Computational Electrosynthesis and Development of Novel DFT-QM/MM Algorithms and Software

Jesse McDaniel
School of Chemistry and Biochemistry,
Georgia Institute of Technology
mcdaniel@gatech.edu

Numerous reactions in organic electrosynthesis proceed via highly reactive, radical ion species with the product yield/selectivity dictated by kinetic competition between various fast reaction pathways. Because these reactive species are electrogenerated within the electrical double layer environment at the working electrode interface, the reaction kinetics may be strongly influenced by large electric fields, high ion concentrations, and modulated solvation behavior. First-principles, quantum chemical description of the microscopic reaction kinetics must both incorporate the complex atomistic description of the electrochemical interface under applied voltage while facilitating statistical sampling to compute reaction free energy profiles; in this regard, quantum mechanics/molecular mechanics or "QM/MM" methods serve as an advantageous, multi-scale resolution approach. In this seminar, we will discuss our work on DFT-QM/MM algorithm and software development, and highlight how such multi-scale, quantum chemical modeling can be utilized to glean detailed understanding of cation radical reaction mechanisms within organic electrosynthesis.