Electronic Structure and Electrochemical Properties of Garnet Type $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ Solid Electrolytes Doped with Ta and Nb

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Received: 8 November 2021 / Accepted: 8 December 2021 / Published: 2 February 2022

The migration of lithium ions in the solid electrolyte is the main limiting factor that determines the rate and cycle performance of lithium-ion batteries. Based on the first-principles method of DFT, the electronic structure of $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ doped with Ta and Nb is studied, and the structure optimization, energy band, density of states and population analysis are carried out. The calculation results show that the lowest total energy of Ta doping system is 47307.28eV, and the structure is more stable. Due to the effect of different orbit electrons of Ta, Nb, the energy required for electron or hole transition is reduced, and the conductivity is improved, and the Ta doping is low to 2.74eV, which is more advantageous to improve the conductivity. The population analysis shows that the Ta-O bond length of the $\text{Li}_7\text{La}_3\text{Zr}_{1.75}\text{Ta}_{0.25}\text{O}_{12}$ system is 2.1717Å, and the stability is better. The Li-Li bond length of the Ta doping system is the shortest, which is beneficial to increase the Li vacancy concentration and thus improve the electrical conductivity.

Keywords: $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$; First principle; Electronic structure; Ionic conductivity

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