Molecular Studies of Supercapacitors: Ionic Liquids Adsorbed into Porous Carbon Electrodes

Peter T. Cummings

Department of Chemical and Biomolecular Engineering, and Multiscale Modeling and Simulation Center, Vanderbilt University, Nashville, TN 37235, USA

Energy storage continues to attract the attention of the scientific and engineering research communities. Renewable energy sources, such as solar and wind, require so-called grid-based energy storage so that energy generated during periods of high energy production can be made available during periods of low or zero energy production. Mobile energy storage (for everything from mobile phones to automobiles and beyond) is clearly increasing exponentially in importance. Among the many choices for energy storage devices, electrical double layer capacitors (EDLCs), also called supercapacitors, are attracting considerable attention. Supercapacitors store electrical energy via ion electrosorption directly in the EDLs at the electrolyte-electrode interface, suggesting that such liquid-solid interfaces play a dominant role in the underlying energy storage mechanism and the resulting device performance. Because electrical energy in supercapacitors is stored based on physical phenomena rather than chemical reaction (as in batteries), supercapacitors have fast rates of charge/discharge and a virtually limitless number of charge cycles (unlike batteries, which are often limited to 10^4 or less cycles). Much of the goal of supercapacitor research is aimed at increasing the amount of energy stored (energy density is the strong point in favor of batteries), which in turn focuses attention on the electrolyte, the nature of the electrode, and the electrode-electrolyte interactions.

To date, ionic liquids (ILs) have become emerging candidates for electrolytes used in supercapacitors, due to their exceptionally wide electrochemical window, excellent thermal stability, nonvolatility, and relatively inert nature; meanwhile carbons are the most widely used electrode materials in supercapacitors, due to their high specific surface area, good electrical conductivity, chemical stability in a variety of electrolytes, and relatively low cost. To improve the energy density and the transport properties of the charge carriers in supercapacitors, carbons have been developed in diverse forms such as activated carbons, carbon nanotubes (CNTs), onion-like carbons (OLCs), carbode-derived carbons and graphene. Using molecular modeling combined with molecular experimental probes, such as SAXS, SANS, NMR, and AFM, we report on our investigations into the interfacial phenomena occurring between the IL electrolytes and electrodes of varying geometries to understand the energy storage mechanism of supercapacitors that rely on EDLs established at IL-electrode interfaces.