INL/CON-22-67294-Revision-0

Unlocking Failure Mechanisms and Improvement of Practical Li-S Pouch Cells Through in Operando Pressure Study

May 2022

Bin Li

Changing the World's Energy Future

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Bin Li

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Idaho National Laboratory
Idaho Falls, Idaho 83415

http://www.inl.gov

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U.S. Department of Energy
Under DOE Idaho Operations Office
Contract Battery 500 Consortium

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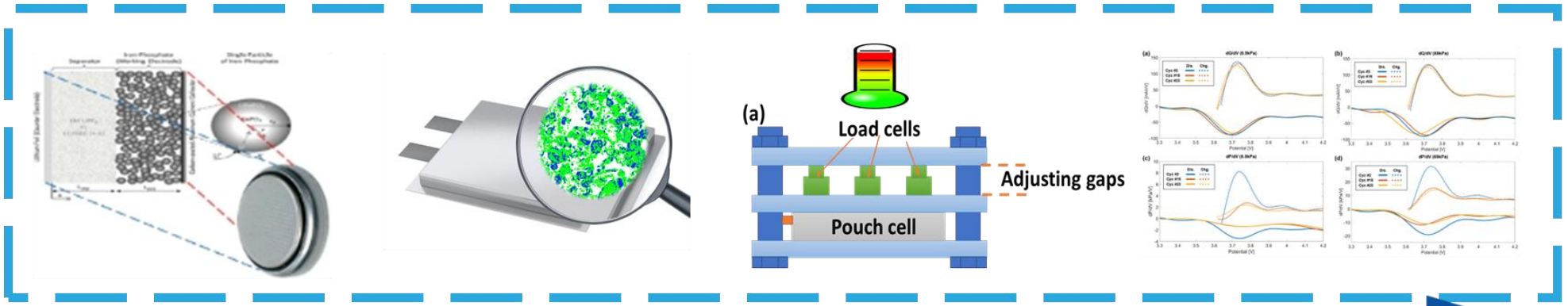
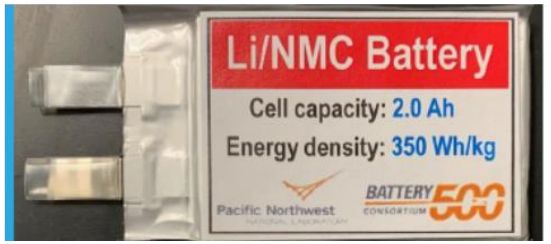
Background

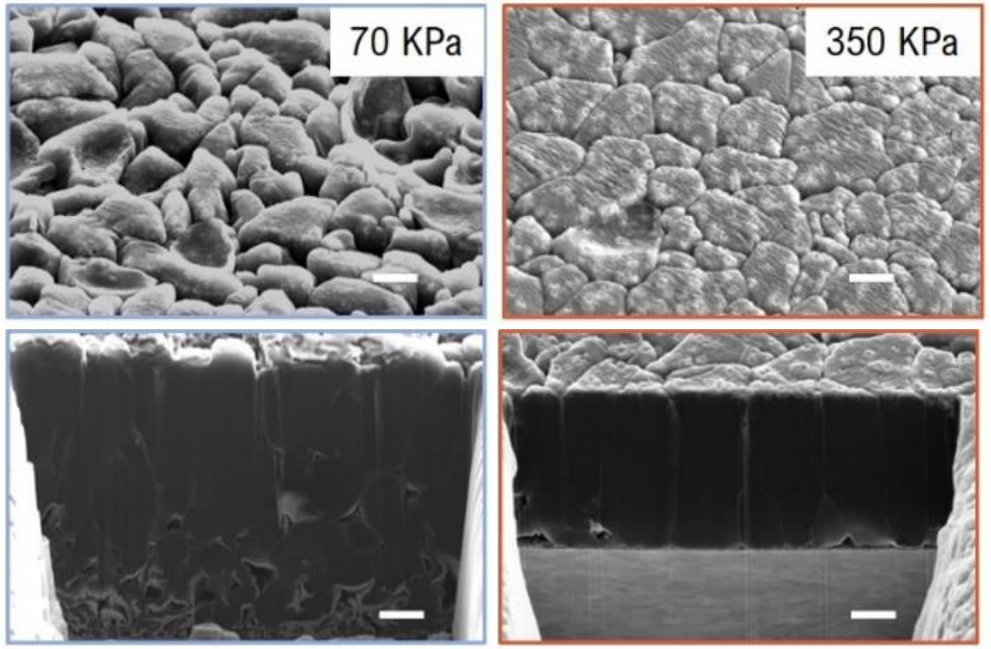
Degradation prevention based on pouch cells



