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Enabling Rechargeable Conversion Cathodes by Modeling the Coupling Between Structure, Electrochemistry, & Mechanics

Scott A. Roberts, Jeffrey S. Horner

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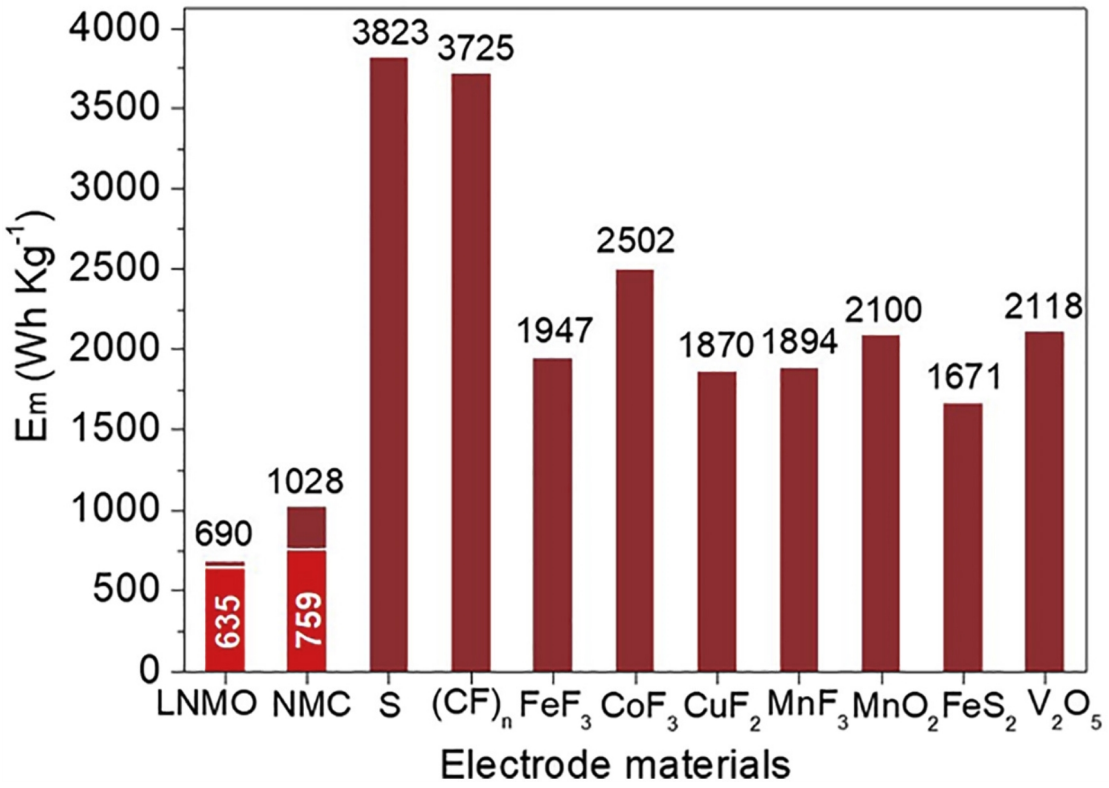
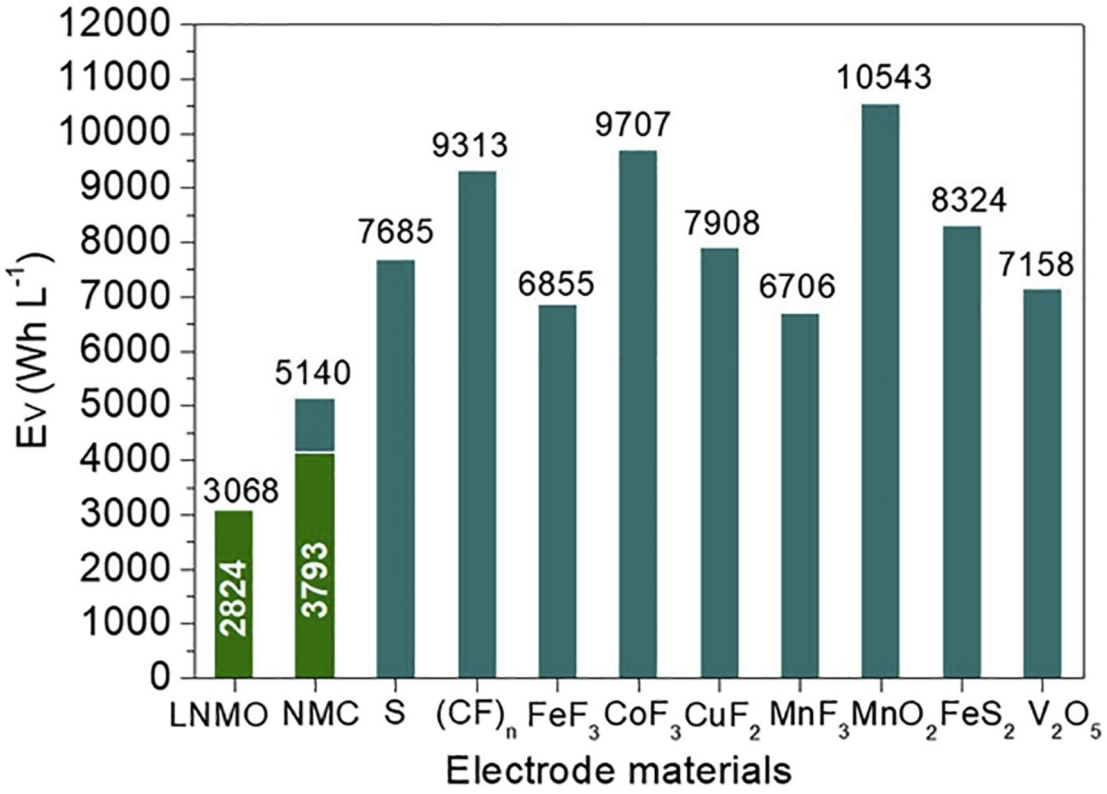
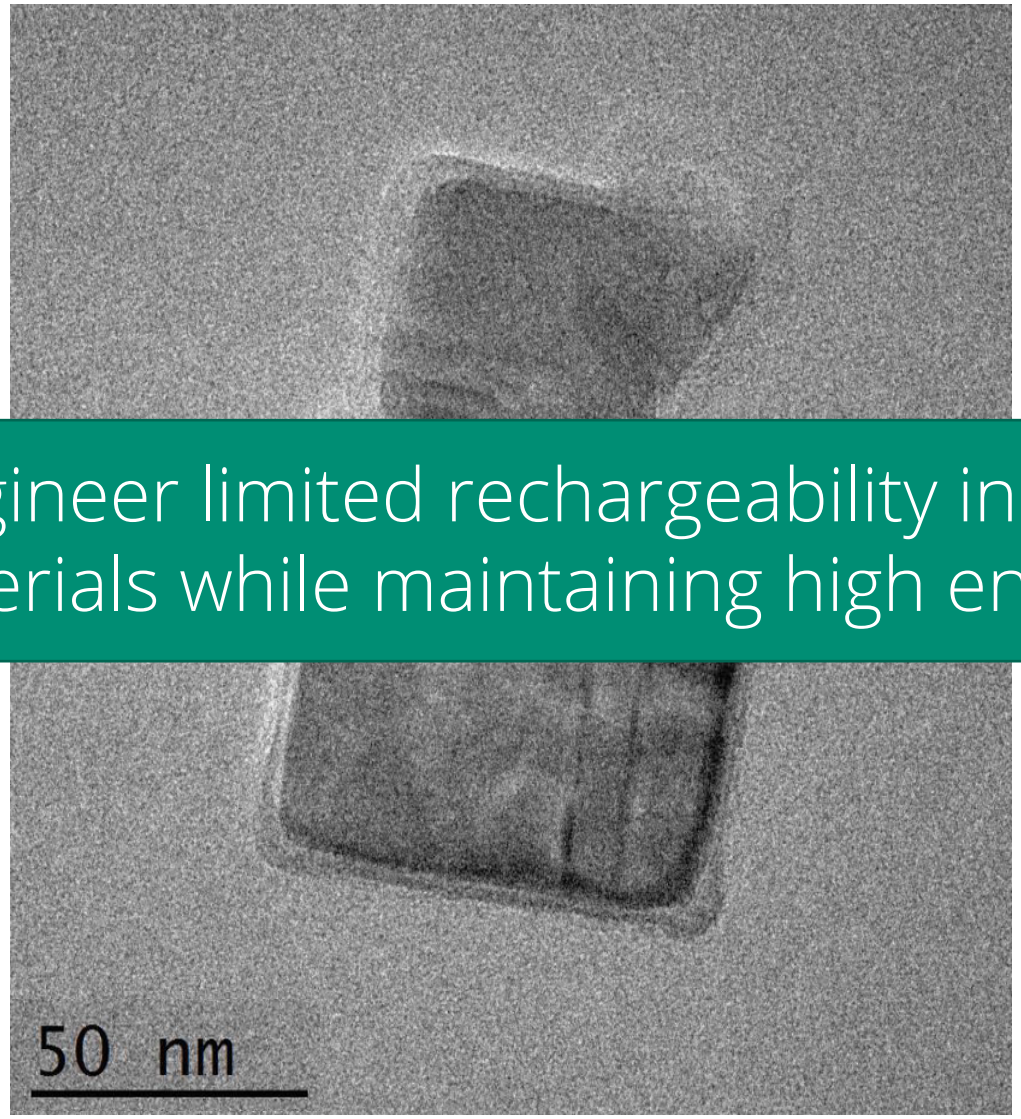
Conversion cathodes hold potential for higher energy densities





Wang et al., Joule (2019)

