

Catalysis Meets Quantum Mechanics: Accurate and Low-Cost Predictions of Complex Solvation and Reactivity Phenomena

The transition from fossil fuels to renewable energy calls for discoveries of new catalytic materials that will enable novel chemical transformations linking renewable energy sources (biomass, water, carbon dioxide, solar light) and high-quality fuels and value-added chemicals. Modern computational methods provide unique atomic-level insights into catalytic processes, often invisible to experimental techniques, and promise to reduce the reliance on tedious “trial-and-error” experimentation in catalyst design. The challenge lies in the high complexity of realistic computational models that comes with a steep increase in computational cost. In this talk, I will describe two research directions showcasing the power of current and emerging computational methods to describe complex phenomena in heterogeneous catalysis at low computational cost and with near-quantitative accuracy.

First, I will highlight our efforts in making reliable predictions of adsorption enthalpies of up to 55-atom molecules in hydrophilic (defected) and hydrophobic (pristine) microporous Lewis-acidic zeolites from dilute liquid solvent mixtures in complete agreement with calorimetry measurements. The structure of the complex environment inside the pores, observed with an atomistic resolution, is shown to influence the rates of zeolite-catalyzed reactions, such as alkene epoxidation by H_2O_2 .

Second, I will discuss how applying ideas from thermodynamics to quantum chemistry leads to a simple, highly accurate, and non-empirical method for making predictions of chemical reactivity at a low cost with broad implications across chemistry, catalysis, and materials science.