Quantification, verification and validation across temporal and spatial domains

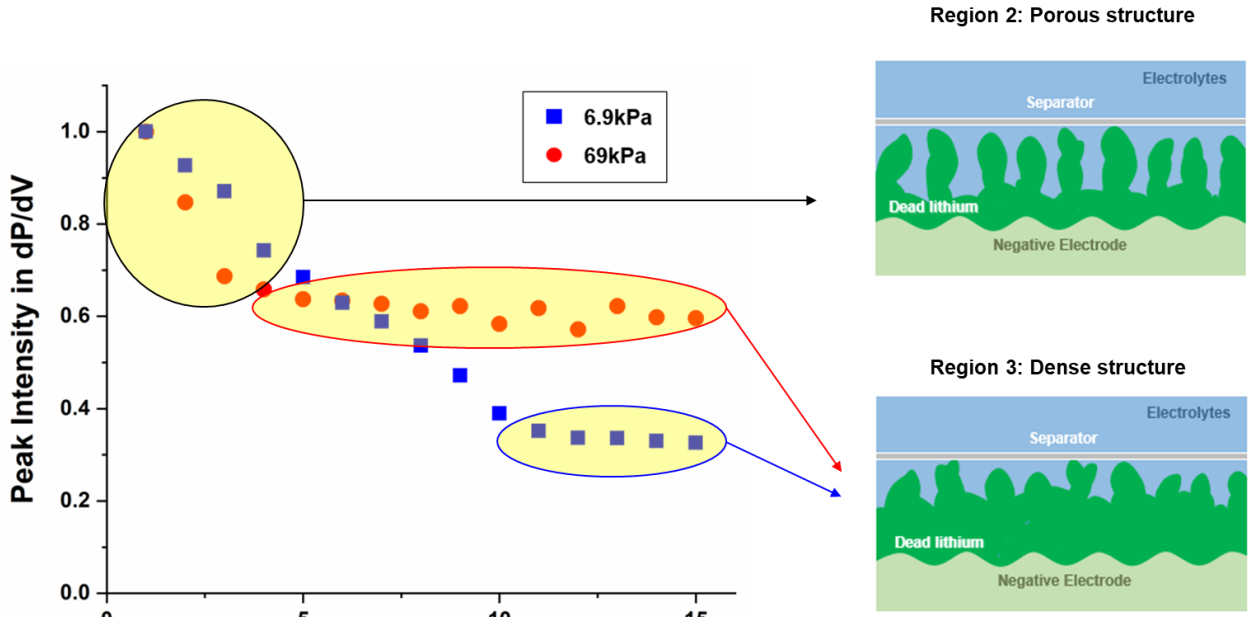


Previous Work on Pressure Effect



- Li morphology becomes denser and more uniform as pressure increase;
- At 350KPa, the Li is plated in a columnar structure.

INL Publications: Fang, et al. *Nature Energy* 6.10 (2021): 987-994.



- dP/dV can be used as a tool to diagnose the Li metal anode evolution in real time.

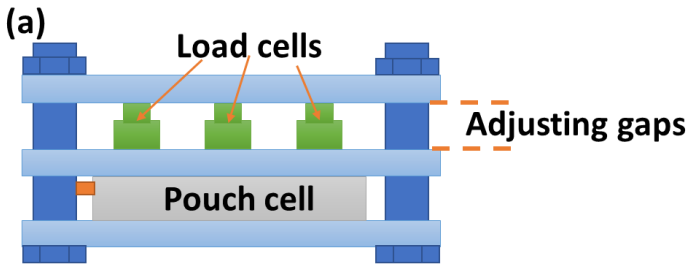
INL Publications: Kim, et al *Journal of Power Sources* 463 (2020): 228180

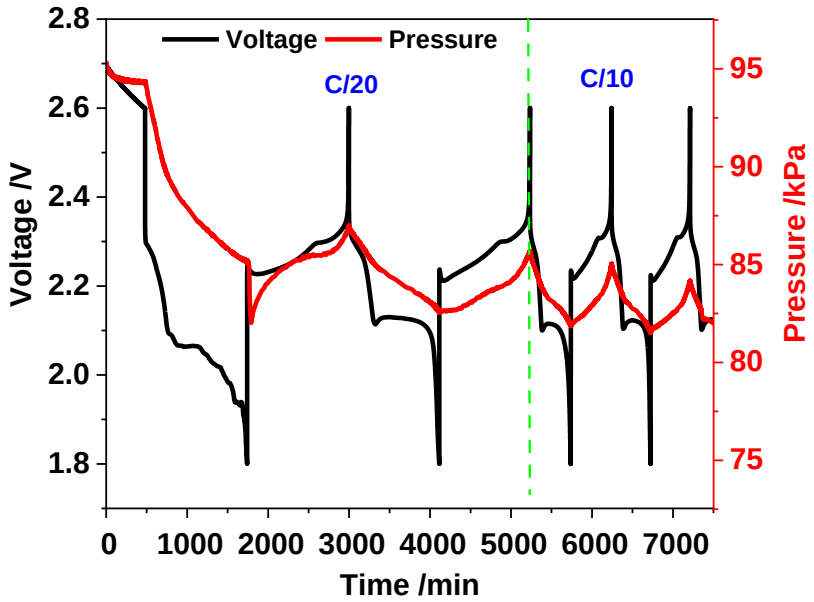
Li-S SLPC Study Through Operando Pressure Monitoring

- Wetting Issues
- Pressure Effect
- Cathode Structure (e.g. Porosity, Tortuosity)
- Real-time Diagnosis

INL Publications:
Chinnam et al, ***Advanced Energy Materials*** 12(7),2270027. Published Date: Dec. 2021

