

ChBE 995 Autumn 2009



**The William G. Lowrie Department of Chemical and Biomolecular Engineering  
Graduate Program**

Cordially invites you to attend a seminar on

## **Oxidation and Reactivity of Transition Metal Surfaces**

**Thursday, October 29th, 11:30 a.m.**

Room 207 Koffolt Labs, 140 W. 19th Avenue  
Reception before the Seminar in Room 336 Koffolt Labs, at 11:00 a.m.

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### **Abstract**

Transition metals (TM) serve as catalysts under oxygen-rich conditions in applications such as natural gas combustion, exhaust gas remediation in lean-burn engines, and the selective oxidation of organic compounds. Under oxygen-rich conditions the metal surface can undergo several structural changes as it begins to oxidize, which in turn can dramatically modify the reactivity of the catalyst. Despite advances in our understanding of the oxidation of several catalytically important TM surfaces there is still disagreement in the exact surface phase that is associated with enhanced reactivity in systems such as CO oxidation on Pt. Therefore there is a need to better understand (1) the oxidation process and the structure of the oxygen phases that develop under various conditions (temperature, partial pressures) and (2) the resulting modifications in reactivity of the catalyst.

In this talk, I will first discuss work in our group examining the initial atomic-level steps in the oxidation of Pt and Pd(111) surfaces using Density Functional Theory (DFT), an accurate first-principles method. We have found a novel mechanism for the initiation of oxidation on Pt(111) that results in strongly buckled 1-D oxide chains on the Pt(111) surface. On Pd(111) this mechanism does not occur but instead subsurface oxygen becomes stable at lower oxygen concentration. I will discuss the differences in Pt and Pd that lead to these differences in oxidation mechanisms. I will also present some preliminary results in understanding the reactivity of CO and NO on the 1-D oxide chains on Pt(111). In the second part of my talk I will present examples from our DFT study of several small molecules ( $\text{H}_2\text{O}$ ,  $\text{H}_2$ , CO, and  $\text{CH}_4$ ) on the major oxide surfaces that form on Pd(111). We have found dramatic differences in reactivity between the 2D oxide phase that initially forms on Pd(111) and the bulk oxide that develops at higher oxygen concentrations. These differences can be attributed to changes in both the geometric and electronic structure of the different oxide surfaces. Our work provides new insight into the kinetics of oxidation of TM surfaces and demonstrates the sensitive link between atomic-level structure of the oxide and the reactivity of the oxide phase.