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Title:	The Functionalized Graphene-Ionic Liquid Interface for Electrochemical Double-Layer Capacitors
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Abstract:	Functionalized graphene sheets (FGSs) are promising electrode materials for energy-dense electrochemical double-layer capacitors (EDLCs) as they can exhibit high electrical conductivity, specific surface area and intrinsic capacitance. However, engineering electrodes that capitalize on these attributes is challenging as FGSs tend to re-stack during processing, reducing the surface area available for capacitive energy storage. Furthermore, though room temperature ionic liquids (RTILs) have been introduced as next-generation electrolytes for EDLCs, a fundamental understanding of the capacitance of the FGS-electrolyte interface for concentrated electrolytes is lacking. This dissertation aims to develop approaches to produce energy-dense, FGS-based EDLC electrodes and elucidate the fundamental behavior of the electrochemical double-layer of the FGS-electrolyte interface. A simple "bottom-up" approach was developed to evaporatively consolidate mixtures of graphene oxide, alcohols, and RTILs, producing thin-film electrodes having an energy density as high as 6.3 Wh/kg at 3.0 V, based on the total mass of the electrode. To improve the energy density, dehydrated sucrose nanoparticles (0.5-2.5 nm) were added to serve as inter-sheet spacers. Liquid-cell atomic force microscopy revealed that these nanoparticles adsorb onto graphene oxide in an aqueous environment, providing a physical barrier to inter-sheet collapse due to van der Waals and capillary forces during solvent evaporation. Thin film electrodes were prepared having a gravimetric capacitance as high as 330 F/gFGS and, by tuning the nanoparticle and RTIL contents, an energy density of 13.3 Wh/kg was achieved. This energy density is the highest reported to date for an FGS-based electrode with a solid spacer. Lastly, electrochemical impedance spectroscopy was used to measure the effect of diluting an RTIL with miscible organic solvents on the intrinsic capacitance of the glassy carbon-electrolyte interface. Diluting 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide with organic solvents revealed a maximum in the minimum differential capacitance near 5-10 mol% RTIL that was not predicted by classical electrochemical double-layer theory. Additionally, diluting with a low-dielectric constant solvent, 1,2-dichloroethane, resulted in the largest increases in capacitance near the open circuit potential, compared to acetonitrile and propylene carbonate. These results provide insight into the electrochemical double-layer for concentrated electrolytes and reveal the potential for the enhancement of double-layer capacitance through dilution.
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