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Challenges at the Li Metal/solid electrolyte interfaces for solid-state lithium batteries

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Both LLZO and LATP are leading solid state electrolyte candidates because of their excellent characteristics, such as high ionic conductivity and wide electrochemical stability windows. However, their interfaces with Li metal anodes face stability challenges which hinders their application in solid-state lithium batteries.
In case of LLZO, we explore the differences between Al- and Ga-doped LLZO when interfaced with Li metal, and 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. Neutron diffraction reveals that doping of LLZO with Ga results in complete transformation of the cubic phase to tetragonal phase when in contact with lithium metal.
In case of LATP, we overcome the chemical instability of LATP against Li, using a ultrathin PSiO polymer, which serves as multifunctional protection layer to enhance the interfacial stability between LATP and Li. It effectively blocks the direct contact between Li and LATP, regulates the homogeneous Li+ flux at the interface, promotes the intimate contact between PSiO and Li0 by forming Si-O-Li bonds, and generates an LiF-enriched interphase. As a result, it enables superior rate capability and cycling stability. Neutron depth profiling proves capability to measure the thickness of such a ultrathin layer.

Primary author: Dr PAUL, Neelima (Technical University of Munich, Heinz Maier-Leibnitz Zentrum (MLZ))

Presenter: Dr PAUL, Neelima (Technical University of Munich, Heinz Maier-Leibnitz Zentrum (MLZ))

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