Advancing Structural Supercapacitors with Hydrated Polymer Electrolytes

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Structural supercapacitors, capable of bearing mechanical loads while storing electrical energy, hold great promise for enhancing mobile system efficiencies. However, developing practical structural supercapacitors often involves a challenging balance between mechanical and electrochemical performance, particularly in their electrolytes. Traditional research has focused on bi-continuous phase electrolytes (BPEs), which typically comprise high liquid content that weakens mechanical strength, and inert solid phases that hinder ion conduction and block electrode surfaces. Our previous work introduces a novel approach with a hydrated polymer electrolyte, demonstrating enhanced multifunctionality. This electrolyte, derived from controlled hydration of PET-LiClO4, forms a trihydrate (LiClO4·3H2O) structure, where water molecules bond with ions without forming a liquid phase, thereby improving ion mobility while maintaining the base polymer's mechanical properties. This new design also promotes better electrochemical interfaces with electrodes, a significant advancement over traditional BPEs.

In this study, we further enhance the performance and processability of such hydrated polymer electrolytes by incorporating polylactic acid (PLA) as the base polymer and lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) as the salt. The electrolyte, prepared through solution casting and subsequent controlled hydration, consistently remains an amorphous solid solution in both dry and hydrated states, as confirmed by DSC, XRD, and FTIR analyses. Our tests on ionic conductivity and mechanical properties reveal that adding water to the polymer electrolyte substantially increases ionic conductivity while retaining mechanical properties. A specific composition demonstrated a remarkable increase in ionic conductivity coupled with superior toughness surpassing the base polymer. Furthermore, we successfully fabricated and tested structural supercapacitor devices made of composites of carbon fibers and these new electrolytes. The prototypes presented enhanced toughness with significant energy storage performance, demonstrating their vast application potential due to their outstanding multifunctionality.