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First-Principle Cluster Expansion Study of Ta-Doped Tetragonal Li7La3Zr2O12 (t-LLZO) Oxide-Garnet Solid-State Electrolyte

The oxide-garnet tetragonal Li7La3Zr2O12 (t-LLZO) is a candidate material for use as a solid electrolyte in Li-ion batteries. In order to use t-LLZO in practical devices, it must be able to withstand high temperatures with good phase stability. Thus, doping with a supervalent cation, such as Ta, at the Zr-site of t-LLZO is an effective way to enhance the low-temperature tetragonal crystal structure. Using first-principle calculations combined with cluster expansion, this study generates new possible stable phases of Ta-doped LLZO. Furthermore, Monte-Carlo simulation was used to better understand the behavior of the Ta-doped phase as a function of temperature under the canonical ensemble. The cluster expansion generated 36 new multi-component Li5La3Zr2-xTaxO12 structures, where all of the new structures are thermodynamically stable with a negative enthalpy of formation. The Monte-Carlo temperature profiles have a miscible gap, indicating that there is no phase separation and the system mixes very well at ~900K. Further density functional theory calculations were performed on the most stable generated Ta-doped LLZO structures to determine the structural, mechanical, and electronic properties of the structures for their application as active solid-state electrolytes. It was found that the generated structures exhibit good structural stability due to the smooth decrease in calculated lattice parameters. Moreover, the structures show good elastic stability against deformation and exhibit magnetic separator behavior, which encourages their use as the next-generation solid electrolytes. Hence, the phase stability insight is crucial for the development of these promising solid-state electrolytes for all-solid-state Li-ion batteries.

Keywords: Solid Electrolyte, Doping, Phase stability, Cluster Expansion, Monte Carlo, DFT

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