HAJIM SCHOOL OF ENGINEERING & APPLIED SCIENCES
UNIVERSITY of ROCHESTER

Department of Chemical Engineering

Dr. Wyatt E. Tenhaeff

Staff Scientist, Oak Ridge National Laboratory

Solid State Electrolytes and Lignin-based Carbon Fibers for Safe and Inexpensive Lithium Batteries

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Before electric vehicles can become a viable option for mainstream consumers, several significant challenges in lithium batteries must be solved. Cost, safety, and range (i.e. total stored energy) are paramount among these challenges, and research groups across the globe are developing new materials and battery designs to address them. Two ongoing projects at Oak Ridge National Laboratory concerning solid state electrolytes and lithium ion battery anodes will be discussed.

Lithium metal offers a specific charge storage capacity that is an order of magnitude greater than graphite's (3860 vs. 372 mAh g^{-1}). Despite the lower specific capacity, conventional secondary lithium ion battery cells employ graphitic anodes due to safety considerations. We are developing solid electrolytes to enable safe, reversible cycling of lithium metal. To date, a solid electrolyte material with adequate ionic conductivity, compatibility with lithium metal, and mechanical robustness does not exist. Mechanical properties are a key consideration given their role in suppressing lithium dendrite formation, which has plagued lithium metal anodes to date and ultimately must be solved to enable their safe commercial implementation. Fabrication of composite materials where both phases contribute to Li cation transport can address these requirements. Our approach to understanding the transport of Li cations at interfaces between polymeric and inorganic solid electrolytes will be discussed. Initially, charge transport was studied in laminated electrolyte layers. Typical polymer electrolytes were poly (ethylene oxide) (PEO) or PEO-based copolymers mixed with a lithium salt, such as $LiClO_4$ or $LiCF_3SO_3$. Inorganic glasses (e.g. lithium phosphate oxynitride) and ceramics (e.g. $Li_7La_3Zr_2O_{12}$) were employed as the hard phase. It was discovered that the interfacial resistance was dominant in these systems but could be controlled and essentially eliminated through particular fabrication approaches. After understanding the charge transport at planar interfaces with well-defined areas, the study was extended to nanoparticulate composite solid electrolytes designed to optimize conductivities and mechanical properties. Initial results suggested that negligible lithium transport occurred through the particulate phases with higher intrinsic conductivities. Efforts to understand and reduce the interfacial impedance in these composites will be presented.

In the second project, lignin-derived carbon fibers (LCFs) were developed as low cost lithium ion battery anodes. Lignin is the second most abundant naturally occurring biopolymer and comprises 18-35% of wood by weight. LCFs were synthesized via industrially scalable melt-spinning and melt-blowing processes. Synthetic techniques were developed to fuse fibers into monolithic mats where the fibers were electrically interconnected. By virtue of their mixed electronic/ionic conductivities, the LCFs functioned as both current collector and active material, eliminating the need for massive copper foil current collectors. LCF mats with thicknesses of several hundred micrometers were galvanostatically cycled in half cells against Li, and specific capacities as high as 250 mAh g^{-1} were achieved. Lithiation and delithiation of the LCFs proceeded with coulombic efficiencies greater than 99.9%, and the capacity retention was greater than 99% over 70 cycles at a rate of 15 mA g^{-1}. LCFs carbonized at 2000°C were also shown to cycle reversibly in propylene carbonate electrolyte.

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