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Formation of Nanosized Cubic Lithium Lanthanum Zirconate (LZZO) for High-Performance Lithium-Ion Batteries

Lithium lanthanum zirconate (Li7La3Zr2O12, LLZO) is a highly promising solid electrolyte for lithium-ion batteries in that it possesses high conductivity (σ ~104 S/cm) and chemical stability against elemental Li, potentially allowing pure Li anodes and greatly increasing energy capacity of the batteries. LLZO may have a cubic or tetragonal crystal structure. The cubic phase has high lithium-ion (Li-ion) conductivity, but is only thermodynamically favorable at high temperatures (>600° C). The tetragonal phase is stable at room temperature, but has poor ionic conductivity. Currently, most LLZO is synthesized through solid state reactions, typically requiring high temperatures, repeated heat treatments, ball milling, or other mixing methods, as well as carefully controlled addition of extrinsic dopants to stabilize the cubic phase at room temperature. Dopants used in these processes, however, often form impurity phases, particularly at interfaces and grain boundaries, resulting in reduced overall conductivity.

Researchers at Arizona State University have developed a method to synthesize nanosized cubic LLZO by forming a solution that includes an organic compound and compounds of lithium, lanthanum, and zirconium. The solution is dried to yield a solid, and the solid is heated in the presence of oxygen to pyrolyze the organic compound to yield a product including nanosized cubic LLZO.

Reducing the crystallite size of LLZO below a certain threshold (e.g., to nanometric dimensions) can stabilize the cubic phase at low temperatures without the use of extrinsic dopants. In addition to stabilization of the cubic crystalline phase, nanosized ceramics have some particular advantages. For example, the onset temperature of sintering is substantially lower for nanosized ceramic particles due to the relatively higher surface energy of nanoparticles compared to bulk particles. Further, smaller grain size in dense LLZO ceramics yields higher overall conductivity since Li-ion conduction is higher along grain boundaries.

This invention is covered by U.S. Pat. No. 10,858,263 and U.S. Pat. No. 11,597,658.

Potential Applications:

- Solid electrolyte for lithium-ion batteries

Benefits and Advantages:

- Rapid, simple, and inexpensive method to synthesize nanocrystals of LLZO
- Bypasses need for extrinsic dopants
- May significantly increase safety and energy capacity of Li-ion batteries