Abstract

Energy storage is a fundamental and essential component of modern society, driving technological breakthroughs, sustainability, and daily activities. The stabilization of power services, the improvement of portable electronic devices, and the development of electric vehicles and renewable energy sources are all largely dependent on the availability of reliable and efficient energy storage systems. In this context, this thesis investigates the development and characterization of novel electrolytes for solid-state lithium-metal battery synthesized through UV/thermal *in-situ* polymerization. By developing ionogels (ion gels) and hybrid/composite ionogels as the electrolytes, the study aims to enhance the cells performance at room temperature, focusing on improving ionic conductivity, thermal stability, cyclability, etc., and thus, to contribute to the development of safer, more efficient battery systems, pushing the boundaries of current lithium battery technology and paving the way for next-generation safe and high energy density energy storage solutions.

Chapter 1 provides an overview of energy storage solutions/technologies, with a specific emphasis on electrochemical energy storage, and progressively delves into the specifics of batteries. It explains the main parameters crucial for understanding battery performance and sets the stage for the subsequent detailed investigations.

In Chapter 2, the focus shifts to the introduction to battery electrolyte systems, which discusses their critical role, the importance of ion transport, and electrolyte/electrolyte interphase in a battery cell. Solid-state electrolytes are introduced, with an emphasis on polymer-based electrolyte systems and their synthesis methods, including polymer-in-salt electrolytes, ionogel electrolytes, composite/hybrid ionogel electrolytes, and free radical polymerization techniques such as UV/thermal crosslinking. This discussion highlights the significance of these materials in advancing lithium-metal battery technology.

Chapter 3 details the methodologies employed in this research, covering a range of materials characterization techniques, including infrared spectroscopies, thermal-mechanical analyses, gel content analysis, rheology and photo-rheology, Raman spectroscopy, X-ray photoelectron spectroscopy, and scanning electron microscopy. Additionally, electrochemical characterizations such as electrochemical impedance spectroscopy, cyclic voltammetry and charge/discharge cycling are outlined. These approaches aim to offer a strong framework for assessing the characteristics and effectiveness of the developed electrolyte membranes.

The initial experimental work, as discussed in Chapter 4, presents the results of two types of ionogel electrolytes composed of different room temperature ionic liquids, salt, and mono/bi-functional methacrylate oligomer mixtures, synthesized through a rapid and solvent-free *in-situ* UV-curing. Furthermore, the addition of micro-scale LLZO particles showed significant preliminary improvements in cycling stability compared to ionogels, even with high-energy cathodes, laying the groundwork for further investigations.

Chapter 5 expands the study to hybrid ionogel membranes that include large proportion of LLZO nano particles. The membranes are prepared through solvent-free *in-situ* thermal-polymerization, and the results showcase notable ionic conductivities. This chapter also examines the incorporation of various 1M ionic liquid electrolyte solutions to create diverse anionic environments within the hybrid ionogel systems, investigating their impact on the properties and electrochemical performance of resulting electrolyte membranes.

The Thesis Conclusion summarizes the main findings and achievements, with confirmation of advantages of the preparation methods and emphasis on the enhanced performance of the developed electrolytes. Potential future directions are proposed. Altogether this work should help design safer, more efficient Li-metal batteries and encourage future research in this intriguing subject, which may allow the development of high-performing, safe and sustainable next-generation solid-state energy storage solutions.