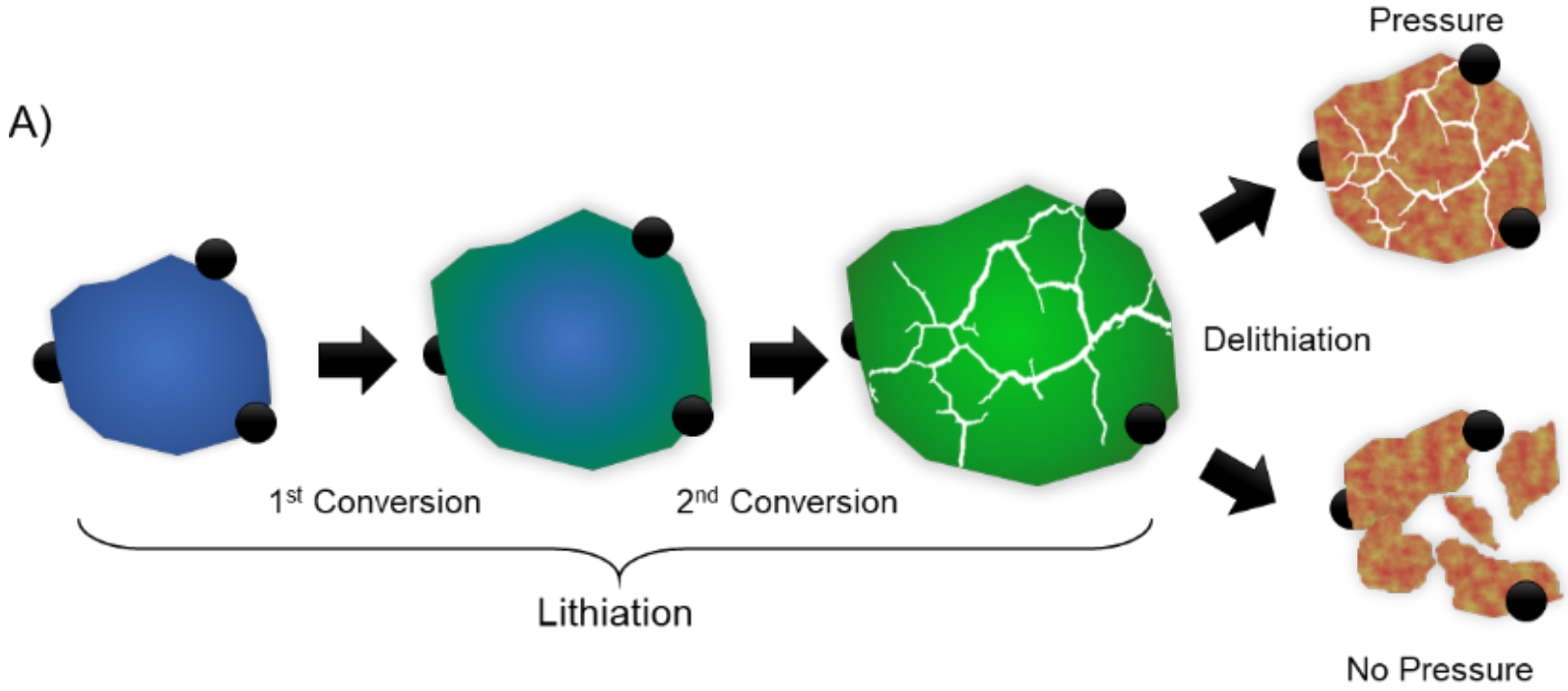
Mechanical degradation can limit conversion secondary cells



Boebinger et al, Joule (2018)

Can we engineer limited rechargeability in conversion cathode materials while maintaining high energy density?

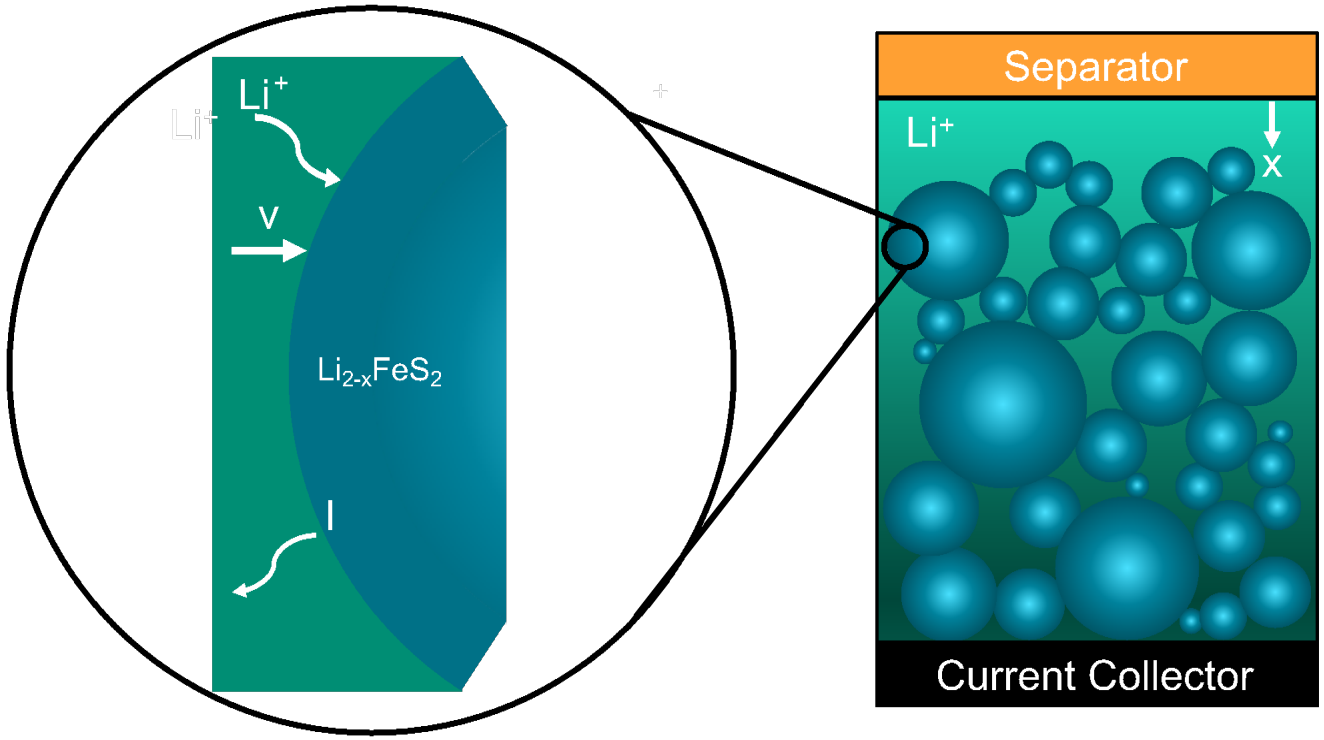
FeS_2 reaction pathways



Generalized Reaction Mechanism		
(1)	$FeS + S + x\,Li^+ + xe^- \rightleftharpoons FeS + x\,Li_2S$	Conversion
(2)	$FeS + x\,Li_2S + y\,Li^+ + y\,e^- \rightleftharpoons y\,LiFeS + x\,Li_2S$	Intercalation
(3)	$y\,LiFeS + x\,Li_2S + z\,Li^+ + z\,e^- \rightleftharpoons Fe^0 + 2\,Li_2S$	Conversion

Ashby et al, ACS AMI (2022), 10.1021/acsami.2c01021

Pseudo 2D model for conversion electrodes



Solid potential (V_s)

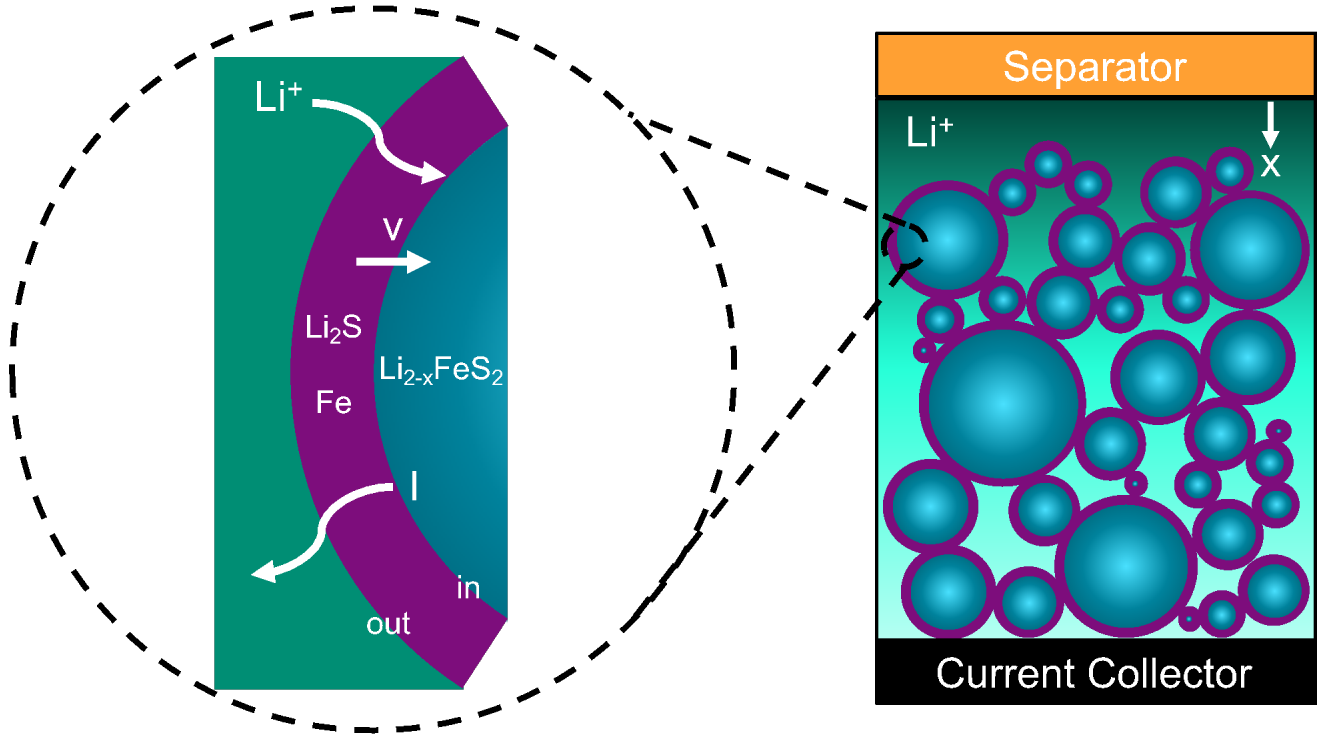
Solid Li concentration (C_{Li})

Electrolyte potential (V_l)

Electrolyte concentration (C_{Li^+})

Horner et al, J. Power Sources (2022), 10.1016/j.jpowsour.2022.231893

Pseudo 2D model for conversion electrodes



Solid potential (V_s)

Solid Li concentration (C_{Li})