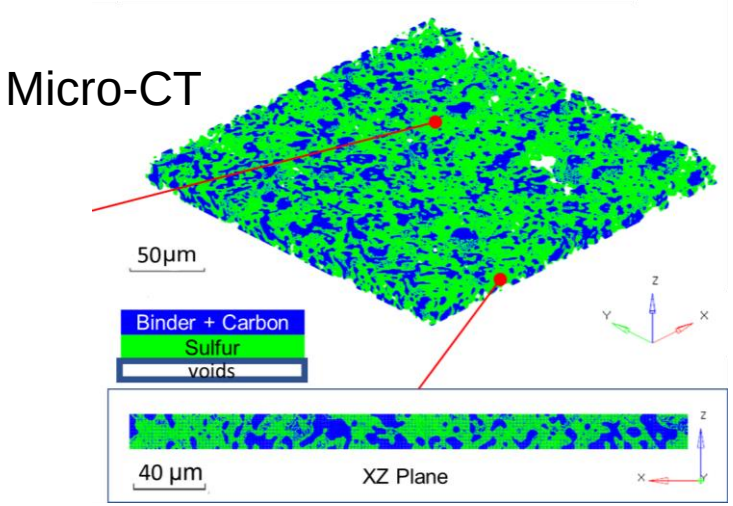
Pressure Set-Up





❖ Pressure variation is dominated by Li metal anode thickness change.

Cathode Structure Characterization



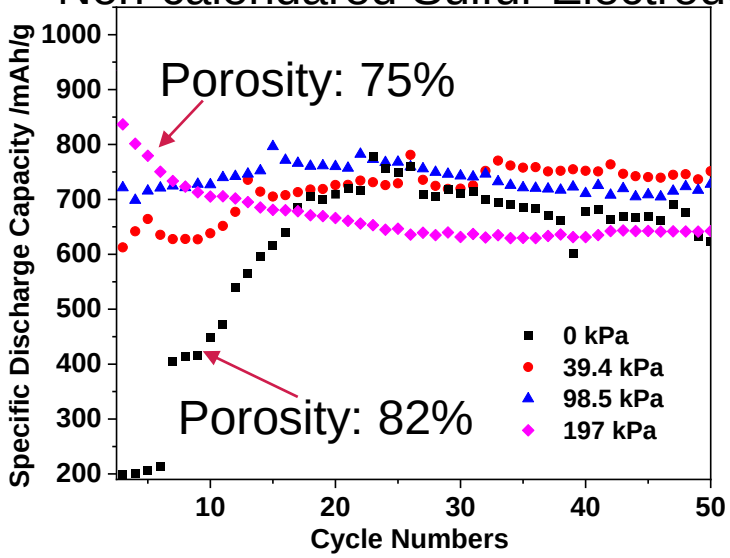
A higher Porosity/Tortuosity (ε/τ) ratio represents faster electrolyte (PS dissolved) diffusion through the cathode structure according to equation (1):

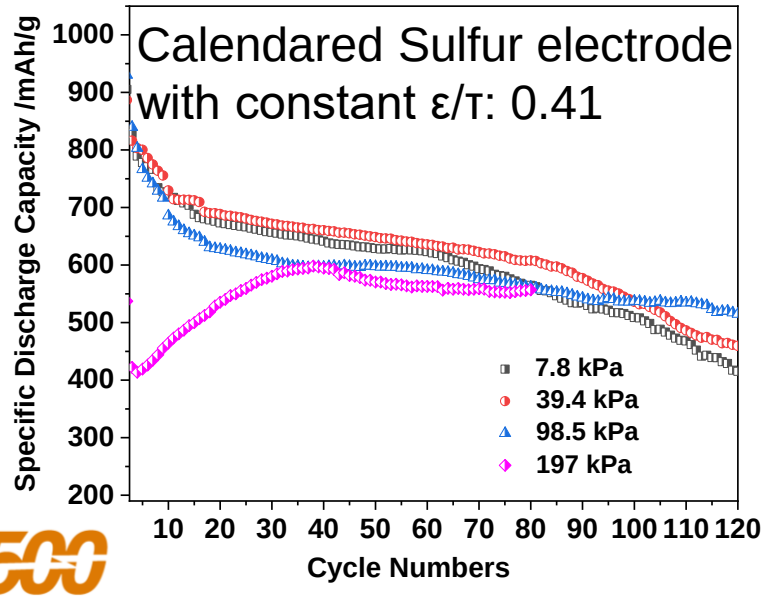
$$D_{eff} = \frac{D_{int} \times \varepsilon}{\tau \times A} \qquad (1)$$

❖ Tortuosity of sulfur cathodes can be characterized by micro-CT.

Wetting Issues

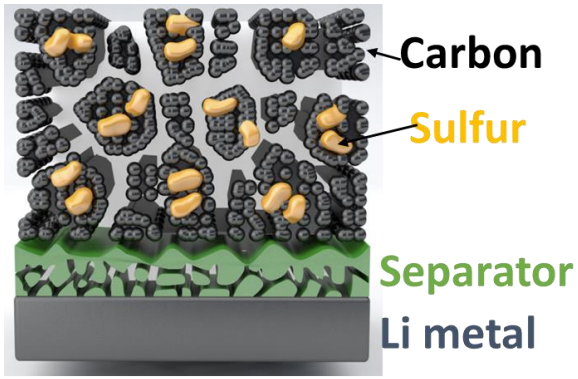
Non-calendared Sulfur Electrode



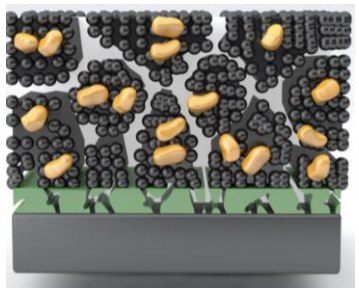


Highly Porous

Poor (low P)



