Overcoming Interfacial Impedance in Solid State Batteries

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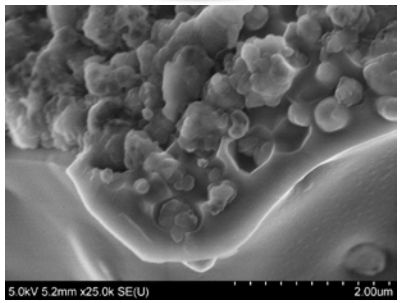
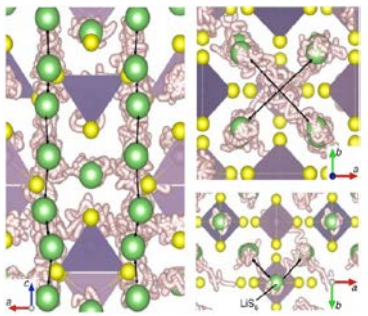
Overcoming Interfacial Impedance in Garnet-Based SSLiBs

WACHSMAN
Garnet structure fabrication & interfacial impedance analysis

HU
Interface layer development, cell fabrication, & evaluation

MO
Computational solid-state ion transport and interfaces





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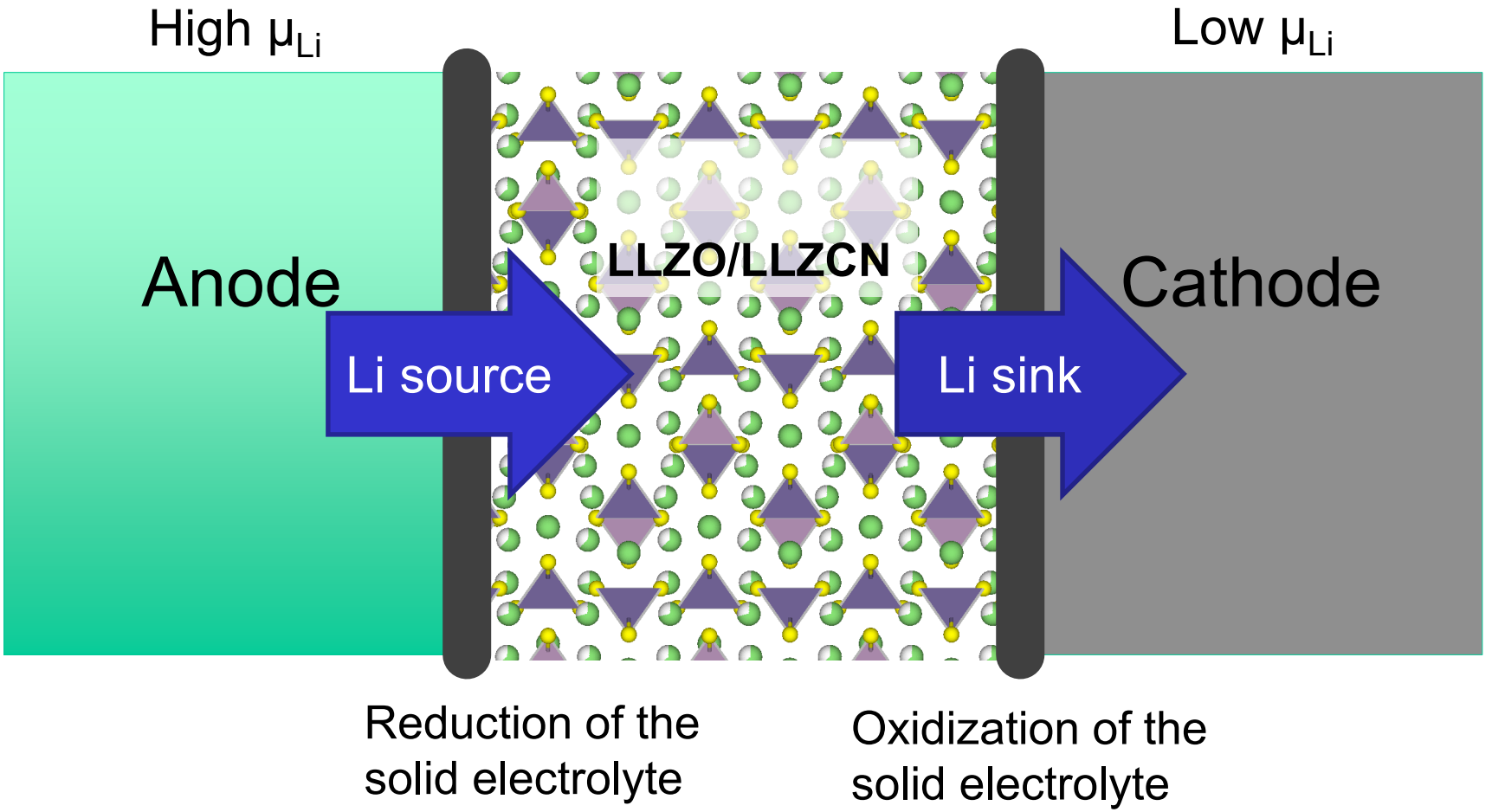
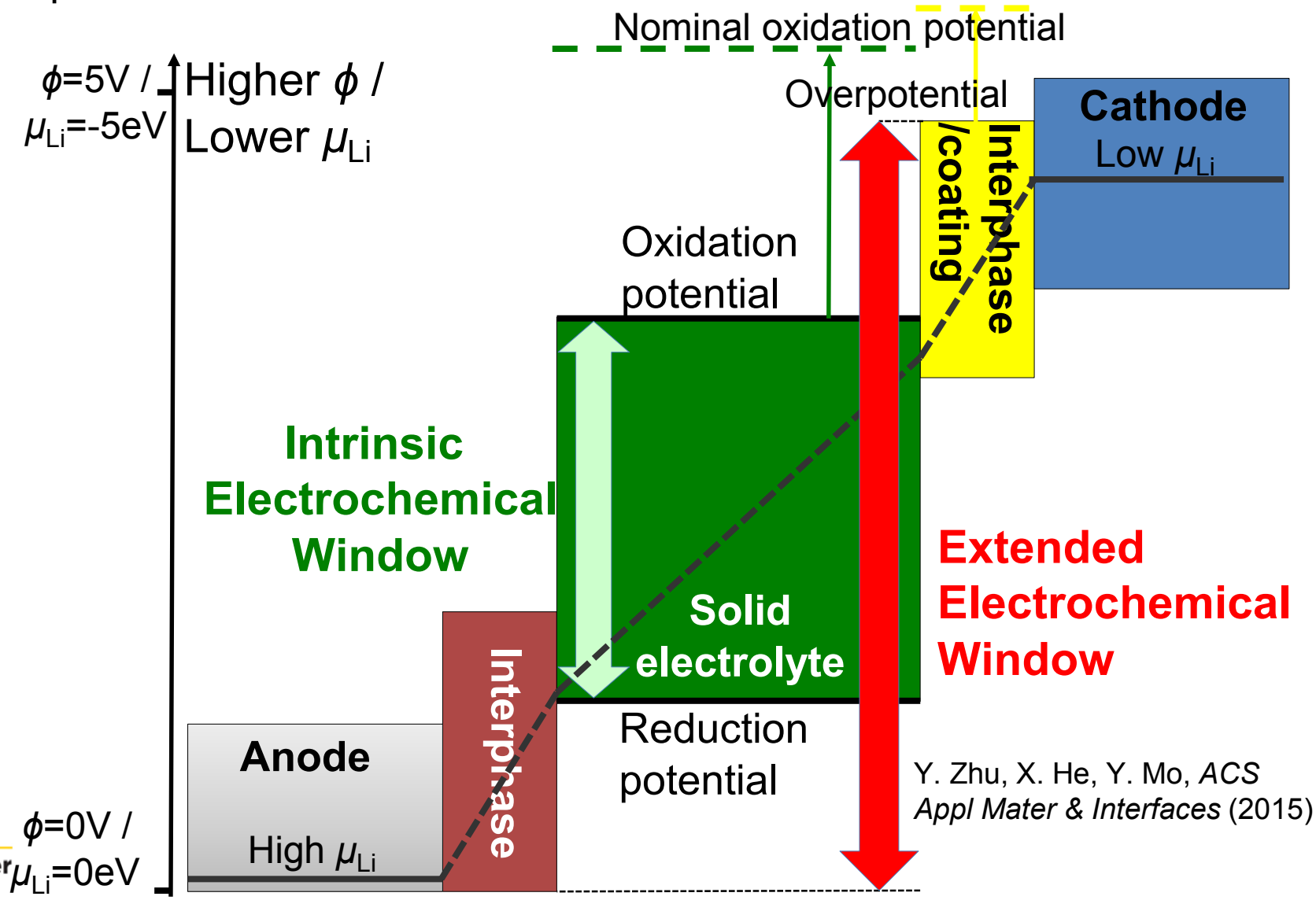
Interfacial Stability Using First Principles Computations