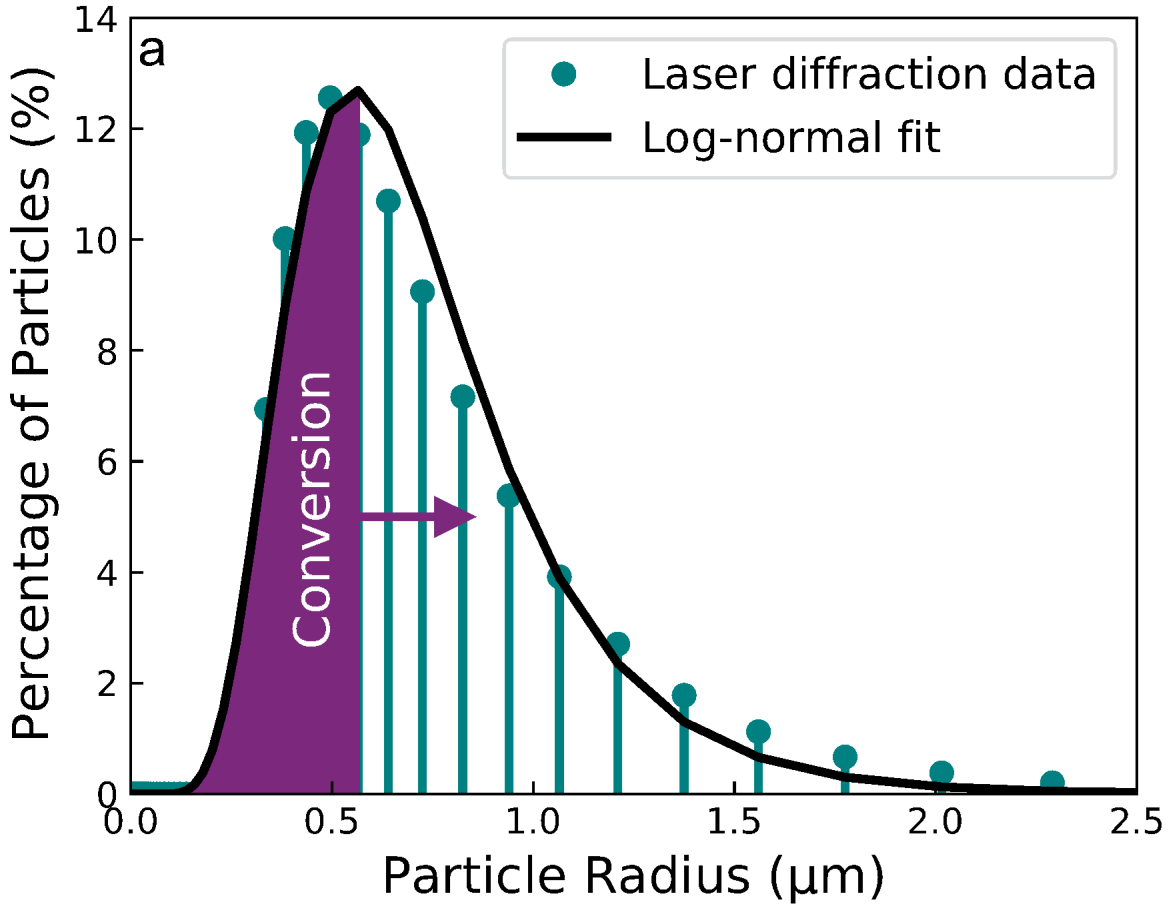
Electrolyte potential (V_l)

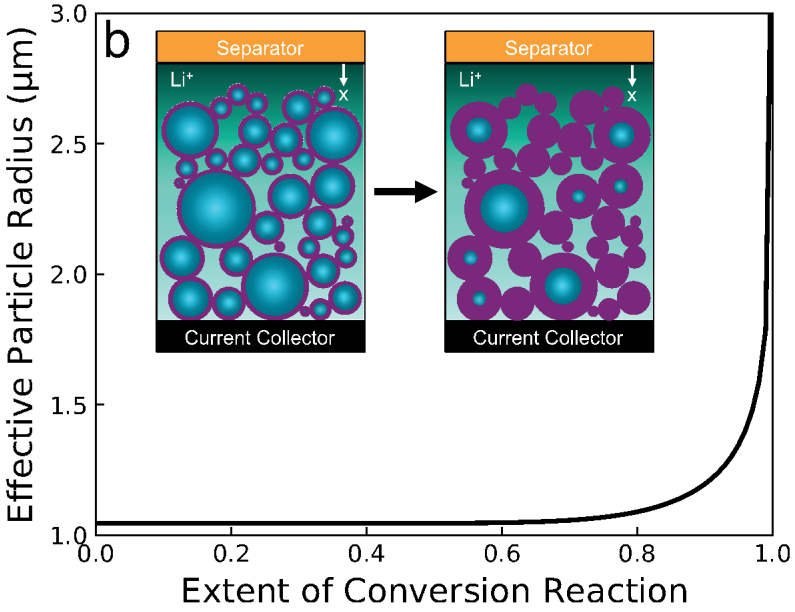
Electrolyte concentration (C_{Li^+})

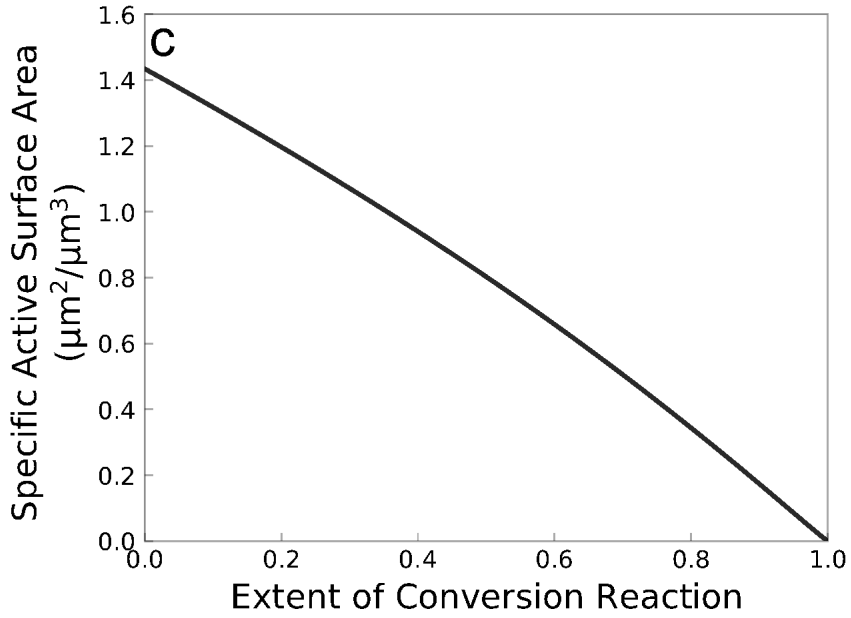
Shell thickness (δ)