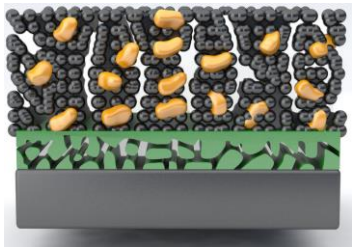
Good (high P)



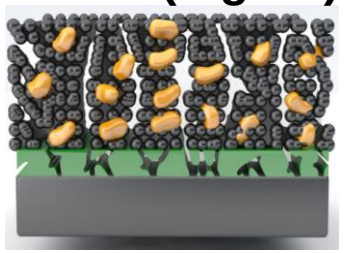
- External Pressure can change the structure of non-calendared cathode
- Larger porosity would cause initial capacity loss.

Slow Diffusion

Good (low P)



Poor (high P)

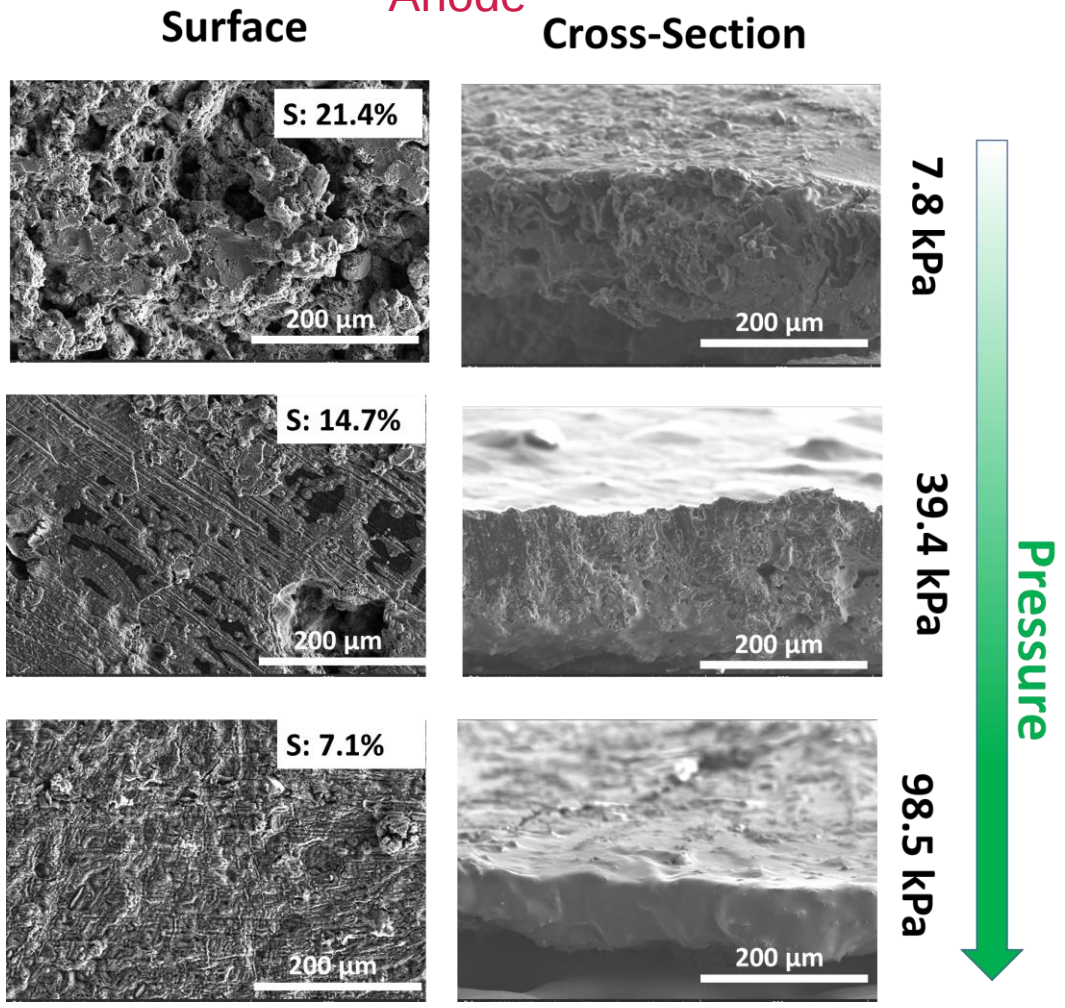


Compressed Separators

- Optimal pressure is required.
- The separator with higher mechanical property and stable porous structures is highly required

