LLZO Ceramic Electrolytes: A Path Forward to Solid-State Batteries?

The appeal of solid-state batteries is clear; there are no flammable liquid components to leak or catch fire, and the possibility of using lithium anodes suggests that higher energy density than currently obtainable with Li-ion configurations should be possible. The emergence of garnet-structured electrolytes based on $Li_7La_3Zr_2O_{12}$ (LLZO) are particularly attractive for such devices because of their high conductivities and apparent stability against lithium. Still, there are many challenges associated with LLZO, including difficulties in densifying the materials and high interfacial resistances at the lithium electrodes. Work in our group has been directed towards overcoming these obstacles. Classical ceramics processing techniques have been used to produce samples up to 94% dense, as thin as 200 microns, with excellent control over grain size and grain boundary chemistry. Synchrotron techniques such as soft x-ray absorption spectroscopy (XAS) and x-ray photoemission spectroscopy (XPS) reveal that the main culprit leading to high interfacial resistance at lithium electrodes is lithium carbonate, either present as a by-product of synthesis or produced upon exposure to air. Polishing in inert atmosphere removes this impurity and lowers the interfacial impedance, but is not practical commercially. Through grain boundary engineering, it is possible to substantially reduce the sensitivity of samples to air exposure, greatly reducing the tendency towards formation of lithium carbonate and resulting in much lower interfacial impedances at lithium electrodes.