Horner et al, J. Power Sources (2022), 10.1016/j.jpowsour.2022.231893

Effective particle size increases as cathode discharges

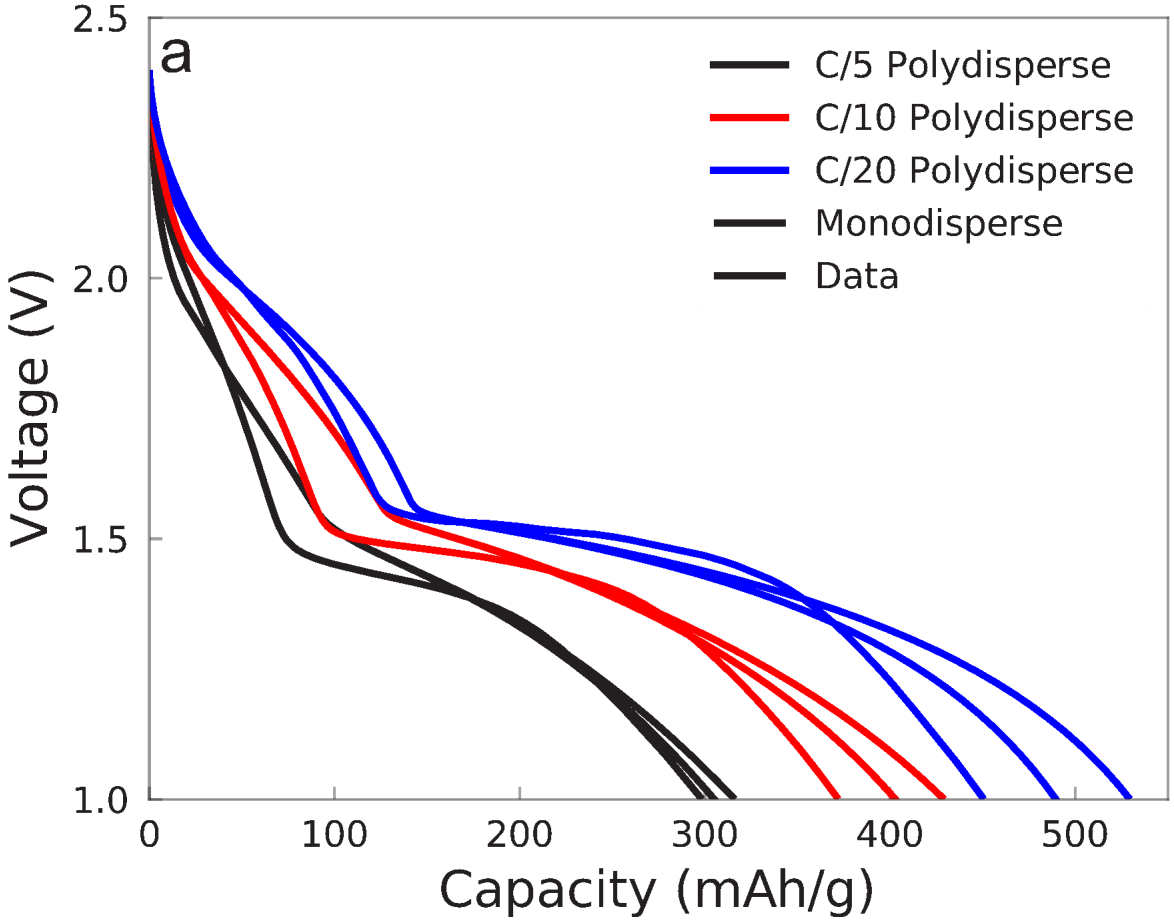


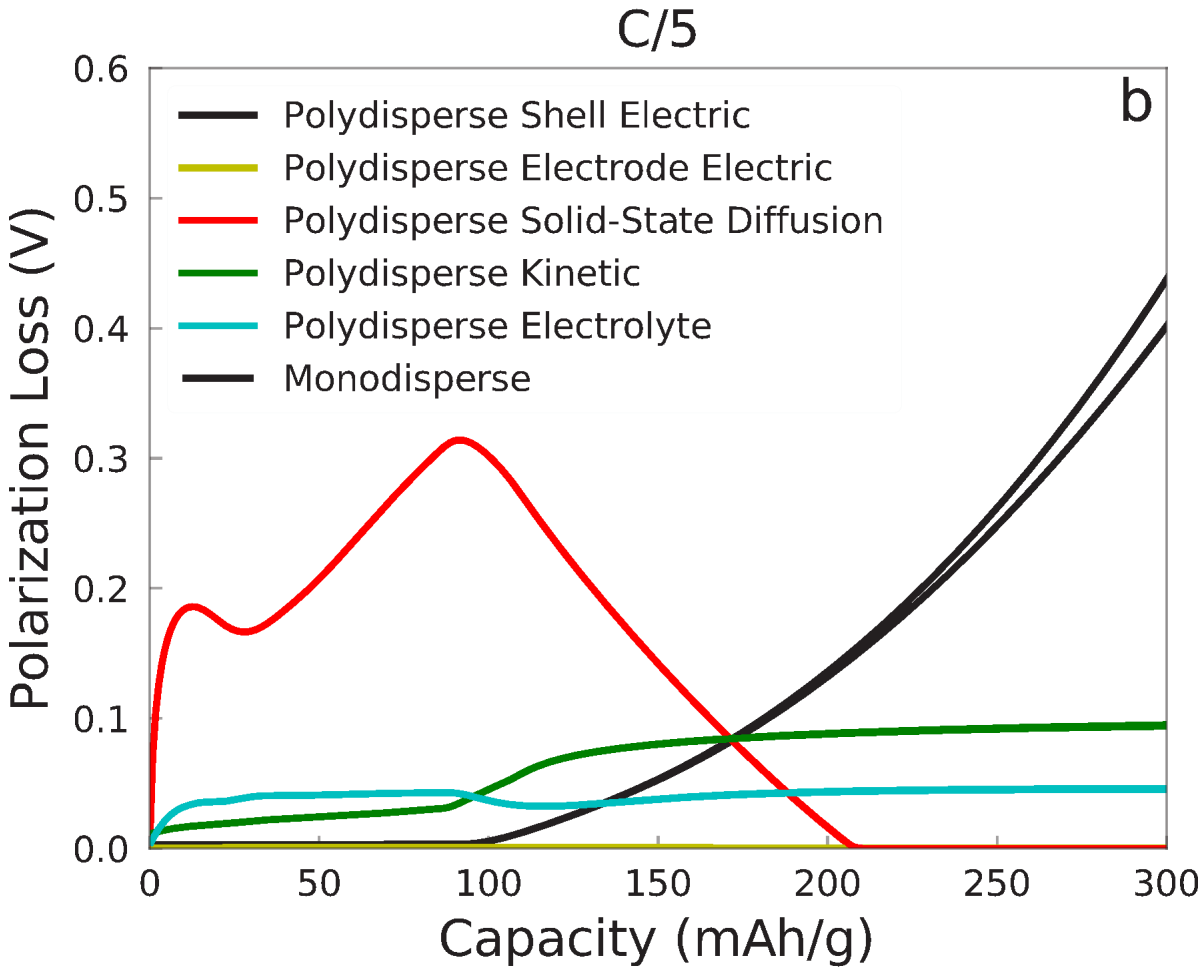




Horner et al, J. Power Sources (2022), 10.1016/j.jpowsour.2022.231893

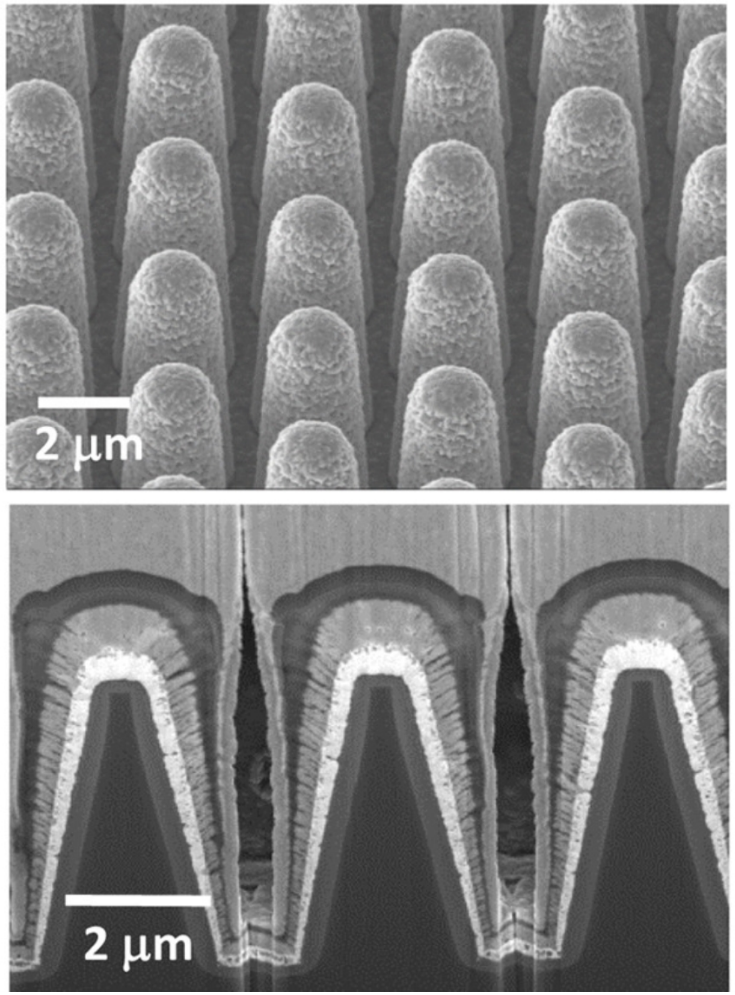
