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Tuning Ion Mobility in Lithium Argyrodite Solid Electrolytes via Entropy Engineering

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Solid-state batteries (SSBs) are attracting great interest leading to potentially higher energy and power densities compared to conventional Li-ion batteries based on liquid electrolytes. However, they are plagued by the development of advanced solid electrolytes (SEs), mainly lacking in ionic conductivity and electrochemical stability; thus, the ongoing quest for exploration of new materials and compositions. Inducing a large structural disorder, i.e. high configurational entropy, has recently emerged as a new strategy to overcome limitations of conventional SE materials. In this regard, few multicomponent SE materials have been reported up to now, showing favorable charge-transport properties [1,2]. However, a thorough understanding on how configurational entropy affects ionic conductivity is lacking. In this regard, we have investigated a large series of multication- and anion substituted lithium argyrodites. Structure-property relationships related to ion mobility were probed using a combination of powder diffraction techniques, solid-state nuclear magnetic resonance spectroscopy, and charge-transport measurements.
In this contribution, we present an overview of our recent work and show correlations between configurational entropy and ionic conductivity [1,3]. To the best of our knowledge our results present the first experimental evidence of a direct correlation between occupational disorder in the cationic, or anionic host lattice and lithium transport. By controlling the configurational entropy through the composition, i.e. entropy engineering, high bulk ionic conductivities up to 20 mS/cm at room temperature were achieved for optimized lithium argyrodite compositions. Our results indicate the possibility of improving ionic conductivity in ceramic ion conductors via entropy engineering, unlocking the compositional limitations for the design of advanced electrolytes and opening up new avenues in the field.

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