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Multi-Faceted Approaches To Enhance The $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ Solid Electrolyte Material

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Solid-state technology is by far the most intriguing development to curb the thermal instability associated with the reactivity of liquid electrolytes. Of particular interest is the cubic $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO) solid electrolyte material with ionic conductivity of 10^{-3} to $10^{-2} \text{ S}\cdot\text{cm}^{-1}$. However, it has temperature-dependent phase instability wherein the low conducting tetragonal phase exists below 630°C whilst the highly conductive cubic phase is only stable at high temperatures. Herein, we explore several strategies incorporated in the fundamental enhancement of the LLZO solid electrolyte i.e. supervalent doping, nanosizing, and subsection to pressure. These were carried out through the exploration of various computational approaches: cluster expansion, ab-initio simulations, and molecular dynamics. The cluster expansion approach was employed to assist in generating new stable structures for the supervalent doping with Ta and W on the Zr site. The Ta-doped structures' electronic and mechanical properties were studied and demonstrated the presence of a band gap of 4.6 eV. alternatively, as an initial approach to understanding the thermodynamic properties of LLZO, the bulk structure was subjected to small pressure during molecular dynamics simulations. Systematic induction of pressure yielded a transition of the tetragonal phase to the cubic phase at 2 GPa pressure. The lattice parameters for the cubic and tetragonal phases, acquired in the study were within 0.38 % agreement with the literature. Furthermore, the XRD graphs confirmed varying phases under different pressure conditions. The temperature phase diagram for 0 GPa structure agreed well with the literature trends and interestingly, the 2 GPa structure retained the cubic phase at various temperatures and was confirmed in the XRDs and temperature phase diagram. The final approach was systematic temperature variation on the bulk and nanostructured LLZO under various temperature conditions in the range 300 – 1500K to understand the LLZO high temperature behaviour and monitor the temperature-induced diffusion. The calculations revealed two distinct patterns of Li-ion transport on the diffusion coefficient plots. The trends in conductive ion diffusions demonstrated that nanostructured materials yielded higher diffusion coefficients than those of the bulk structure. These findings have laid a basis and will contribute towards the evaluation of ionic conductivity and structural stability retention aided by computational simulations.

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Primary author: Prof. LEDWABA, Raesibe (University of Limpopo)

Presenter: Prof. LEDWABA, Raesibe (University of Limpopo)

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