USC Viterbi School of Engineering

MORK FAMILY DEPARTMENT OF CHEMICAL ENGINEERING AND MATERIALS SCIENCE

Distinguished Lecture Series

Presents

Single atom catalysts on amorphous supports: a wild frontier for *ab initio* calculations

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Abstract

Several industrially important catalysts for olefin polymerization, metathesis, and epoxidation are atomically dispersed metal centers on amorphous supports, e.g. the Phillips ethylene polymerization catalyst, Cr/SiO_2). These heterogeneous catalysts elude the usual experimental and theoretical analyses because every site on an amorphous catalyst is different. Moreover, the disorder is "quenched" unlike the predictable dynamical disorder of a liquid environment. We discuss the mysterious ethylene-induced initiation in the Phillips catalyst at molecular and catalyst pellet scales. At the molecular scale, we show how prevalent *bis*(ethylene)Cr(II) complexes can generate active Cr(III)-alkyl sites via a new tethered homolysis mechanism. The computationally predicted initiation times and fractions of active sites compare well to experimental estimates. At the pellet scale, the porous support pulverizes itself upon initiation and the Cr/SiO_2 catalyst becomes diluted in an expanding sphere of polyethylene. Catalyst dilution leads to an effectiveness factor that *increases* with size of the expanding polyethylene/catalyst spheres. Thus, the observed activity increases with polymerization time may, in part, be due to early transport limitations. Finally, we present a new "Importance Learning" algorithm that combines machine learning and importance sampling methods to properly and efficiently average over the site disorder in *ab initio* studies of these catalysts.

Tuesday, November 19. 2019
4:00 pm – SLH 102
The Scientific Community is Cordially Invited.