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Comparative Analysis of Reactivity of Al and Ga Doped Garnet Solid State Electrolyte at the Interface with Li Metal

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The lithium garnet Li7La3Zr2O12 (LLZO) is a leading solid state electrolyte candidate for an All-solid-state battery (ASSB) containing Li as anode because of its high ionic conductivity, high toughness and wide electrochemical stability. Undoped LLZO exists in two polymorphic phases; a low conductivity tetragonal phase and a high conductivity cubic phase. Dopants in LLZO play a critical role in determining Li/LLZO interface stability and influencing the overall performance of an ASSB. In this work, we explore the differences between Al- and Ga-doped LLZO when interfaced with Li metal, using a combination of several techniques. We show that formation of Li metal interface with Ga-doped LLZO leads to a propensity of Ga to move from LLZO and form Ga-Li alloy layers, resulting in loss of dopant and associated changes in structure and electrochemical behavior not present in Al-doped LLZO. X-ray photoelectron spectroscopy with in situ lithium deposition shows the formation of a Ga-Li alloy at the interface, while Al is shown to be stable to reduction by lithium metal. Through neutron diffraction we observe that doping of LLZO with Ga results in complete transformation of the cubic phase to tetragonal phase when in contact with lithium metal, and although some performance characteristics may be enhanced through Ga doping, the materials stability is compromised significantly compared to Al doping.

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