High-performance supercapacitors based on $[N_{114}][NTf2]$ ionic liquid

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Achieving high energy density supercapacitors are the main objective of current electrochemical storage research. However, the electrochemical stability of these devices is primarily dependent on the synergy of their main components that summarizes the ion/solvent and ion/pore interaction. In this way, we aim to decipher the mechanism behind the charge accumulation process in supercapacitors filled with ionic liquids conductors and complex microstructured electrodes. Furthermore, we analyzed the fundaments of supercapacitors by *in-situ* electrochemical techniques of [N1114][NTf2] ionic liquid combined with $TEABF_4$-PC organic electrolyte in four different concentrations, assembled with *high-surface-area* activated carbon electrodes in a symmetrical two-electrode system. The electrochemical findings indicate that these ionic conductors' maximum work voltage window reaches 3.6 V with the irregular pores carbon-based electrode. We determined the best concentration of 1:1 ratio, in which the device reached over 600 Wh kg^{-1} of energy density and an incredible gravimetric capacitance of ~1,900 F g^{-1} at 2.6 A g^{-1}. Nevertheless, the analyses of the Mott-Schottky test and Potentiostatic Intermittent Titration Technique determined that the preferable charge accumulation process that forms the electrical double-layer occurs by crowding effect at high voltages due to a formation of the one-hump camel shape curve predicted by the Mean Field theory. In addition to that, the bare ions are confined in the narrow pores emerging into a superionic state that enhances the capacitance. Overall, we have determined the best electrochemical methodology to characterize ILs with carbon-based electrodes with a pure electrostatic process, describing the mechanism that reduces the equivalent series resistance in high gravimetric currents (> 1 A g^{-1}), positively affecting the resulting capacitance, energy, and power. This fundamental study highlights that *in-situ* electrochemistry could offer essential information about the energy storage mechanism for EDLC, which allows for improving the system response.

Keywords: Ionic Liquids, High-Density devices, Supercapacitors, Carbon Electrodes, Superionic State.

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