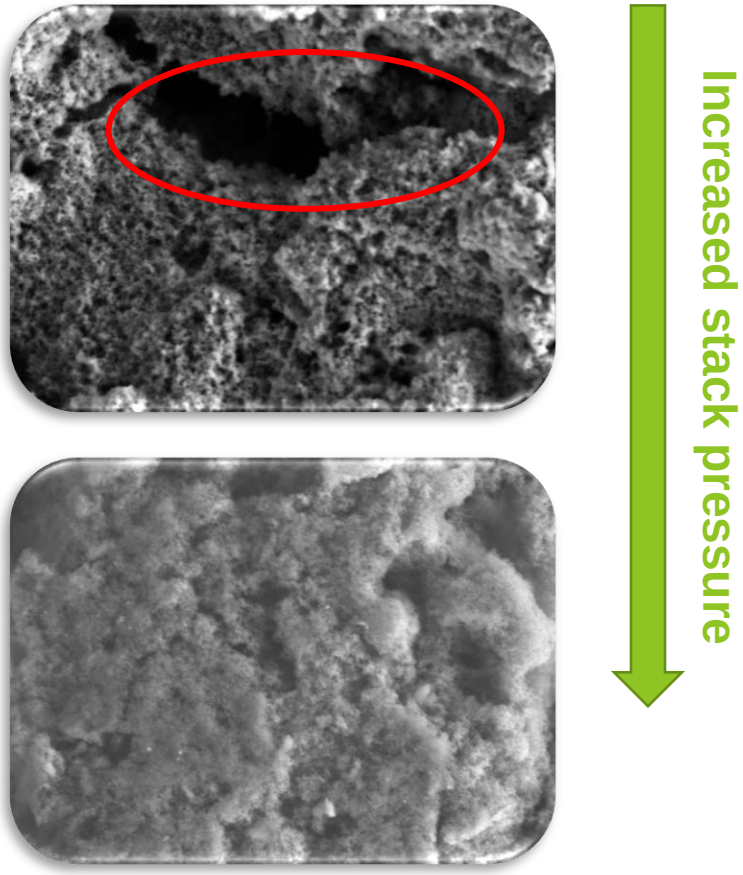
Pressure roles

Anode



- Higher pressure can promote uniform metallic Li growth
- Lower S content on the surface under higher pressure indicates less shuttle.

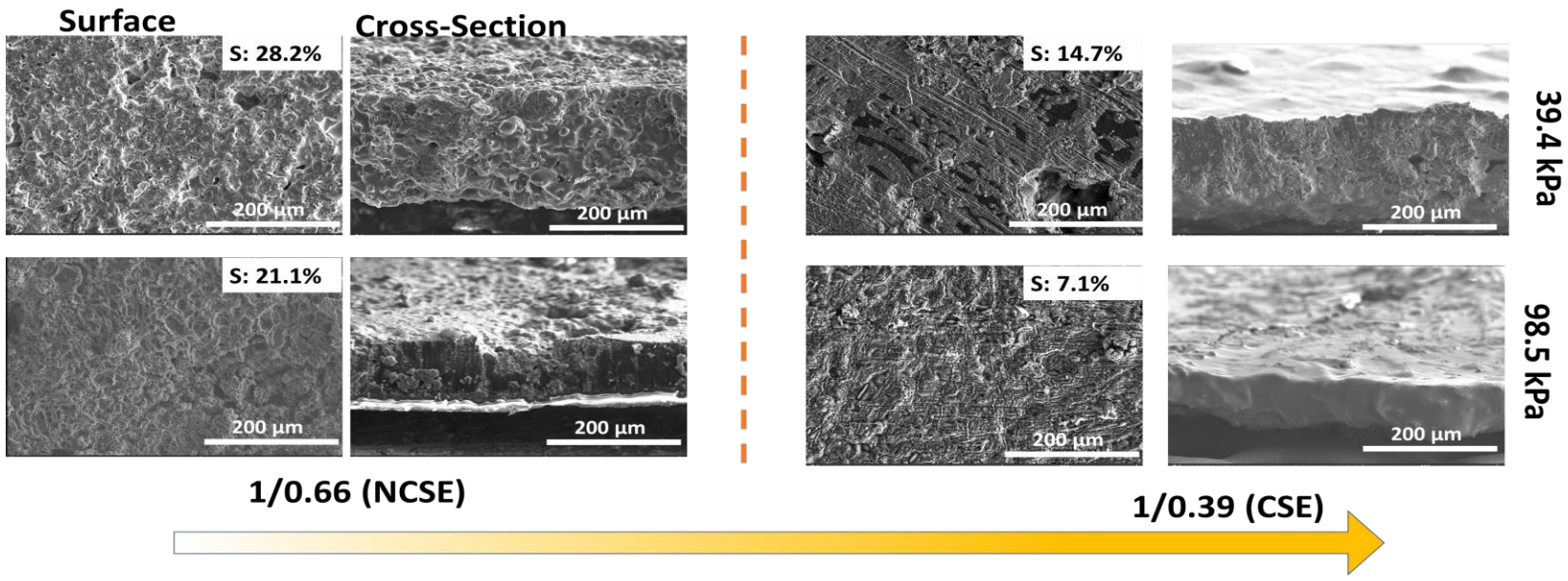
Cathode



- Higher pressure can suppress cracking at the cathode side, which would benefit the weight reduction of conductive agents and binders used in the sulfur cathodes.

