Tracking Lithium Transport Pathways in Solid Electrolytes for Batteries

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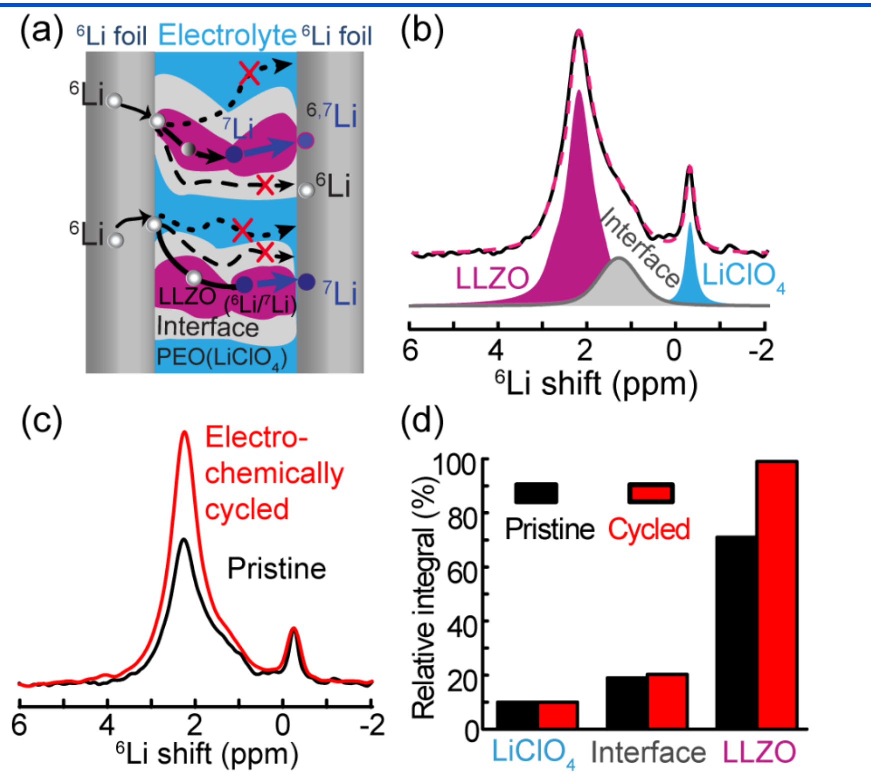
2. **Funding Grants:** G.S. Boebinger (NSF DMR-1157490); Y.-Y. Hu (NSF DMR 1508404)

Most commercially-available rechargeable lithium batteries contain liquid-based electrolytes and, thus, face several challenges, including safety issues, energy density, and cost. New all-solid-state batteries are addressing these challenges, yet fundamental research is necessary at this early stage to help realize their potential.

By developing a new method to track lithium (Li) transport pathways in solid electrolytes, this work develops a fundamental understanding that facilitates improvement of Li ion conductivity for future technologies. Experts in electrolyte synthesis produced our high-performance solid electrolytes. *These new Lithium-6 → Lithium-7 isotope replacement experiments reveal that Li transport pathways vary significantly, depending on the composition and structure of the solid electrolytes.* The experiments were performed in the MagLab's 11.7-T magnet with a 2.5 mm magic-angle-spinning NMR probe and an *in operando* NMR probe designed by the MagLab for battery research.

The in operando NMR measurements provide real-time tracking of Li ion transport under real battery operating conditions. The gained knowledge will help guide battery materials design and device fabrication for future high-performance, all-solid-state rechargeable batteries.



(a) A symmetric solid-state battery was made with two ^{6}Li metal electrodes and a polymer (PEO)-ceramic (LLZO) composite electrolyte. (b) Three different Li environments in the electrolyte were identified with high-resolution ^{6}Li NMR. A biased electric potential drives ^{6}Li ions from ^{6}Li metal into the electrolyte to replace ^{7}Li. An increase in the ^{6}Li amount of the LLZO phase after the ^{6}Li → ^{7}Li replacement suggests Li transports through a percolated network of LLZO ceramic particles, instead of the polymer or along interfaces.

Facilities: NMR/MRI Facility at the NHMFL/FSU

Citation: Zheng, J.; Tang, M. and Hu, Y.Y., *Lithium Ion Pathway within Li7 La3 Zr2 O12 -Polyethylene Oxide Composite Electrolytes.*, **Angew. Chem. Int. Ed.**, **55** (40), 12538-12542 (2016)