ABSTRACT

This dissertation investigates the operational challenges faced by lithium-ion batteries (LIBs) in ultra low-temperature (ULT) environments, which are critical for LIBs to be fully implemented in extreme environments like aerospace, outer space, and artic conditions. The thesis primarily addresses the enhancement of the electrolyte, one of the two main limiting factors for LIBs poor performance, in conjunction with using more suitable electrodes for optimal functionality at ULTs, facilitating their integration into relevant systems.

The initial phase of the research on ULT LIBs centers on a locally highly concentrated electrolyte (LHCE). This electrolyte comprises a concentrated solution of lithium bis(fluorosulfonyl)imide (LiFSI) salt dissolved in a blend of two to three fluorinated carbonates, subsequently diluted with nonafluorobutyl methyl ether (NONA), a nearly non-polar diluent. The advantage of LHCEs over highly concentrated electrolytes (HCEs) lies in its maintaining the advantages of a HCE with its FSI^- anion rich solvation shell while also not having some of the adverse side effects as LHCEs have a reduced viscosity and freezing point (FP) due to the addition of NONA as a diluent. This novel electrolyte formulation enables LIBs to surpass traditional electrolytes at ULTs, exhibiting low desolvation energy, high capacity retention, and a stable, uniform inorganic solid electrolyte interface (SEI).. Further research explored the ideal concentration ratios of LiFSI vs. lithium bis(trifluoromethane)sulfonimide (LiTFSI), alongside variations of ratios for the chosen fluorinated carbonates and diluent. These adjustments significantly enhanced the electrochemical performance of LIBs at ULT, ensuring a high capacity retention at ULT.

The collected data facilitated the development of a hybrid photovoltaic (PV)-LIB system for energy generation and storage, designed to emulate operational scenarios in extreme environments like aerospace, outer space, and artic conditions. The system charges the LIB using a PV module prior to the controlled discharge of the LIB, whose discharge procedure can be modeled after whatever intended device or application. This design aids in the testing and scaling of novel ULT batteries, particularly when integrated with PV cells, to assess their combined efficacy at ULTs. Given that ULTs natural lead themselves to uses where a PV cell would be the major source of its energy, this system provides valuable insights into the individual and collective performance of both components.

Future research directions, as indicated in this study, include exploration on more

suitable ULT electrodes as only preliminary studies were conducted with more suitable electrodes and further investigation on a more fundamental level regarding how the electrolyte composition effects both the solvation structure and Li^+ transport inside the battery.

In summary, this dissertation delves into how electrolyte modification can enhance the performance at ULTs and when paired with suitable electrodes for these temperatures, is capable of producing a LIB that has exceptional performance at ULTs. The works focus primarily on deeply exploring the effects of electrolyte modification on ULT performance and on how LIBs actually operate when combined into more systematic testing at ULTs. This work is on laying a solid groundwork for future researchers to build off and advance the field further to allow for LIBs to see further use in extreme environments.