Quantum Mechanics based mechanisms for Catalysis and Electrocatalysis with extensions to large scale reactive simulations of Electrocatalysis

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Advances in theory and methods of quantum mechanics and in supercomputers are making it practical to consider first principles (de novo) predictions of the mechanisms of complex catalytic reactions.

We will highlight some recent advances in such methodologies including:

- New methods of continuum solvation for electrochemical reactions (CANDLE)
- Grand canonical QM calculations of electrochemical catalysis at constant potential (instead of constant numbers of electrons)
- QM Metadynamics calculations of free energies of electrocatalysis at operational temperature and potential
- Multiscale reaction simulations using Reactive Force Fields including polarization

which we will illustrate with recent applications to such catalytic systems as:

- The reaction mechanism for the Electrocatalytic oxygen reduction reaction (ORR) on metals alloys. This is the critical problem for H_2 fuel cells.
- Onset potentials and product distributions for electrochemical reduction of CO_2 to form hydrocarbon and oxygenated products. Cleaning up CO_2.
- The reaction mechanisms for the heterogenious selective oxidation of ethane, propane, and butane to chemical feedstocks. Decreased waste and energy
- Large Large scale (million atom) simulations of electrocatalysis. Realistic systems.