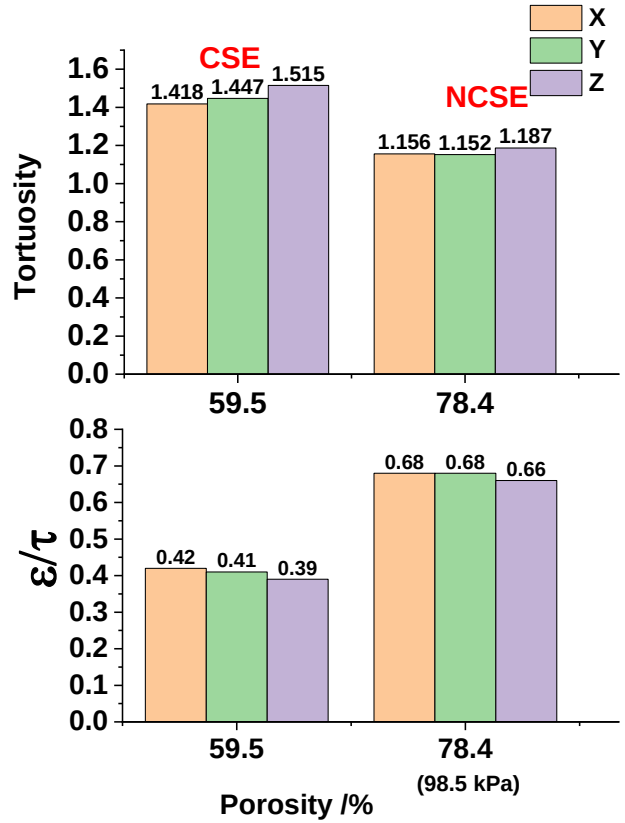
Sulfur cathode structure
------- Porosity (ε)/Tortuosity (τ)

CSE: Calendared Sulfur Electrode
NCSE: Non-Calendared Sulfur Electrode



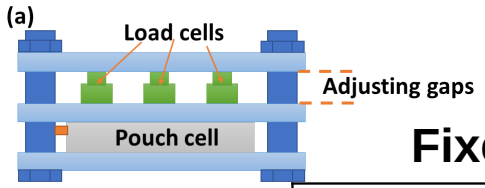
Porous and thick Li metal anode with high S content

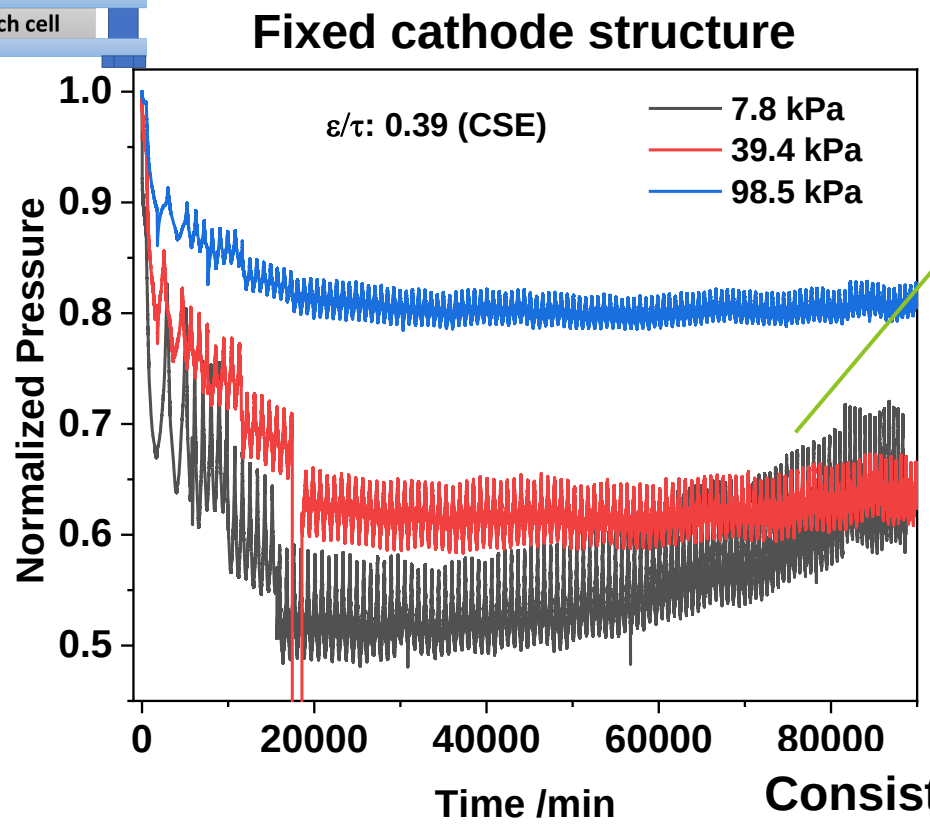
Dense and thin Li metal anode with low S content