FeS_2 leading loss mechanisms



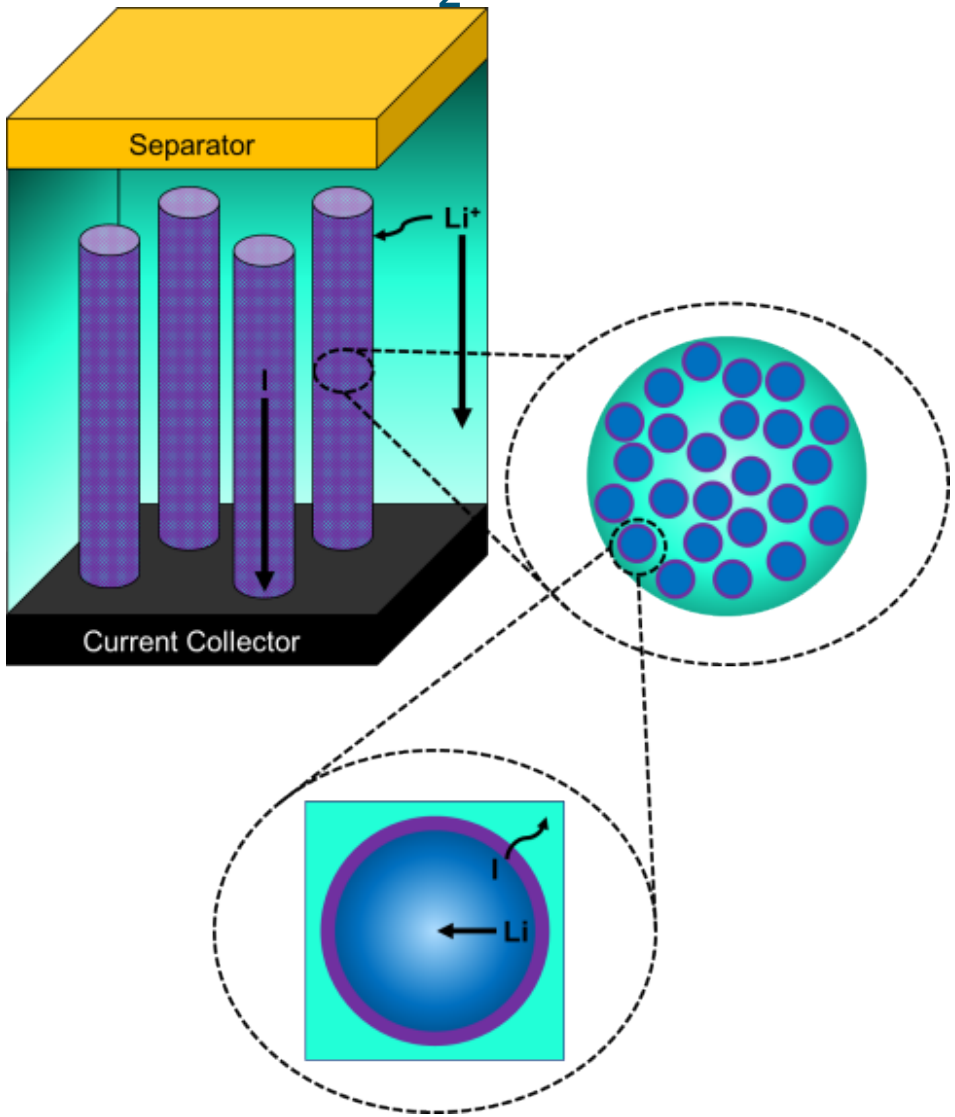


Horner et al, J. Power Sources (2022), 10.1016/j.jpowsour.2022.231893

Extending the P2D model to 2.5D features: P3.5D FeS_2 models

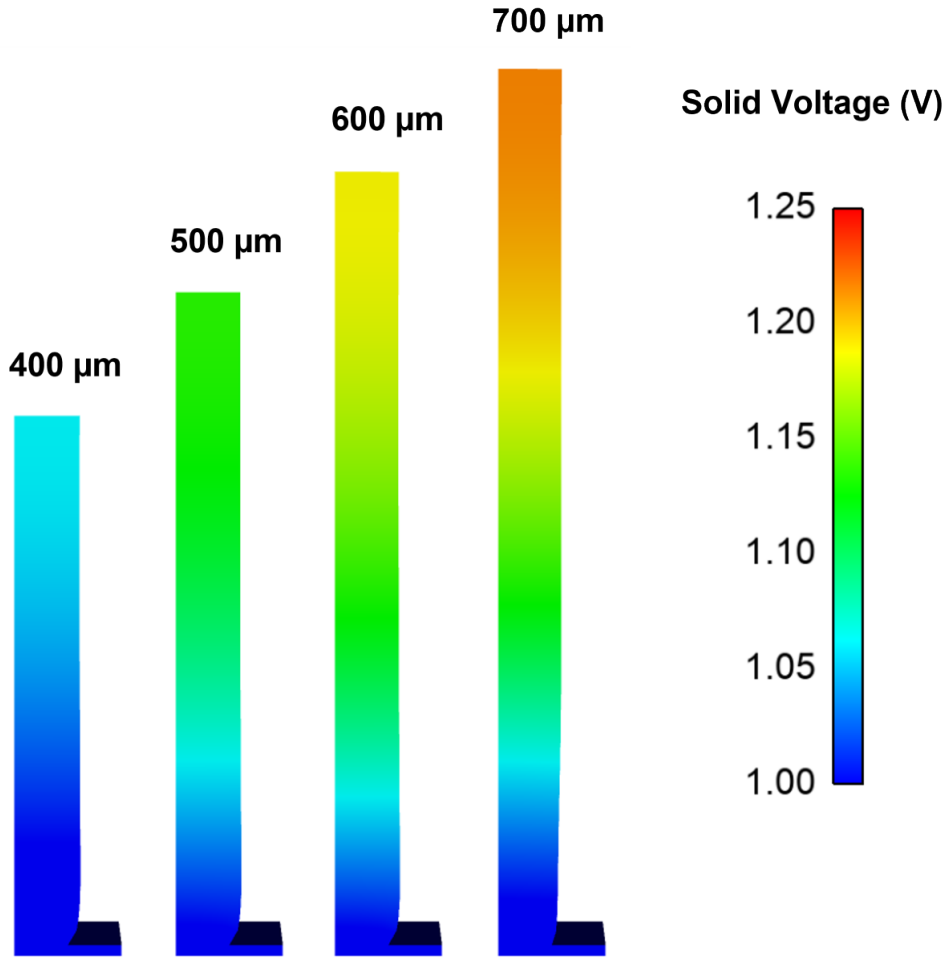


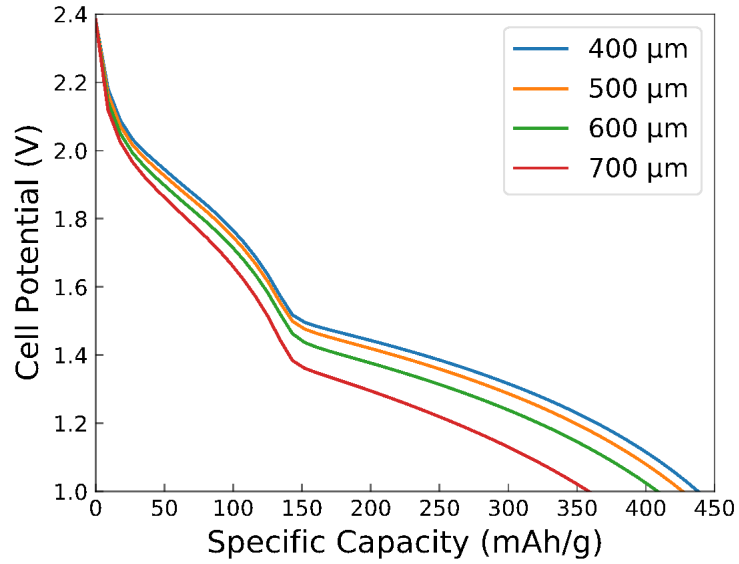
Talin et al, ACS AMI (2016)

