Improving Cell-Level Specific Energy for All-Solid-State Lithium Sulfur Batteries

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Why is NASA Interested in Solid-State Batteries?

UAM: Urban Air Mobility
eVTOL: electric Vertical Take-Off and Landing
SOA: State-of-the-Art
R&D: Research & Development
Why is NASA Interested in Solid-State Batteries?

SABERS: Solid-state Architecture Batteries for Enhanced Rechargeability and Safety

Combination of unique materials technologies to achieve performance goals
Dry-Pressed Electrodes Enabled by Holey Graphene

Li ion can conduct through the thickness of holey graphene (hG) – as long as the holes are at least 25% in size of the solid-state electrolyte particles.

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Composite Solid-State Cathode Powder

- Active material: S
- Solid electrolyte (SE): Li$_6$PS$_5$Cl (LPSC)
- Carbon: CB (carbon black) vs hG (holey graphene)

CB: Super C45
All-Solid-State S Cathodes

- hG: holey graphene
- CB: carbon black

Diagram:
- "Coating" and "Scaffolding" processes
- Sulfur, LPSC, CB, and hG in the mixture
- Cold Press steps to create S-LPSC-hG and S-LPSC-CB cathodes
Both composites are compressible to form robust cathode/SE bilayer discs

LPSC glass electrolyte serves as binder

hG as “cold pressable hosts” is not an obvious advantage…?
Ionic conductivity of solid electrolytes from different fabrication pressure can be described using a particle dynamics model.
Dry-Pressed Cathode/SE Bilayers
Cathode Microstructures
The use of hG provides much lower impedance, especially in low frequency region.
The use of hG allows one magnitude higher Li ion diffusion through the cathode.
All-Solid-State Li-S Cell Performance

- Li metal anode
- No additional stack pressure

Room Temperature
- 12 mg/cm²

60°C
- 5 mg/cm²
Strategies toward High S Utilization

Increase Operation Temperature

- 60°C
- S mass loading = 5 mg/cm²

S Melt Infiltration

- 60°C
- S mass loading = 5 mg/cm²
- 1319 mAh/gₜ (79% utilization)
- 913 mAh/gₜ (55% utilization)
A Design-of-Experiment (DOE) study
20 unique compositions
- S: 10 – 50%
- hG₁+hG₂: 5-20%; hG₁: 0-15%; hG₂: 0-20%
- SE₁+SE₂: 30-85%; SE₁: 0-75%; SE₂: 0-70%
- No hG₁ = no melt infiltration
S Cathode Design Principles from DOE

- **High Discharge Capacity**
  - Low S content
  - High hG:S ratio in melt infiltration

- **Low Overpotential**
  - Low S content
  - Medium hG:S ratio in melt infiltration

- **Low impedance**
  - High scaffolding-step hG content
  - High coating-step SE content

- **High Li\(^+\) Diffusion Coefficient**
  - High hG:S ratio during melt infiltration
  - High coating-step SE content
Reducing SE Thickness in Dry-Press

\begin{itemize}
  \item 114 mg/cm\(^2\)
  \item 57 mg/cm\(^2\)
  \item 28 mg/cm\(^2\)
  \item 20 mg/cm\(^2\)
\end{itemize}

\begin{figure}
\centering
\includegraphics[width=\textwidth]{diagram}
\caption{Graph showing the relationship between LPSC Loading (mg cm\(^{-2}\)) and Thickness (μm). The line suggests that thickness increases linearly with LPSC loading.}
\end{figure}

\textbf{~1.45 g cm\(^{-3}\)}
Improved Energy Density

- Reduction in SE thickness + Increasing cathode S content with retained S utilization = **Improved Energy Density**

- All dry-pressed cathodes
- No additional stack pressure
- 0.032 mA/cm²

![Graph showing areal capacity vs. LPSC loading and voltage vs. specific capacity.](image-url)
Reliable Reductions of SE Thickness

Free-standing LPSC Thin Films
(~26 μm in thickness)

Tape-Casted LPSC Films
(~25 μm in thickness)

Future development and direction: 5-10 μm

Feasible? Reliable? Safe?
Solid-state S cathodes were prepared by **solvent-free pressing** a mixture of S, solid electrolyte, and carbon.

**Holey graphene** provides robust composite cathode architecture, thus enhanced electrochemical performance (in comparison to carbon black).

**High S utilization** was achieved at **high mass loading (> 5 mg/cm²)** in all-solid-state cells.

**Optimization** of all-solid-state S cathodes was achieved via **DOE studies**.

Cell-level (electrochemical) **energy density** was improved by **reducing solid electrolyte thickness and increased cathode S content**.
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