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Zhi Deng Dissertation Defense
NanoEngineering Ph.D. Candidate
Materials Virtual Lab

"First Principles Modeling of Lithium Solid Electrolytes"

PI: Dr. Shyue Ping Ong
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Abstract: Developing all-solid-state lithium batteries with inorganic solid electrolytes can potentially address the safety concerns caused by using flammable organic liquid electrolytes in traditional lithium-ion batteries. Though the discovery of new solid electrolytes with exceptionally high (on par or even exceeding organic solvents) ionic conductivities have re-energized all-solid-state lithium battery research in recent years, many practical challenges remain, hindering large-scale applications. In this thesis, we demonstrate how density functional theory (DFT) calculations can be used to provide crucial materials insights to address these challenges. This thesis is broadly divided into two topics. In the first topic (Chapters 3 and 4), we will investigate bulk solid electrolyte properties such as ionic conductivity, diffusion mechanisms, electrochemical stability and mechanical properties using DFT calculations. We will show that Li excess interstitials are crucial to achieving reasonable ionic conductivity in Li_6PS_5Cl by promoting diffusion between Li_6S cages. Li_6PS_5Cl is also shown to be metastable with limited intrinsic electrochemical window. We have also carried out a large-scale study of the elastic properties of most known alkali solid electrolyte candidates, quantifying relationships between the chemistry and mechanical properties. In the second topic (Chapters 5 and 6), we develop approaches to apply atomistic-scale DFT calculated data to probe diffusion at much larger length scales. By combining bond percolation analysis with DFT-calculated local-environment dependent diffusion barriers, we identify composition ranges with potentially improved ionic conductivities in the anti-perovskite $Li_3OCl_{1-x}Br_x$ superionic conductor. We also demonstrate how large-scale DFT calculations can be used to train a quantum-accurate interatomic potential for Li_3N. This electrostatic Spectral Neighbor Analysis Potential (eSNAP), which combines a rigorously defined local environment descriptor with an electrostatic model, is then applied to large scale transport studies that are well outside the accessibility of expensive *ab initio* molecular dynamics (AIMD), such as the computation of thermodynamic factors and grain boundary diffusivity.