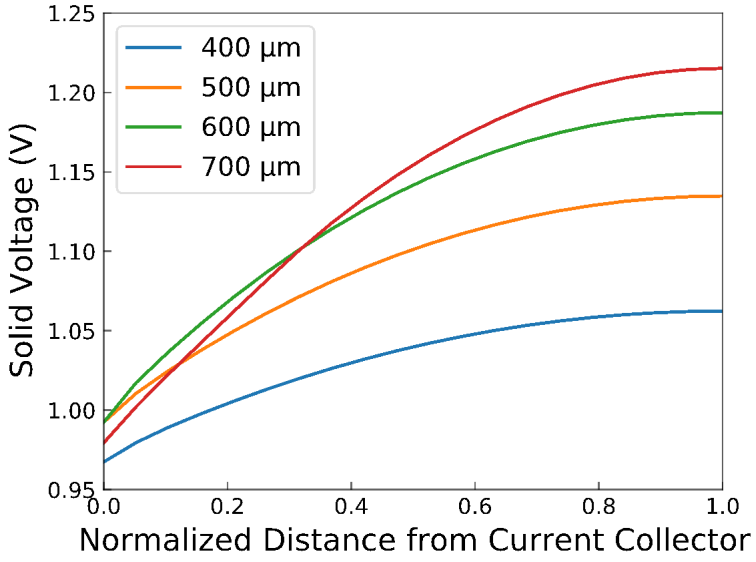


2.5D architected cells can provide high power and energy density solutions

Pillars can be very tall before limiting performance

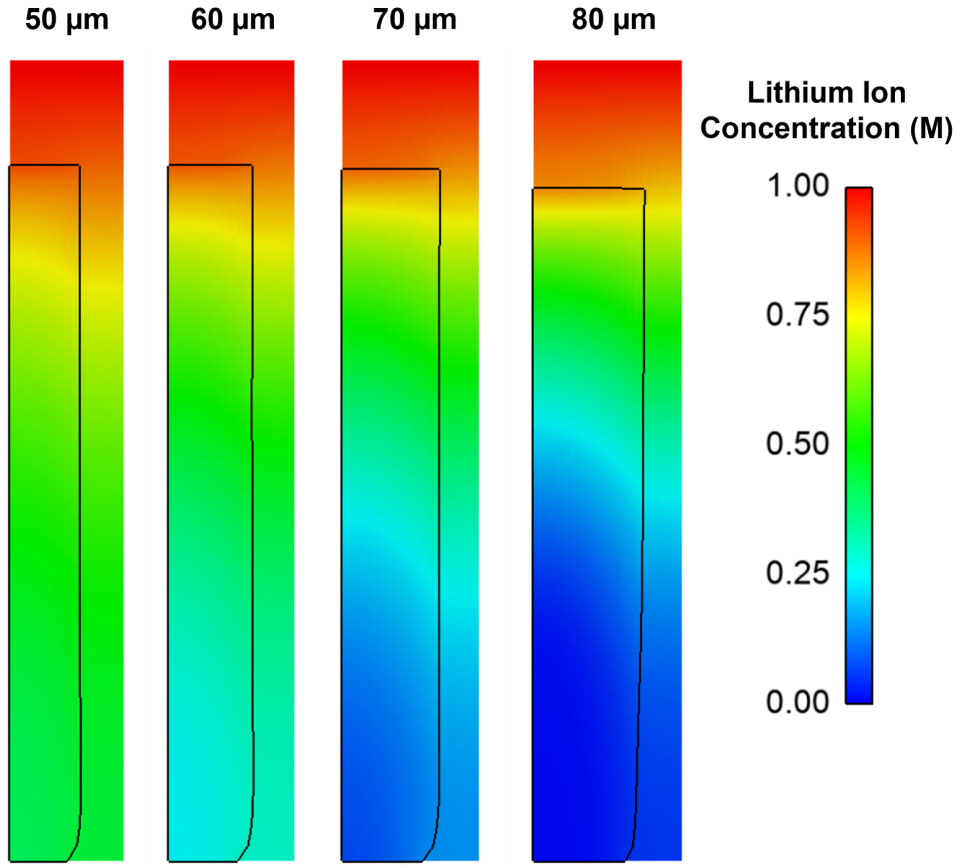


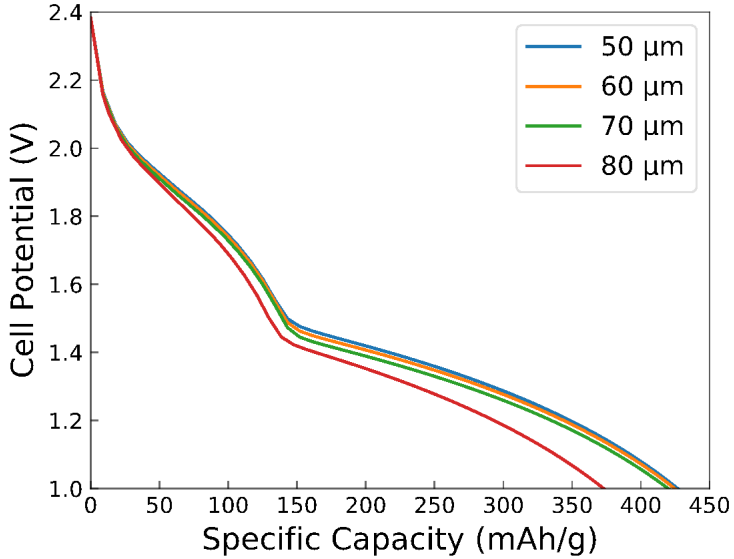


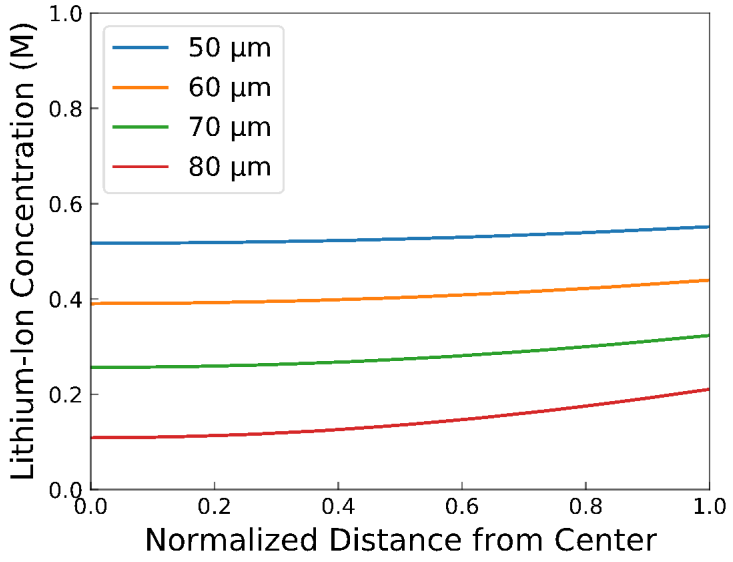


Pillar radius and gap are 50 µm, C/20 discharge

Pillars rely on radial transport to maintain performance

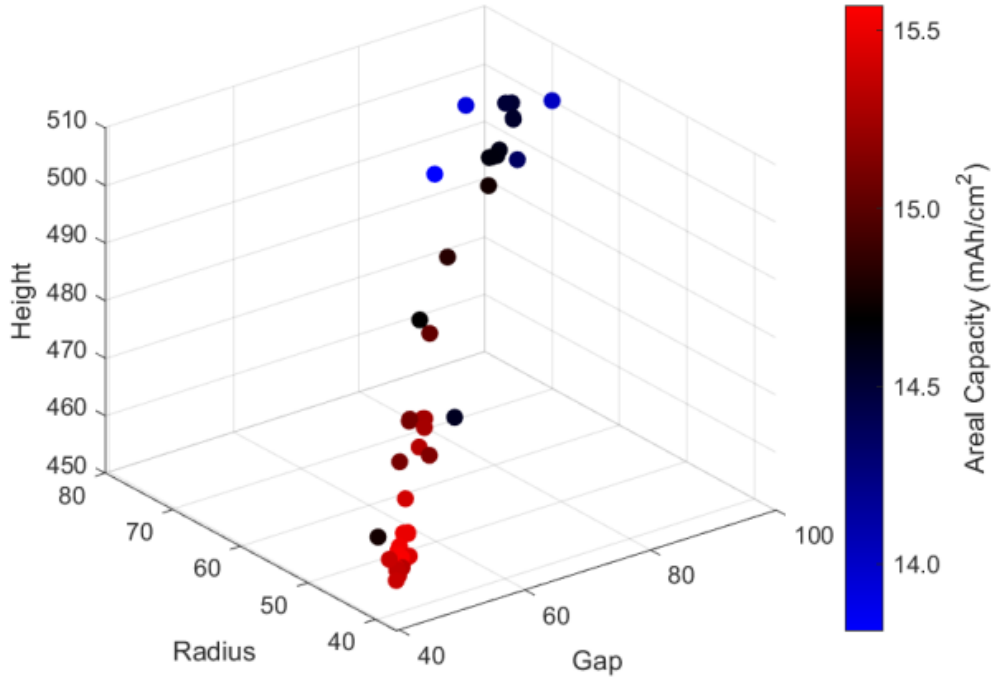






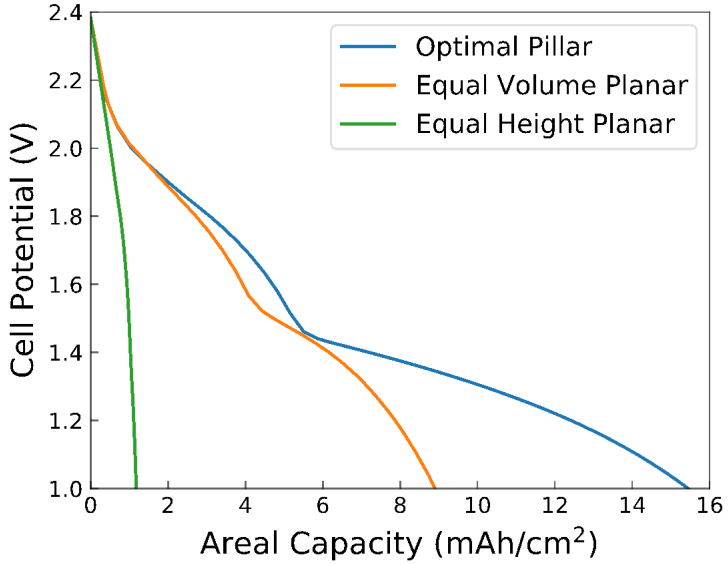
Pillar gap is 50 µm and height is 500 µm , C/20 discharge

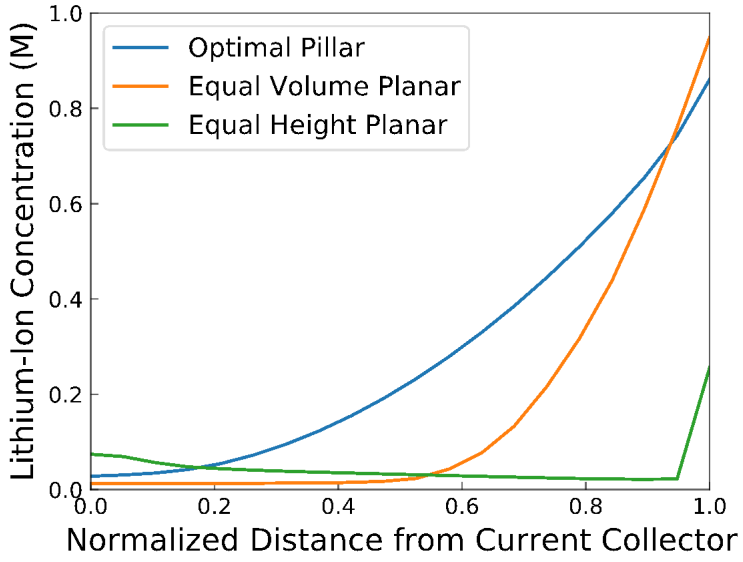
Optimal pillar geometry greatly improves performance compared to planar electrodes



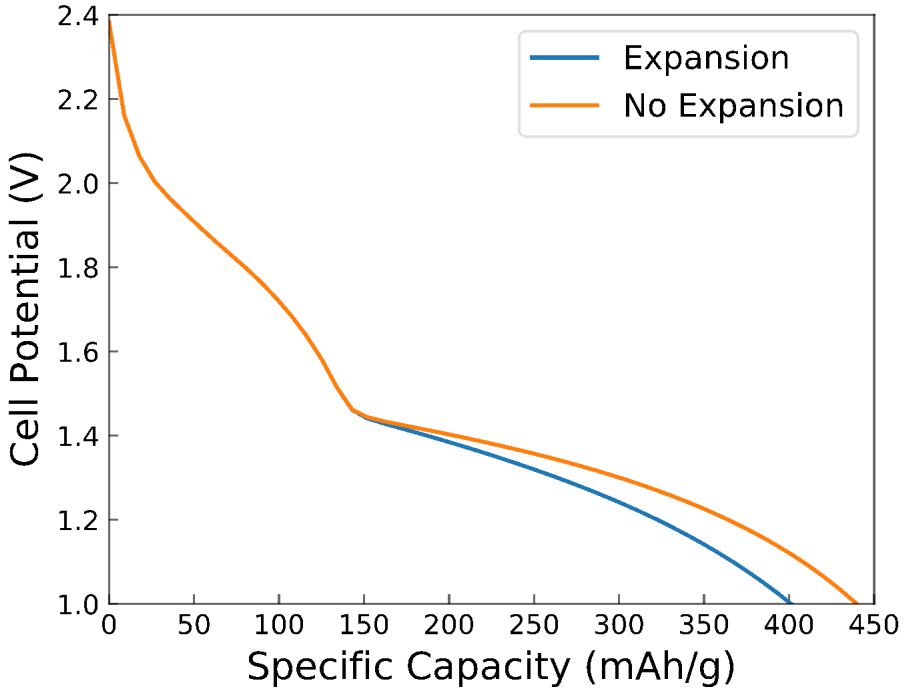
Optimal geometry, C/20

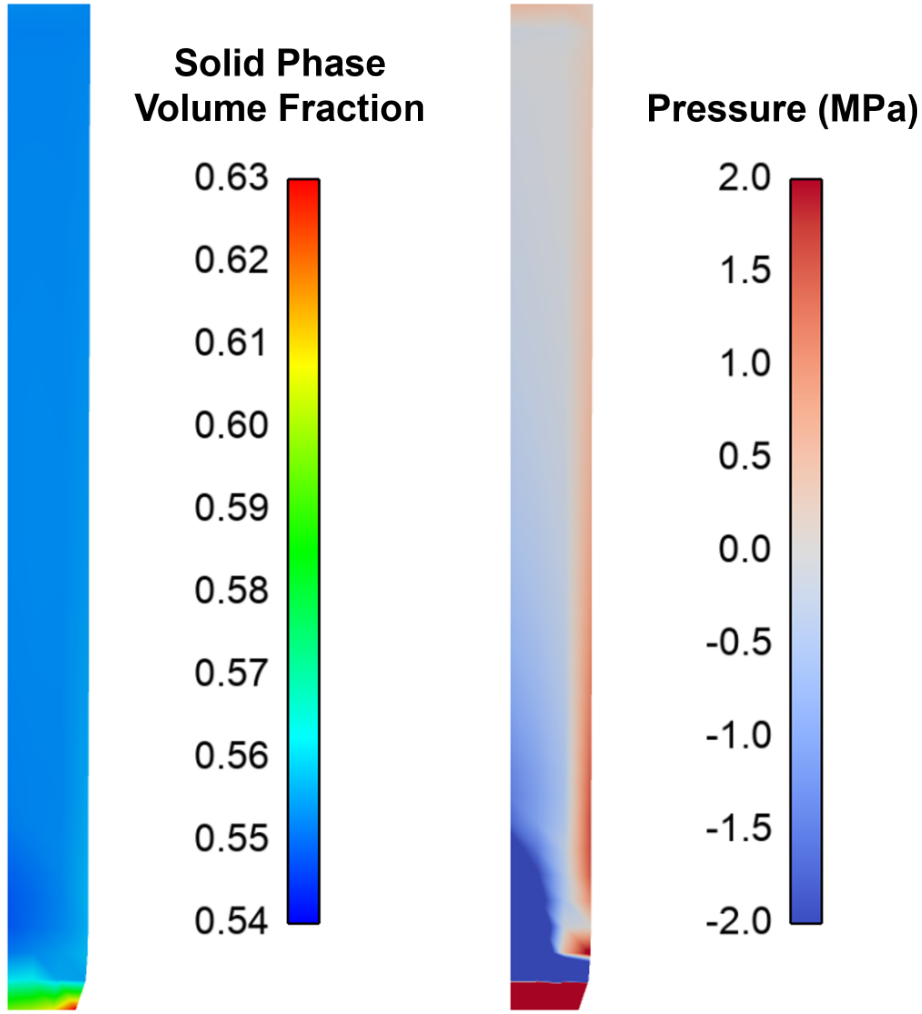
Dimension	µm
Height	460
Radius	37
Gap	40



