Multiscale Modeling of Polymer Electrolyte Membrane Fuel Cell

Seung Soon Jang

Computational NanoBio Technology Laboratory, School of Materials Science and Engineering, Georgia Institute of Technology,771 Ferst Drive NW, Atlanta, GA 30332-0245, USA

We present what we have learned from our researches on polymer electrolyte membrane fuel cell systems using density functional theory (DFT) and molecular dynamics (MD) simulation method. For the polymer electrolyte membrane, our effort has been focused on the relationship between nanophase-segregation and transport properties. From our proton exchange membrane studies, it is clear that the interaction of polymer backbone with water molecules determines the nanophase-segregation, and such nanophase-segregation determines the internal structure of water phase as well as the distribution and dimension of the water phase. Based on these understanding on the proton exchange membranes, we pursue the working principles in anion exchange membranes. For fair comparison, we simulate proton and anion exchange membrane fuel cell using the same polymer backbone with the same molecular variables such as molecular weight, equivalent weight, water contents, and so on. The only difference is the ionic entities: the proton exchange membrane has sulfonate group with hydronium, whereas the anion exchange membrane has trimethyl ammonium group with hydroxide. From MD simulations, we investigate the relationship between nanophase-segregation and transport properties in the membranes. The nanophase-segregation is quantitatively evaluated by the structure factor analysis which characterizes the correlation length in reciprocal space. The transport of molecules such as water, hydronium and hydroxide is analyzed using the mean-square displacement.

Next, we expand the scope of this modeling study by including the electrode that has catalyst, carbon support, oxygen molecules etc. It is desirable to achieve fundamental understanding of the three-phase system consisting of carbon support, polymeric ionomer, and Pt catalyst nanoparticle where the most essential electrochemical events for fuel cell operation take place. Although variety of cutting-edge in-situ and time-resolved probing techniques would be available to investigate nanometer-scale systems, the structure and dynamic behavior of the three-phase system of fuel cell has not been thoroughly analyzed at molecular level, which is mainly due to such complicated multi-component feature of the three-phase system. In this context, we have simulated the three-phase systems using multi-scale first-principles modeling approach consisting of quantum mechanical density functional theory (DFT) and molecular dynamics (MD) simulations in order to investigate the nanophase-segregation of polymeric ionomers and water molecules, surrounding Pt nanoparticle on graphitic carbon support. For this, we develop a new force field based on DFT computations and run large-scale MD simulations. Main focus is to characterize the structure and transport properties in the three-phase interfacial system.