The project involves an ongoing collaboration of 20+ researchers from University of Minnesota, University of South Carolina, Massachusetts Institute of Technology, and Pacific Northwest National Laboratory. The main goal of this project is to develop and apply innovative high-performance computing techniques and simulation methods for reactivity and dynamics of complex systems with special emphasis on the EMSL theme of the science of interfacial phenomena. The present proposal is concerned with application of advanced computational techniques to several fundamental fields of research including charge transport processes at interfaces, chemical and phase equilibria, thermochemical kinetics and rate constants, photochemistry, and spectroscopy. In particular, the proposal targets the following topics: (1) rational design of novel electrodes for solid oxide fuel cells, (2) nucleation and structure of aerosols, (3) metal-organic frameworks (MOFs) for adsorption, fractionation, and catalysis, (4) combustion mechanism of biofuels, and (5) spectroscopy of selected radical and ionic species in solution. The long-term goal of the first topic is to develop and apply more efficient computational methods and models for the design of complex oxide and metal-oxide surfaces and interfaces for high temperature electrochemical applications. In particular, we propose a computational study of the oxidation mechanism of hydrogen and natural gas and its reverse process, the reduction of water and carbon dioxide, on traditional nickel/yttria-stabilized-zirconia (Ni/YSZ) electrodes and novel electrodes based on doped perovskite crystals. The ultimate goal of the second topic is to better understand how liquid particles nucleate and grow in a multi-component gaseous mixture, and this is closely related to one area of the EMSL’s current interest which is the formation and evolution of aerosol chemistry in the environment. We are interested in studying the formation of atmospheric sulfuric-acid–water–ammonia nanoparticles and nucleation phenomena in silane-based dusty plasmas. Our work on gaseous adsorption on MOFs includes alkanes, olefins, and oxygen molecules on Fe-MOF-74 and other MOFs with a similar structure. The purpose of this project is to study the ability of MOFs to adsorb and separate a mixture of gases which involves high costs and energy inputs when performed by currently used techniques such as cryogenic distillation and also study their catalytic properties. This work is being pursued in collaboration with U. S. Department of Energy’ Nanoporous Materials Genome Center (NMGC). Our computational study of the combustion mechanism of biofuels carried out in collaboration with U. S. Department of Energy’ Combustion Energy Frontier Research Center will eventually lead to developing predictive, multi-scale models with relevance to practical fuel combustion. Our work on the spectroscopy of selected radical and ionic species in solution will involve several species that are important intermediates in biological and environmental reductive chemistries of NO, and this work will be carried out in close collaboration with the pulse radiolysis lab at Brookhaven National Laboratory. Overall, the proposed work will eventually lead to more accurate and comprehensive computational models and protocols, and the design of such models and protocols will allow for handling new challenging problems in the future.