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Ph.D. DISSERTATION DEFENSE

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Degree:	Doctor of Philosophy
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Title:	**Advanced Strategies for High Performance Sulfide All-Solid-State Batteries**
Chairperson:	**Dr. Jae Chul Kim,** Department of Chemical Engineering and Materials Science.
Committee Members:	**Dr. Alyssa Hensley,** Department of Chemical Engineering and Materials Science. **Dr. Benjamin Paren,** Department of Chemical Engineering and Materials Science. **Dr. Weina Meng,** Department of Civil Environmental & Ocean

ABSTRACT

Recent advancements in lithium-ion battery technology have made zero-emission electric vehicles (EVs) a reality, increasing the demand for next-generation batteries like all-solid-state batteries (ASSBs). These batteries, leveraging nonflammable solid electrolytes and lithium metal, offer higher safety and energy density. However, the introduction of solid-state electrolytes leads to solid–solid interfaces, where mechanical degradation from volume changes in the cathode during cycling significantly limits battery lifespan. Additionally, the low conductivity of solid-state electrolytes reduces the overall capacity.

This dissertation identifies key scientific solutions to address these limitations. The introduction of zero-strain cathodes in combination with amorphous Li_3PS_4 (LPS) solid electrolytes significantly reduces interface degradation and void formation due to negligible volume change (0.12%), leading to improved mechanical stability and extended cycle life. Moreover, our study reveals that the electrochemical performance of sulfide-based noncrystalline solid electrolytes is governed by their mid-range structural order, local chemical environments and Li pathway percolation, with these factors playing a critical role in reducing activation barriers and improving ionic conductivity. This insight allows for the enhancement of battery performance by optimizing noncrystalline solid electrolyte structure. Furthermore, a novel "mid-range peak area fraction index" is proposed to quantify the simplified medium-range structure of noncrystalline materials, providing a new tool for analyzing noncrystalline materials systems.

These findings offer significant implications for the development of high-performance solid-state batteries, providing pathways for their practical application in electric vehicle technologies.