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Article / Book Information

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Category(English)	Doctoral Thesis
種別(和文)	論文要旨
Type(English)	Summary

（博士課程）
Doctoral Program

論 文 要 旨

THESIS SUMMARY

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要旨（英文800語程度）
Thesis Summary（approx.800 English Words ）

The increasing demand for high-performance energy storage systems has driven extensive research into next-generation batteries, with lithium-sulfur (Li-S) batteries emerging as a promising candidate. Among these, all-solid-state lithium-sulfur batteries (ASSLSBs) have gained particular attention for their potential to overcome the challenges of liquid-electrolyte-based systems, such as safety concerns, polysulfide dissolution, and dendrite growth. However, the inherent limitations of sulfur-based active materials, solid electrolytes, and anode configurations necessitate innovative approaches to achieve practical performance. This thesis focuses on addressing these challenges by designing novel materials and configurations for ASSLSBs, particularly in anode-free architectures, and demonstrates their potential through systematic exploration of experimental and theoretical approaches.

Chapter 1 provided a comprehensive overview of secondary batteries, emphasizing the evolution and challenges of lithium-sulfur batteries and all-solid-state lithium-sulfur batteries (ASSLSBs). Key limitations, such as low ionic and electronic conductivities, significant volume changes, and interfacial instability, were identified as critical barriers to achieving high energy density and cycle life. These insights framed the research objectives, which included enhancing the mixed conductivity of Li_2S-based cathodes, constructing a stable cathode framework with 3D conductive pathways, improving interfacial contact, and optimizing anode-free configurations for ASSLSBs.

Chapter 2 detailed the methodologies and theoretical bases employed, including advanced liquid-phase synthesis techniques, density functional theory calculations, and sophisticated characterization tools. These methods formed the foundation for the subsequent experimental and computational investigations.

Chapter 3 focused on the development of the ACSV (Active material–Carbon replica 10–Solid electrolyte–Vapor-grown carbon fiber) cathode composite based on liquid-phase techniques. By integrating Li_2S-LiI active materials, a porous carbon framework CR10, a high-conductivity liquid-phase synthesized sulfide electrolyte, and conductive vapor-grown carbon fiber, this composite achieved remarkable ionic and electronic conductivity, as well as robust structural stability. Structural and electrochemical analyses demonstrated the effectiveness of this design in addressing the inherent limitations of Li_2S-based cathodes.

Chapter 4 primarily demonstrated the electrochemical performance of all-solid-state lithium-sulfur cells utilizing the ACSV cathode. The results showed significant improvements in capacity retention, cycling stability, and rate capability, highlighting the synergistic effects of the composite design. Through carefully designed control experiments, the chapter systematically evaluated the individual contributions of LiI and CR10 within the ACSV system, providing critical insights into the composite's functional mechanisms.

Chapter 5 explored the development and characterization of metal-coated Cu–X (X = Al, Ag, Au) current collectors for anode-free all-solid-state lithium-sulfur batteries (AFASSLSBs). The research encompassed: phase identification to confirm membrane composition, morphological analysis to validate film thickness, first-principles computational studies to assess the impact of three different metals on lithium deposition, electrochemical performance evaluation of anode-free all-solid-state lithium-sulfur full cells employing the novel Cu/X current collectors, and comparative energy density calculations. As a result, the incorporation of metal coatings facilitated uniform lithium deposition, suppressed dendrite formation, and enhanced interfacial stability. The electrochemical performance of the AFASSLSB full cell employing the Cu–Ag collector and the ACSV cathode validated its potential for achieving high energy density and long-term stability.

Chapter 6 summarized the results of the previous chapters. This thesis successfully tackled key challenges in the development of ASSLSBs by integrating innovative material designs and advanced fabrication strategies. The ACSV cathode composite and Cu–Ag current collectors for anode-free architectures demonstrated remarkable potential for improving ionic/electronic conductivities, mitigating volume changes, and stabilizing interfaces. Furthermore, the research provided valuable insights for understanding electrode material properties through computational science, making significant contributions to fundamental science. These advancements enhance our understanding of material interactions within ASSLSBs and highlight practical pathways for achieving high energy density, safety, and long-term cycling stability.