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Computational Modelling Study of Structure and Stoichiometry of Ta Doped Tetragonal Li7La3Zr2O12 Oxide Garnet for Solid State Batteries.

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Due to the outstanding chemical stability against high voltage electrode, the oxide garnet with tetragonal structure Li7La3Zr2O12 (LLZO), is one of the most promising solid-state electrolytes for li-ion batteries. However, it has low ionic conductivity (˜10-6 S.cm-1) at room temperature, which limits its practical application. Doping with a supervalent cation such as Ta on the Zr site of LLZO is an effective way to improve Li+ conductivity and further stabilize the tetragonal phase. To this end, the fundamental aspects regarding stability of most stable structural configuration of Ta-doped LLZO structures are still not entirely clear.
In this study, we have combined the first-principle calculations within the generalised gradient approximation (GGA) by determining the structural and thermodynamic properties of pure and doped t-LLZO for high ionic conductivity. The negative energy of formation in pure t-LLZO shows that the structure is thermodynamically stable. We further employed the substitutional search (SS) module to identify all possible structures and provide a better understanding of doped supervalent cation Ta on the octahedral 16c Zr site of LLZO. The substitutional search was used to replace a fraction of Zr atoms with Ta atoms, so that it can enable excess Li to occupy the disordered octahedral sites (occupied by Zr atoms), which could facilitate better li-ion transport and increase ionic conductivity. Furthermore, the substitutional search generated 3 new multi-component structures (monoclinic Li28La12Zr7TaO48, orthorhombic Li14La6Zr3TaO24 and triclinic Li28La12Zr7TaO48) of Ta doped LLZO. The calculated lattice parameters of doped LLZO are smaller than that of pure t-LLZO. The results show that the distance between Li-Li in doped Ta-LLZO is smaller than in pure t-LLZO, which indicates that the smaller the difference between the dopant ionic radius and the critical dopant radius, the higher the conductivity. Therefore, the structural properties of tantalum-doped structures are shown to improve, due to the smooth decrease in calculated lattice parameters. Hence, it is important to understand the stability of Ta doped LLZO for the development of all solid-state Li batteries.

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Yes

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MSc

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