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Structure of Room Temperature Ionic Liquids on Charged Graphene: An integrated experimental and computational study¹ AH-MET UYSAL, HUA ZHOU, SANG SOO LEE, PAUL FENTER, Argonne National Laboratory, GUANG FENG, SONG LI, PETER CUMMINGS, Vanderbilt University, PASQUALE FULVIO, SHENG DAI, Oak Ridge National Laboratory, JAKE MCDONOUGH, YURY GOGOTSI, Drexel University — Electrical double layer capacitors (EDLCs) with room temperature ionic liquid (RTIL) electrolytes and carbon electrodes are promising candidates for energy storage devices with high power density and long cycle life. We studied the potential and time dependent changes in the electric double layer (EDL) structure of an imidazolium-based room temperature ionic liquid (RTIL) electrolyte at an epitaxial graphene (EG) surface. We used in situ x-ray reflectivity (XR) to determine the EDL structure at static potentials, during cyclic voltammetry (CV) and potential step measurements. The static potential structures were also investigated with fully atomistic molecular dynamics (MD) simulations. Combined XR and MD results show that the EDL structure has alternating anion/cation layers within the first nanometer of the interface. The dynamical response of the EDL to potential steps has a slow component (>10 s) and the RTIL structure shows hysteresis during CV scans. We propose a conceptual model that connects nanoscale interfacial structure to the macroscopic measurements.

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> Ahmet Uysal Argonne National Laboratory

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