Computational investigation and design of solid lithium-ion electrolytes based on the $Li_{10}GeP_2S_{12}$ superionic conductor

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$Li_{10}GeP_2S_{12}$ (LGPS) is a recently discovered lithium super ionic conductor. This new material has the highest conductivity ever achieved among solid lithium electrolytes of 12 mS/cm at room temperature and outstanding electrochemical performance in Li batteries. Our previous study have shown that LGPS is a metastable phase in the calculated phase diagram and that LGPS is not stable against reduction by lithium at low voltage or extraction of Li with decomposition at high voltage. We also identified that LGPS is a three-dimensional ion conductor rather than a one dimensional ion conductor, and our calculated overall activation barrier and conductivity are in remarkable agreement with the experimental results.[1]

In this talk, we will present our study of lithium superionic conductor electrolytes based on LGPS using first principles calculations. We will demonstrate how first principles molecular dynamics simulations can provide insight into the occupancies of the lithium sites in the LGPS structure, and the mechanisms for diffusion in this structure. We will also demonstrate how first principles calculations can be used to perform materials design by studying how the phase stability, electrochemical stability, and Li+ ionic conductivity of LGPS is modified by cation substitutions (Ge for Si, Sn, Al or P) and anion substitutions (S for O or Se).[2] We will then make recommendations for modifications of LGPS to mitigate some of its limitations.

References:

[1] Mo, Y.; Ong, S.P.; Ceder, G. “First principles study of the $Li_{10}GeP_2S_{12}$ lithium super ionic conductor material” *Chemistry of Materials*, 24, 15-17 (2012)
[2] Ong, S. P., Mo, Y., Richards, W. D., Miara, L., Lee, H. S., & Ceder, G. (2013). Phase stability, electrochemical stability and ionic conductivity of the Li10±1MP2X12 (M = Ge, Si, Sn, Al or P, and X = O, S or Se) family of superionic conductors. *Energy & Environmental Science*, *6*(1), 148.