Structural Effects on Lithium-Ion Diffusion in Perovskite Lithium Lanthanum Titanate: an Ab Initio Molecular Dynamics Study

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All-solid-state lithium-ion batteries are one of the promising next generation battery technologies. The replacement of the flammable organic liquid electrolyte with an inorganic solid electrolyte in a battery cell greatly mitigates the safety issue. Besides, it enables the use of lithium metal anode that has a very high theoretical capacity. The key for the realization of the technology is finding a suitable solid-state lithium-ion conductor with a sufficiently high ionic conductivity. Perovskite lithium lanthanum titanates exhibit high bulk lithium-ion conductivities among oxides [1]. Although a bulk conductivity at room temperature as high as 1×10^{-3} Scm^{-1} has been reported [2], the value is still orders lower than that of liquid electrolytes. There is therefore a need to further improve the lithium-ion conductivity in oxide solid electrolytes.

Extending our previous work on battery materials [3, 4], in this study lithium-ion diffusion in perovskite lithium lanthanum titanate is investigated using ab initio molecular dynamics simulations. It is shown that a small amount of structural change, i.e. 1 % of lattice expansion, can induce a dramatic enhancement of lithium-ion diffusivity. Detailed analyses indicate that lithium-ion diffusion in the perovskite structure is very sensitive to the octahedral tilting. Based on the findings, possible approaches to further improve the lithium-ion conductivity are suggested.

References:

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