ABSTRACT

This dissertation addresses the significant challenge of enhancing the performance of lithium-ion batteries (LIBs) in extremely low-temperature environments, which is critical for applications in defense and space exploration. By innovating both electrolyte formulations and electrode materials, this research extends the operational boundaries of LIBs to temperatures below -100 ℃.

The initial phase of the research involves examining high salt concentration electrolytes using tetrahydrofuran (THF) solvent and lithium bis(fluorosulfonyl)imide (LiFSI) salt. The low freezing point and high solubility for lithium salts of THF are essential for maintaining fluidity and allowing Li^+ transport at low temperatures. The developed electrolytes demonstrate extensive FSI anion engagement in Li^+ solvation, enhancing Li^+ transport and leading to the formation of a stable and inorganic-rich solid electrolyte interphase (SEI). Further research incorporates a non-solvating diluent and fluoroethylene carbonate to reduce electrolyte viscosity without compromising the solvation structure and to improve SEI properties further. These advancements ensure stable Li plating/stripping behavior and enhance performance at temperatures as low as -40 ℃, with operational capabilities extending to -60 ℃.

Concurrently, the dissertation explores novel electrode materials aimed at further widening the low-temperature operability. Materials such as amorphous iron hydroxy selenide and niobium tungsten oxide are developed for their pseudocapacitive properties, which facilitate Li^+ kinetics. This is crucial for improving battery performance at low temperatures, as these materials support rapid Li^+ reactions and significantly enhance charge and discharge capabilities. This approach mitigates kinetic limitations typically seen in conventional intercalation-based electrodes, such as Li^+ desolvation and solid-state diffusion. The integration of these advanced electrode materials with the newly developed electrolytes results in exceptional performance under extremely low temperatures, demonstrating excellent performance at -100 ℃ and extending operability to -120 ℃.

Based on the findings of this dissertation, future research directions are also suggested. Future work should focus on developing electrolytes using commercially available electrode materials to ensure immediate applicability. Prototyping with larger formats and thermal safety validation of these materials for real-world applications are essential next steps. Additionally,

addressing gaps in our fundamental understanding of Li^+ transport mechanisms, particularly when anions are involved in Li^+ solvation, is necessary to further refine electrochemical models and guide the design of next-generation LIBs capable of operating under extreme conditions.

Overall, this dissertation provides a comprehensive study into the enhancement of LIBs for low-temperature applications. This work lays a robust foundation for future research aimed at further advancing LIB technology to ensure reliable performance in frigid environments, such as those found in defense and outer space applications.