Lithium Nucleation of Anode-Free Solid-State Batteries with a Dry Compressible Interlayer

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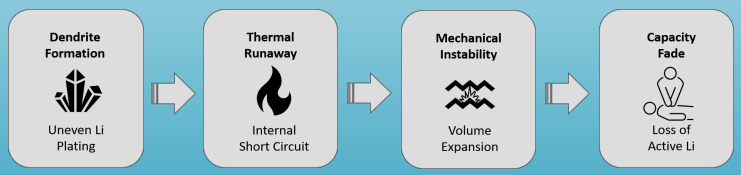
Anode-free lithium batteries offer promising advantages, including increased energy density and the ability to address common mechanistic failures within the cell, thus increasing safety. One way that can make this possible is to control lithium nucleation. This could be achieved by reducing the overpotential required and implanting lithophilic nucleation sites in an anodic interlayer to aid in lithium ion transport and plating. The concept has been utilized in solid-state battery systems where electrolytes are solids with further improved safety and structural longevity. In this presentation, we discuss the use of a silver-holey graphene-based anodic interlayer that can be fabricated via direct dry compression without the use of solvent or binder. The addition of silver to the holey graphene matrix creates a route to lithium plating via a lower-energy intermediate. This idea is supported by the presence of a lithium-silver alloy that forms during the activation step. It is understood that the embedded silver acts as a nucleation site for lithium ions, thereby assisting in even plating. This control, combined with the added cushion of the holey graphene itself, can help reduce dendrite formation, ultimately increasing the safety and lifetime of the solid-state batteries.

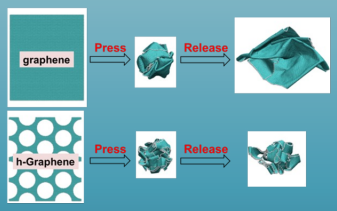
Problem- Uncontrolled Li Deposition

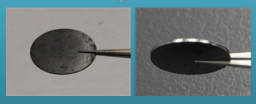


Controlling Li Formation

- ❑ Decreases Dendritic Growth
- ❑ Reduces Short Circuit Risks
- ❑ Minimizes Mechanical Stress
- ❑ Preserves Life Cycle

Proposed Solution- Tunable hG Interlayer





- ❑ The presence of nanometer holes on graphene sheet allows the facile escape of air molecules, enabling the high solvent-free compressibility of holey graphene.

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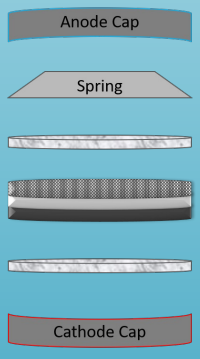
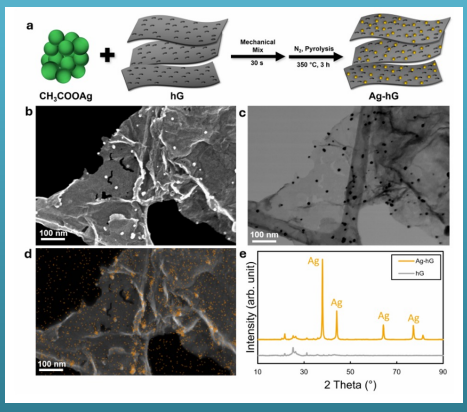
Battery Architecture



- ❑ **Anode Place Holder-** Silver Holey Graphene
- ❑ **Electrolyte-** LPSC (Li_6PS_5Cl)
- ❑ **Cathode-** NMC (Nickel Manganese Cobalt)



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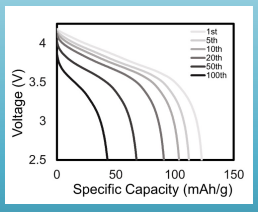
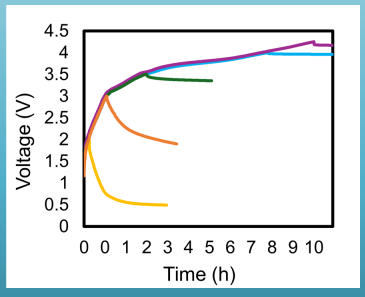
Cycling Performance



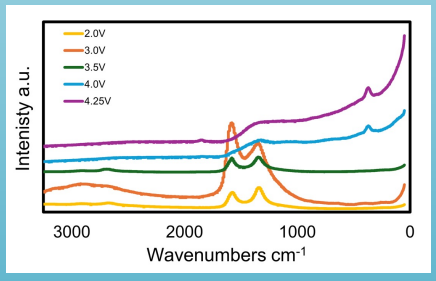
- ❑ **Cell architecture**: 64mg NMC Cathode at 30mg/cm^2 active mass loading, 90mg LPSC, 8mg 1:3 Ag-hG as anode housing.
- ❑ **Cycling conditions:** First charge at C/10 (1.062mA), then cycled at C/5 (2.124mA). 100C

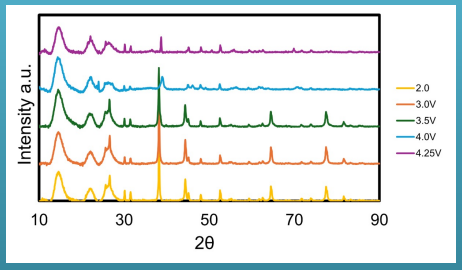
Voltage Monitoring



Testing voltage relaxation behavior at various points of charging voltage suggests SEI formation at ~3.5V.

Mechanistic Insight





Raman spectroscopy and optical photographs suggested Li-Ag alloy forms when the cell was charged to ~3V. X-Ray diffraction (XRD) supports Li-Ag alloy formation

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