt/SnO_2 samples (on a glassy substrate). A low frequency Si-O-Si TO rocking mode was identified in the % transmission spectra of all $Pt/SnO_2/SiO_2$ catalysts examined; this mode was created upon formation of the Pt/Sn bimetallic catalyst and was determined 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; these modes were destroyed by acetone immersion. We measured the normalized peak height (Kubelka-Munk) over time of all molecular species relevant to the CO oxidation reaction, to construct an empirical reaction mechanism from the DRIFTS and % transmission data. Plots of the normalized peak height over time revealed oscillatory and nonlinear dynamics with fractional order reaction curves that predicted anomalous diffusion in the first 30 minutes into testgas reaction. After 30 minutes into reaction conditions, period doubling leading to period tripling, lag and phase synchronization and unidirectional interaction (proposed to be master-slave type) were observed between several different pairs of molecular species. Pairwise plane trajectories for *xy*, *xz* and *yz* phase planes were plotted for two (*x,y,*z) systems; the 2D plot of isolated OH and linear PtCO revealed diffusional jumps that preceded autocatalytic oscillations around two steady state solutions. From this analysis we propose bistable kinetics between linear PtCO and isolated OH over $Pt/SnO_2/SiO_2$. We believe that we have recorded two coupled Lorenz butterfly attractors. Next amplitude analyses and attractor reconstructions were conducted from normalized peak height data, to test for deterministic chaos and underlying structure, respectively. The reconstructed isolated OH attractor displayed a periodic nature throughout CO oxidation; and the reconstructed linear PtCO attractor showed three pronounced lobes in its structure. We also present the unexpected results of a modified experiment where we observed what resembled a phase transition during reductive CO pretreatment that involved both surface molecular species and the lattice modes of the glass substrate. Upon introduction of testgas generalized synchronization was observed in the normalized peak height behavior of most of the molecular species participating in the CO oxidation reaction mechanism.

Tuesday, March 19, 2013 at 12:00 in Education 208