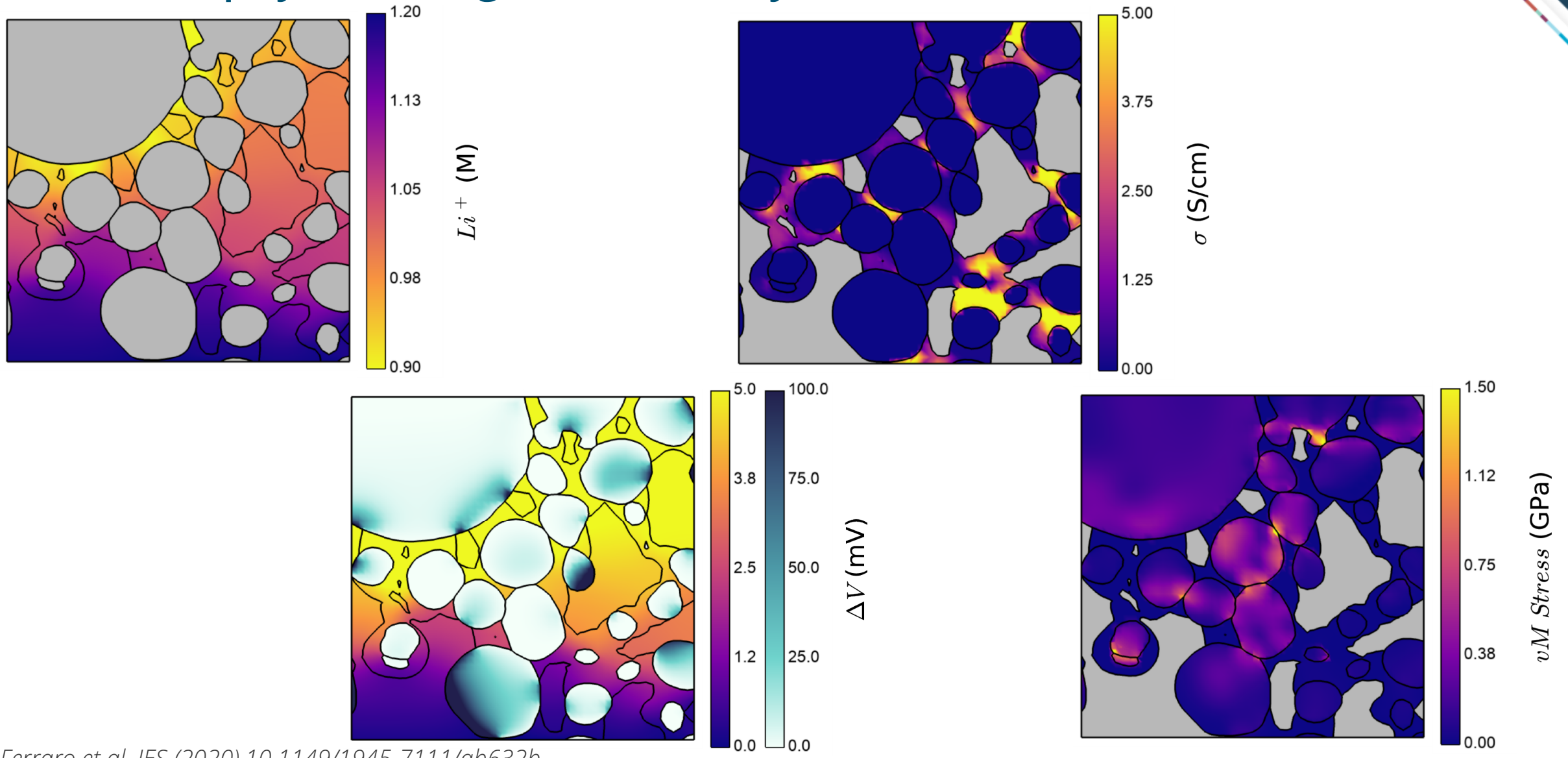


Consideration of particle swelling and mechanical constraints decreases available capacity



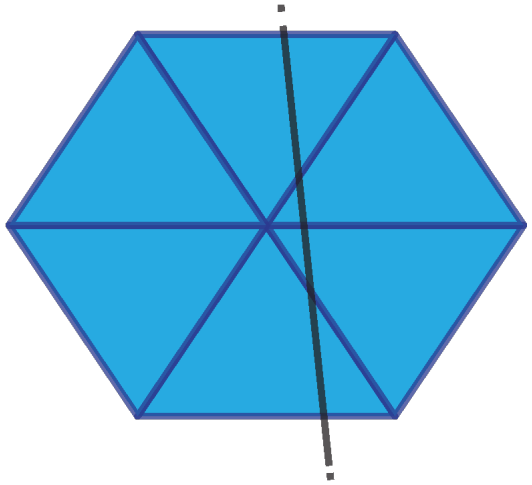


Volume averaging is nice, but mesoscale models can provide additional physical insight and fidelity

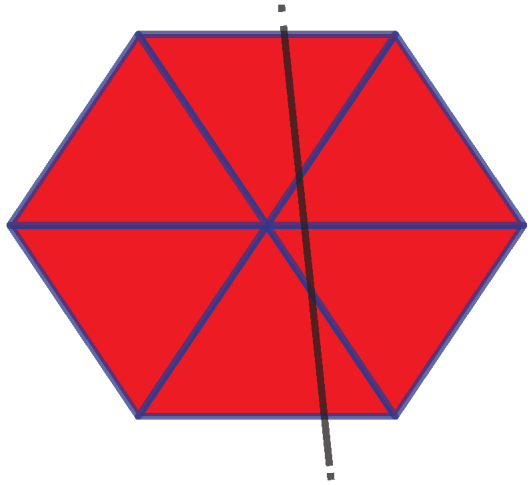


Ferraro et al, JES (2020),10.1149/1945-7111/ab632b

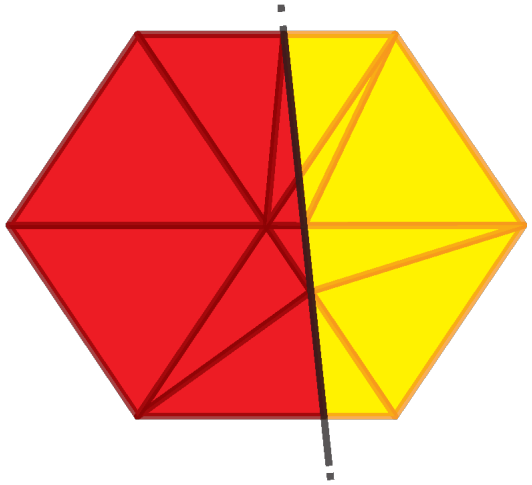
Interface representation – CDFEM/cThruAMR