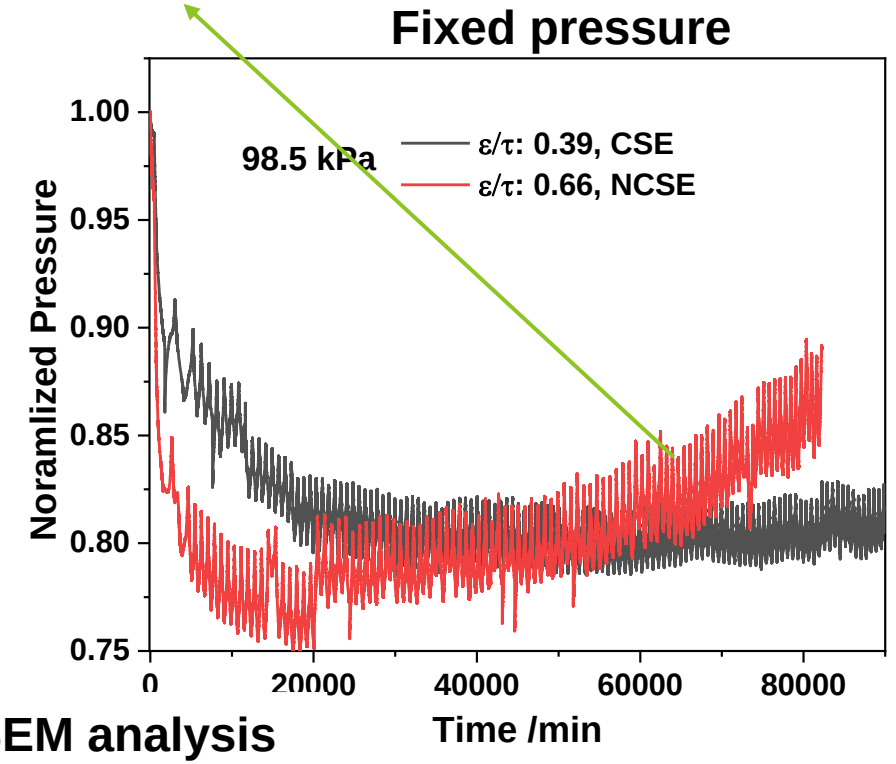
❖ Lower ε/τ of cathodes can reduce shuttle.

Li metal anode evolution by real-time monitoring pressures





Li metal thickness build up



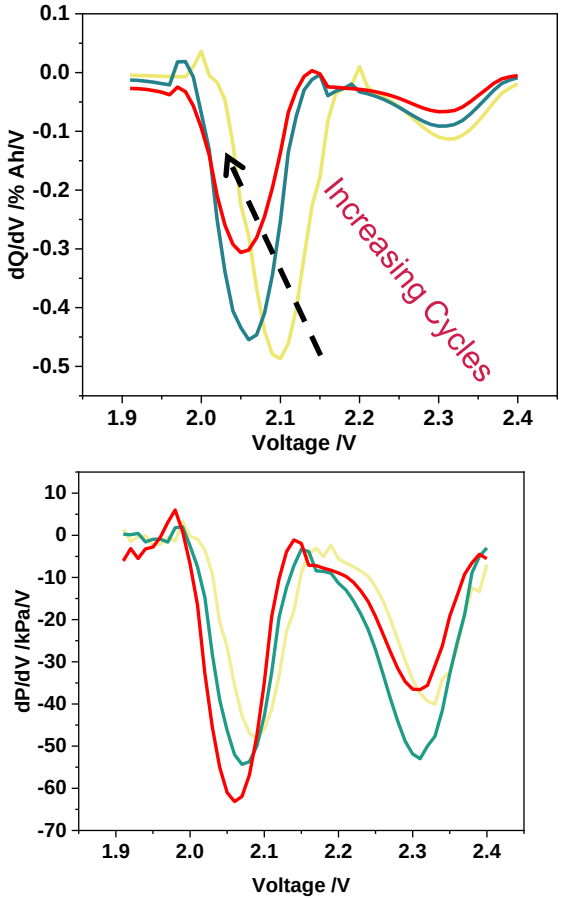
Consistent with SEM analysis

- Lower pressure: Anode Thickness build-up
- Higher ε/τ: Anode Thickness build-up

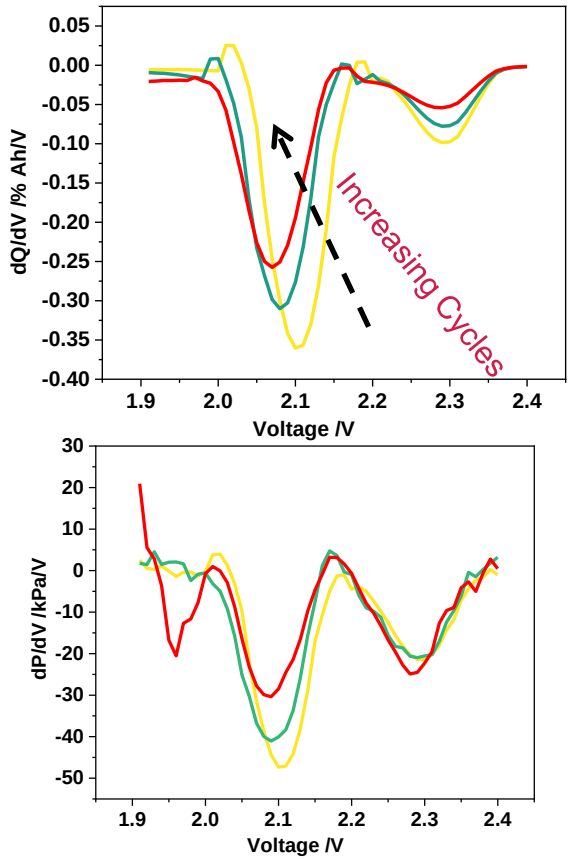
INL Publications:
Chinnam et al, ***Advanced Energy Materials*** 12(7),2270027. Published Date: Dec. 2021

Li metal anode evolution by real-time monitoring pressures

Porous (lower Pressure and larger ε/τ)



Dense (higher Pressure and lower ε/τ)

