Recent Advances on Garnet LLZO Solid Electrolytes for Lithium Batteries

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Abstract: Variants of lithium lanthanum zirconium oxide (LLZO are among the most promising candidates for use as electrolytes in solid-state lithium batteries, based on high conductivities and apparent resistance to reduction by lithium metal. Difficulties in processing and high interfacial resistances at lithium electrodes are among the challenges facing successful deployment of these interesting materials in electrochemical cells. In our laboratory, we have successfully prepared dense Al-substituted LLZO samples and characterized them using synchrotron and other advanced techniques. The main culprit involved in the high interfacial resistance is Li_2CO_3 on pellet surfaces, which can either be left over from processing or formed during even very brief periods of air exposure. The degree of air sensitivity is a function not only of the chemistry but also of the microstructure, so that it is possible to design materials with Li_2CO_3-free surfaces that exhibit low area-specific resistances in electrochemical cells. We have also employed a recently developed technique, intermittent contact alternating current scanning electrochemical microscopy (ic-ac-SECM), to characterize the topography and impedance of LLZO pellets, allowing direct visualization of grain and grain boundary resistances.