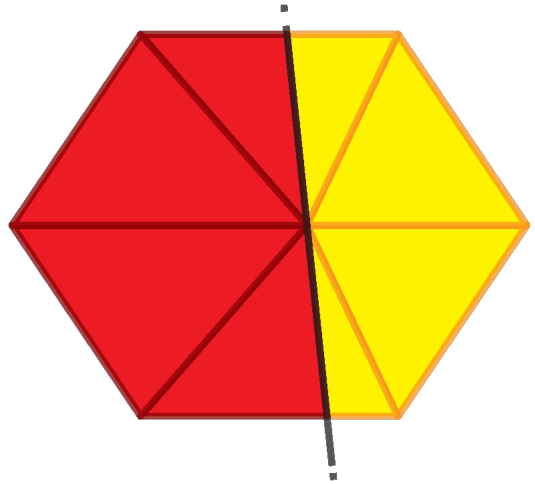
Background Mesh



Level-set or phase field

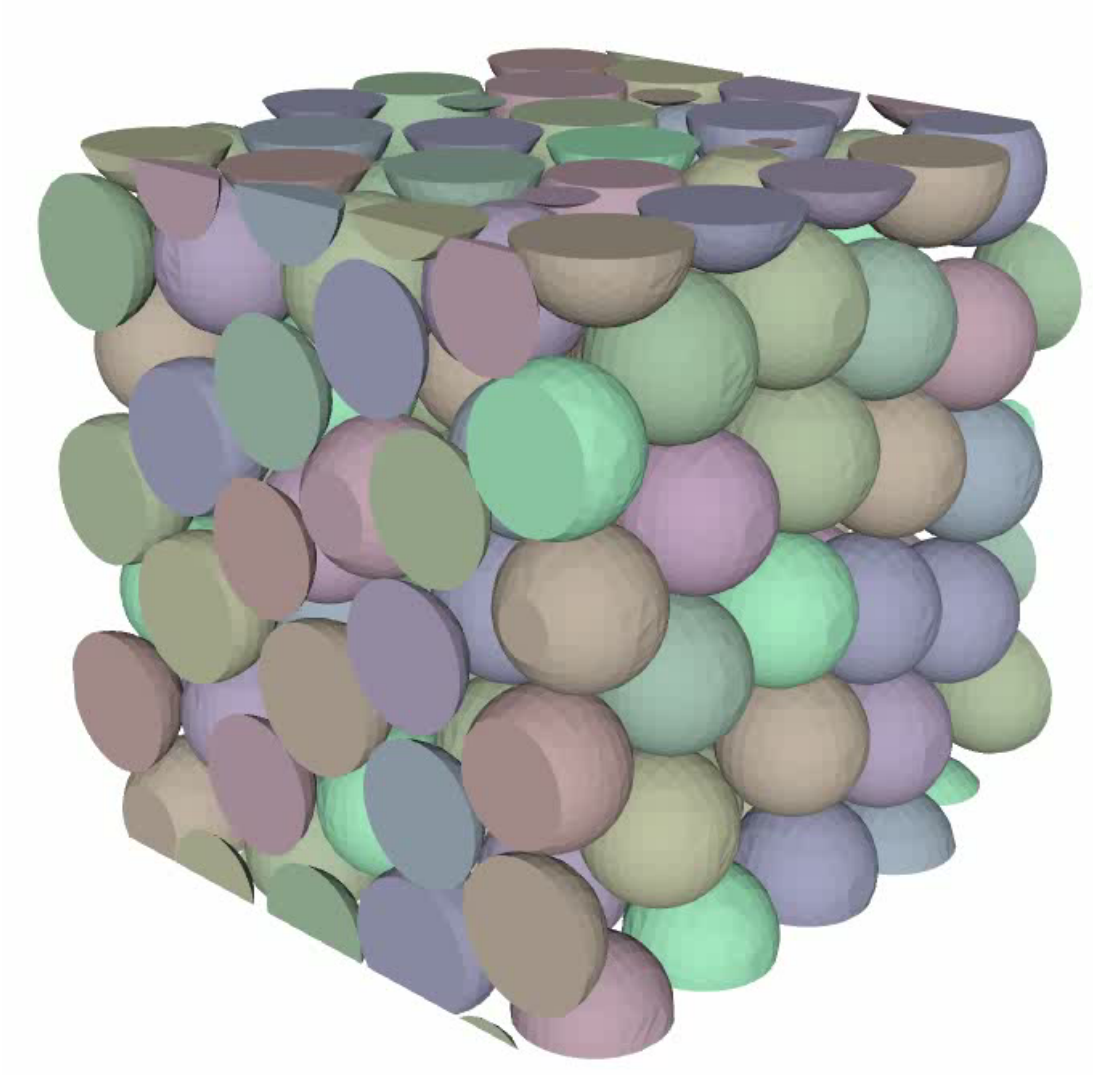


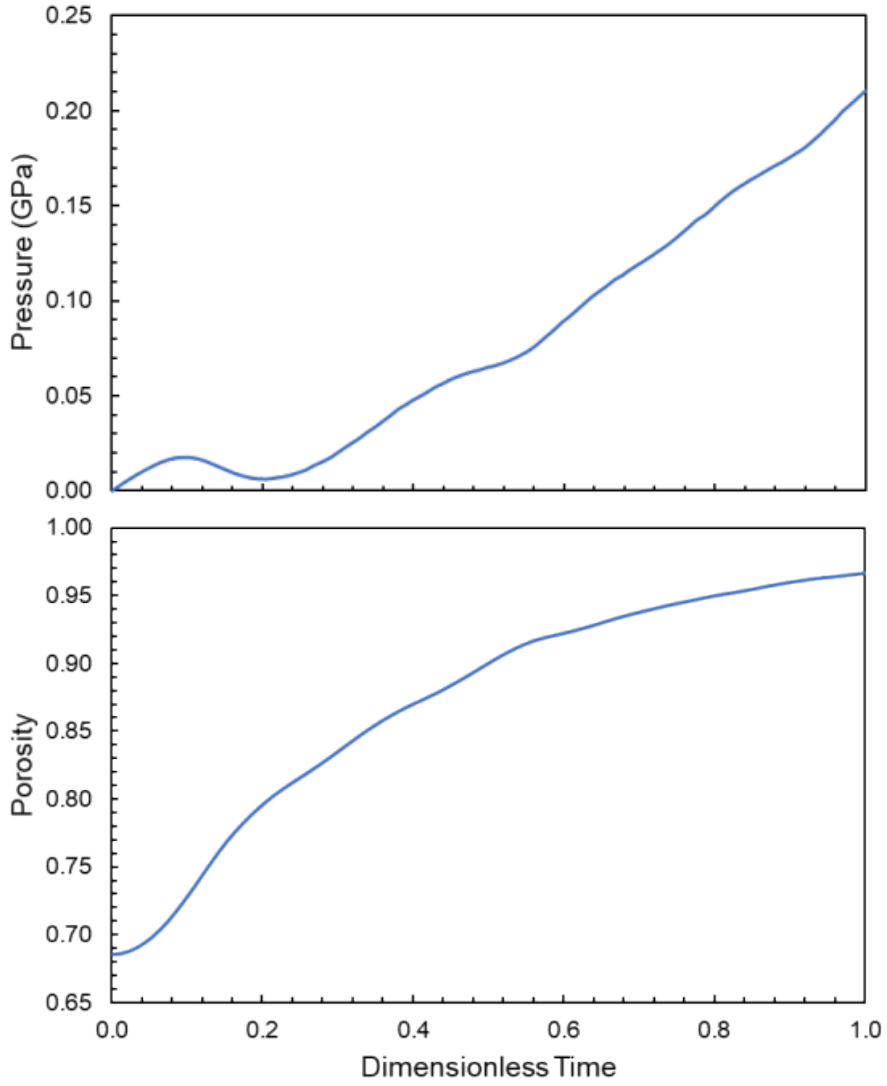
CDFEM



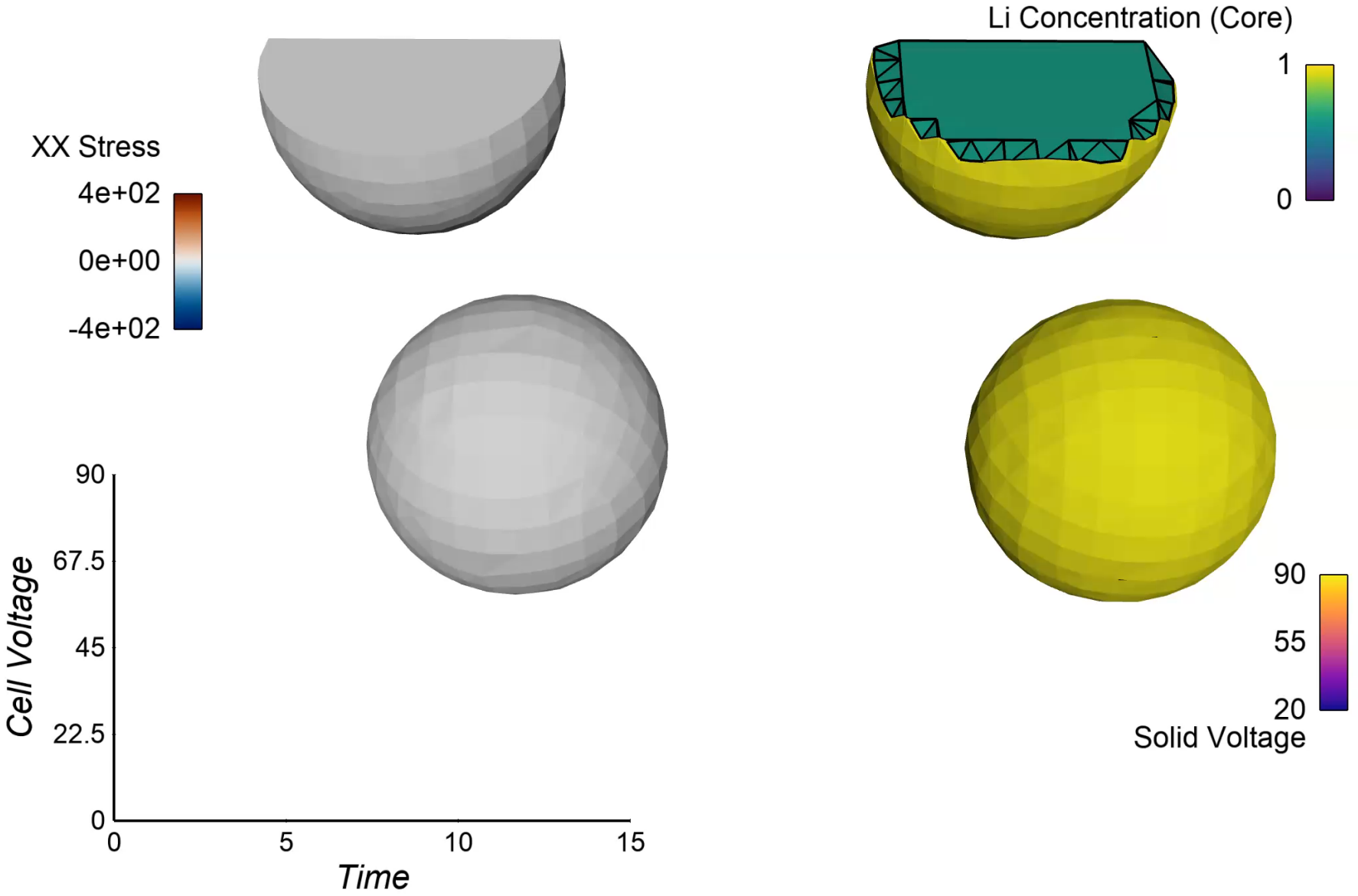
cThruAMR

Swelling of FeS_2 particles impacts ionic transport

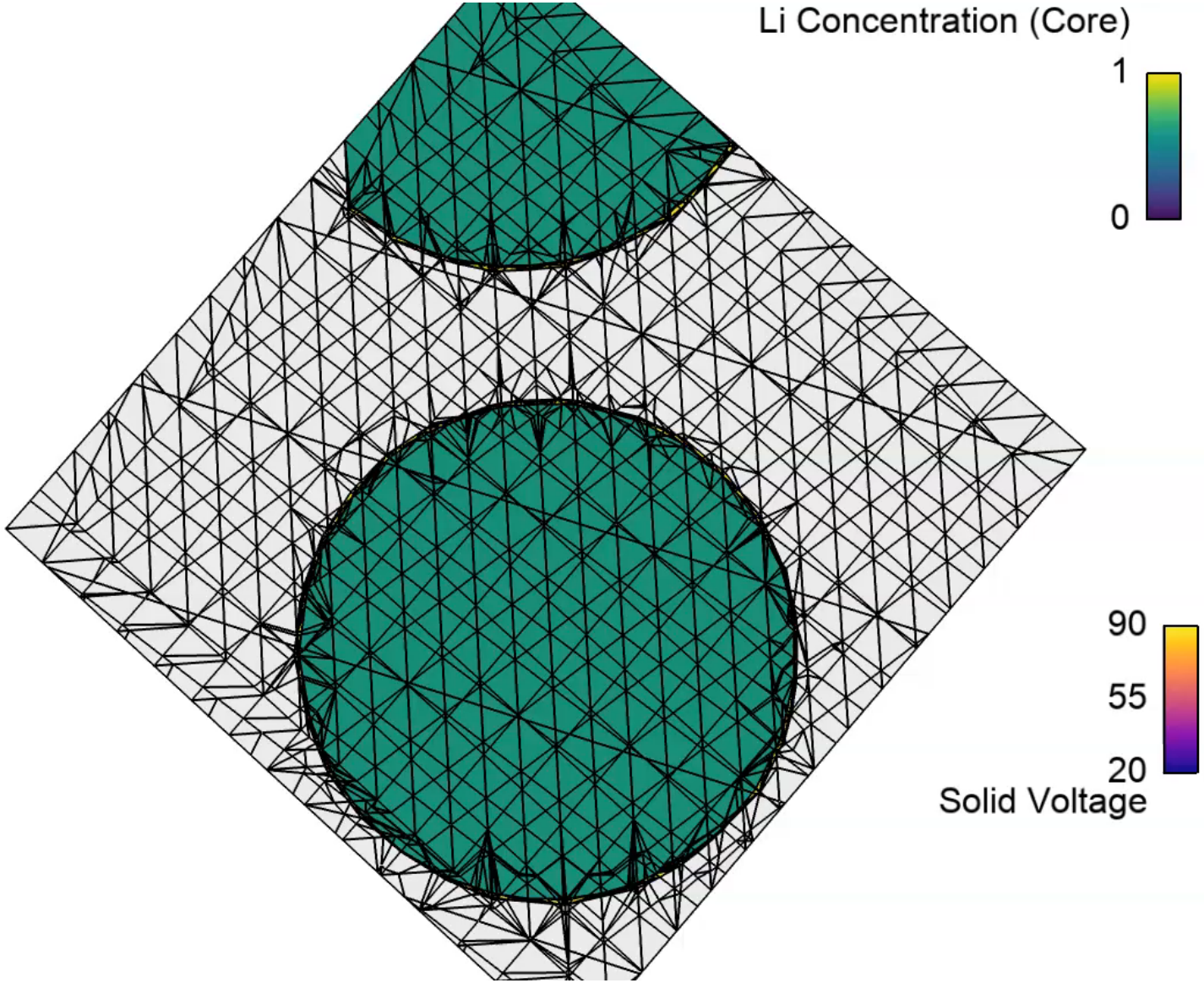




Early work towards a coupled electrochemical-mechanical model of FeS_2 that includes intercalation and conversion



Early work towards a coupled electrochemical-mechanical model of FeS_2 that includes intercalation and conversion



THANK YOU!

Scott A. Roberts, Ph.D.
Distinguished R&D Chemical Engineer
Thermal/Fluid Component Sciences (1513)

Phone: (505) 844-7957

Email: sarober@sandia.gov

Web: https://www.sandia.gov/-sarober