Department of Chemical and Environmental Engineering

2014—2015 Seminar Series

Friday, November 14, 2014

10:00—11:00 AM

Watkins 1000

Eric Schwegler

Quantum Simulations Group Leader

Physical and Life Sciences Directorate

Lawrence Livermore National Laboratory

First-principles modeling of semiconductor/water interfaces

Photoelectrochemical cells hold great promise for the sustainable production of hydrogen fuel from water using sunlight. Currently, the most efficient photocathodes are based on III-V phosphides; however, photocorrosion in the electrolyte solution remains a significant challenge. In an effort to better understand the complex chemistry involved, we have performed extensive first-principles molecular dynamics simulations water in contact with GaP and InP. In addition to explicitly accounting for the presence of liquid water, we have also studied the effect of experimentally reported oxygen-derived surface adsorbates. We find that surface oxygen dramatically alters the nature of the electrode-electrolyte interface, providing a kinetically feasible pathway for water dissociation. This results in the formation of an interfacial hydrogen-bond network. Interestingly, the dynamics of this network are qualitatively different for GaP and InP, with the latter exhibiting much greater network fluidity. We suggest that this translates to the capability of InP for long-range hydrogen transport in a way kinetically inaccessible to GaP. Implications for understanding initial interface formation, as well as the kinetics of photoelectrochemical hydrogen evolution and photocorrosion processes on GaP and InP surfaces, will be discussed in detail. This work was performed under the auspices of the U.S. Department of Energy by LLNL under Contract DE-AC52-07NA27344.

Biosketch: Eric Schwegler received his Ph.D. in Physical Chemistry in 1998 from the University of Minnesota, following undergraduate degrees in computer science and chemistry from Southwestern University. His thesis research was focused on the development of linear scaling electronic structure theory. In 1998, he joined Lawrence Livermore National Laboratory as a postdoctoral researcher and worked under the supervision of Giulia Galli, primarily in the area of first-principle simulations of fluids. In 2001, he was promoted to a staff scientist position at Lawrence Livermore and in 2005 he was appointed as the Quantum Simulations Group Leader in the Materials Science and Physics Divisions. He is also the Crosscutting Research Focus Area Lead for the Critical Materials Institute, a DOE Energy Innovation Hub. The Quantum Simulation Group (www.llnl.gov/qsg), which currently consists of 22 researchers, focuses on the application of state-of-the-art quantum simulation tools to understand and predict the properties of a wide range of materials.

*Light Refreshments will be served.