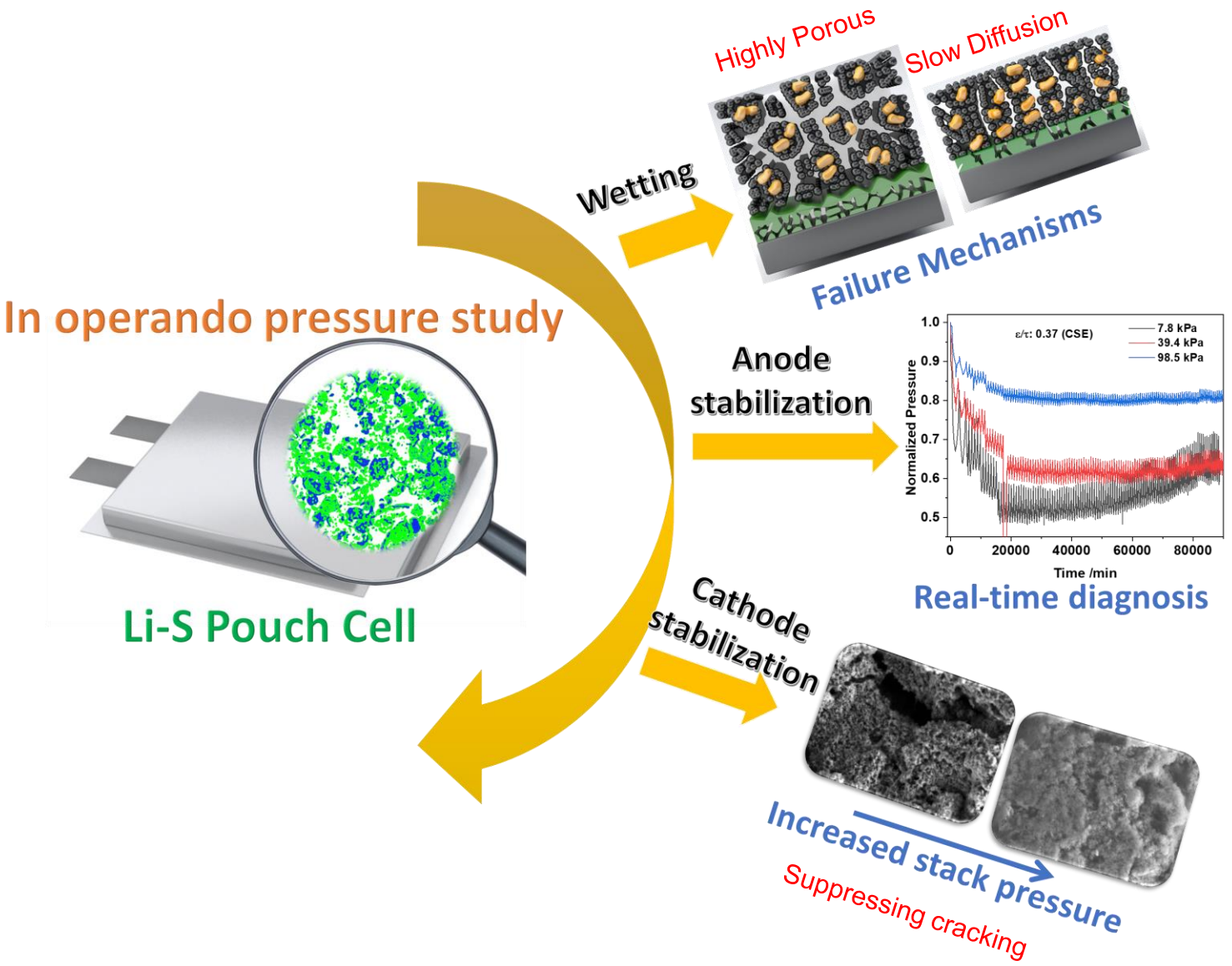


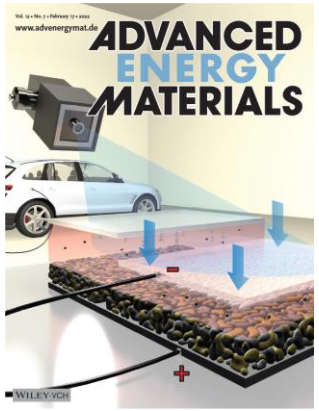
Uniform Li metal plating/stripping: the peak intensity of dP/dV over cycling is expected to follow the dQ/dV trend

Consistent with SEM analysis

- Normalized pressure and dP/dV verified as an effective tool for real-time diagnosis of Li-metal anode degradation (e.g., when significant degradation initiates; where the degradation is located by monitoring pressures at different locations across the large-scale cell) in Li-S pouch cells

Summary


Highly Porous
Slow Diffusion
Wetting
Failure Mechanisms
In operando pressure study
Li-S Pouch Cell
Anode stabilization
ε/τ: 0.37 (CSE)
7.8 kPa
39.4 kPa
98.5 kPa
Normalized Pressure
Time /min
Real-time diagnosis
Cathode stabilization
Increased stack pressure
Suppressing cracking


ADVANCED
ENERGY
MATERIALS

Acknowledgements

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- Battery500 Teams
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- INL team members: Parameswara R. Chinnam,, Lu Cai, Nikolaus L. Cordes, Sangwook Kim, Corey M. Efaw, Eric J. Dufek