

Ph.D. DISSERTATION DEFENSE

Candidate: Ruhao Li
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School/Department: Charles V. Schaefer, Jr. School of Engineering and Science / Chemical Engineering and Materials Science
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Title: Utilizing Interfaces and Nanostructures in New Hybrid Electrolyte Designs for Enhanced Ion Transport

Chairperson: **Dr. Pinar Akçora**, Department of Chemical Engineering and Materials Science, School of Engineering and Science

Committee Members: **Dr. Henry Du**, Department of Chemical Engineering and Materials Science, School of Engineering and Science
Dr. Benjamin Paren, Department of Chemical Engineering and Materials Science, School of Engineering and Science
Dr. Jindal Shah, School of Chemical Engineering, College of Engineering, Architecture and Technology, Oklahoma State University

ABSTRACT

Polymer-based electrolytes address the critical challenge of creating high-performance, safe electrolytes for electrochemical devices by offering enhanced safety and mechanical stability compared to traditional liquid electrolytes; however, they often fall short in achieving comparable ionic conductivity. To address this limitation, this project introduces an innovative design strategy leveraging the self-assembly of polymer-grafted nanoparticles to form efficient ion-conducting pathways for improved ionic transport. This dissertation systematically investigates a series of interrelated systems—ranging from ionomers, polymer/ionic liquid mixtures and poly(ionic liquid) containing block copolymer-grafted particles. Our initial experiments in charged copolymer-grafted nanoparticles show the importance of percolation of ion clusters for ion transport. To investigate the role of the interface between the non-conductive filler and polymer, we design hybrid electrolytes based on single-ion conducting polymers grafted on nanoparticles. Poly(ionic liquid) (PIL)-grafted nanoparticles are synthesized for the first time and PIL chain length and assembled structures are found to improve the ion conductivity compared to that of particle-free poly(ionic liquid) homopolymer. Finally, copolymers comprising a single-ion conducting domain (PIL) and poly(methyl methacrylate) (PMMA) are synthesized, integrating ion-dipole interactions between PMMA and the ionic liquid, along with coordinated anion transport facilitated by the polycations. The variations in the PIL block length, diblock sequence, and the incorporation of ionic liquid into the copolymer hybrids modulate the net repulsion between nanoparticles, resulting in conduction pathways with distinct morphologies. The highest molar conductivity is observed with well-dispersed copolymer grafted nanoparticles, where the PIL coronas establish continuous pathways for ion transport. Moreover, the polarization and rearrangement of charged chains under applied electric fields demonstrate the potential of these hybrid electrolytes for applications in electroactive actuators, sensors and electrochemical devices.