DISSERTATION DEFENSE

Kangmin Xie

THE DONNA CHEN GROUP

“Understanding the Activity of Model Catalyst Surfaces: Ultrahigh Vacuum and Atmospheric Pressure Studies”

Abstract:

In order to provide insight into the development of new catalytic materials, a fundamental understanding of surface reactions is required. Vapor-deposited metal clusters on single-crystal surfaces are well-defined systems that can be used to understand the structure-relationships in commercial catalysts. The pressure gap between fundamental surface analysis under ultra-high vacuum (UHV, P~10^{-10} Torr) conditions and kinetic evaluation of catalysts under realistic pressures has been bridged by constructing a micro-reactor interfaced with UHV chamber: pre- and post-reaction surfaces can be examined by X-ray photoelectron spectroscopy (XPS) without exposing the sample to air. The performance of this micro-reactor has been verified by reproducing kinetic parameters for CO oxidation on Pt/TiO_2(110).

Pt-Re systems were studied in order to understand the nature of the enhanced bimetallic activity for oxidation reactions and the water gas shift (WGS) reaction. Model Pt-Re catalysts were prepared by sequential deposition of Pt and Re on a rutile TiO_2 (110) support. Active sites on the Pt-Re bimetallic clusters were investigated by temperature programmed desorption (TPD) using CO and methanol as probe molecules. At room temperature, Re on top of Pt can diffuse into Pt clusters in a kinetically limited process, and the surface composition of Re can be controlled by varying the Re coverage. Lattice oxygen participates in the recombination of dissociated CO and promotes CO_2 formation on bimetallic surfaces. In methanol oxidation reactions on Pt-Re surface alloys, higher long-term activity is observed for Pt-Re compared to Pt because less carbonaceous deposits are formed on the alloy surface. However, surface Re is unstable due to formation and subsequent sublimation of Re_2O_7. For the WGS reaction, bimetallic surfaces consisting of Pt on Re have higher activity than pure Pt, while Re alone has no activity. Furthermore, XPS studies show that the active species for WGS is metallic Re rather than Re oxides.


www.sc.edu/study/colleges_schools/chemistry_and_biochemistry/index.php

The University of South Carolina is an equal opportunity institution.