- **Electrochemical Stability** are evaluated as the phase stability in equilibrium with a Li reservoir (or applied potential) using **Li grand potential phase diagram**

Stability Achieved by Interphase/Coating Layers

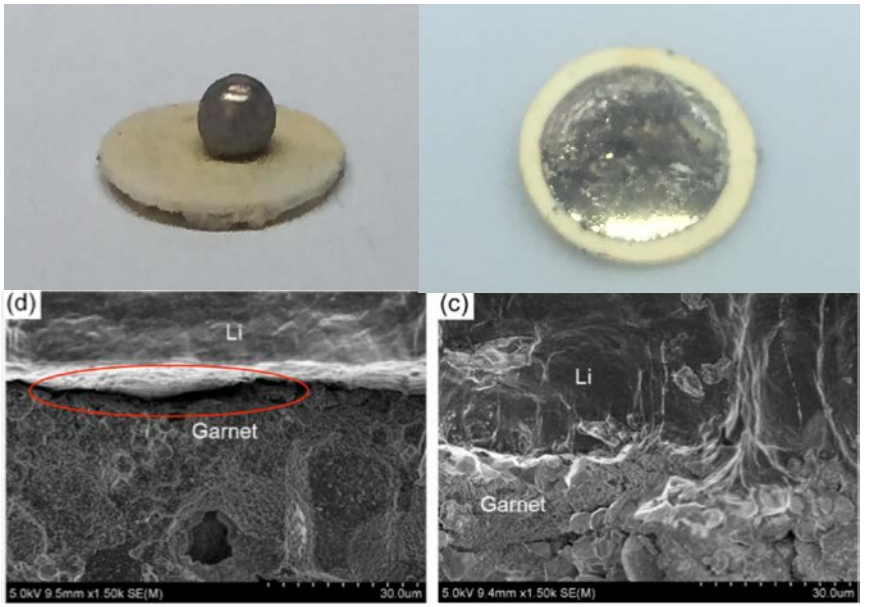
- The electrochemical window of solid electrolyte is usually thermodynamically limited and is extended by the interphase/coating layers.
- The formation of interphase layers at electrolyte-electrode interfaces has significant impact on the interfacial resistance.



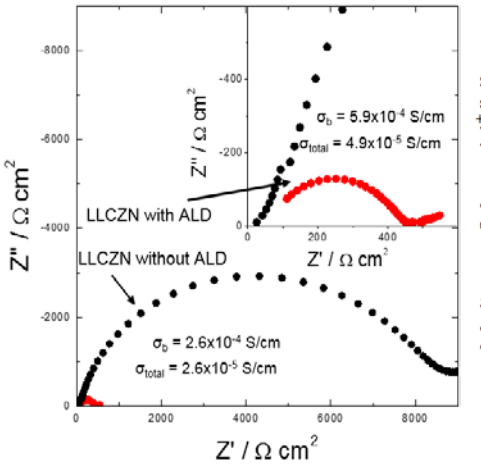
Y. Zhu, X. He, Y. Mo, *ACS Appl Mater & Interfaces* (2015)

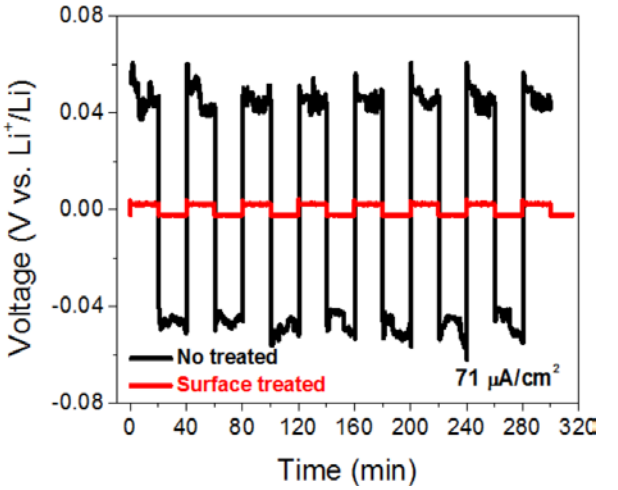
Overcoming Li/Garnet Interfacial Impedance

- Developed surface treatment that allows Li metal to wet garnet surface

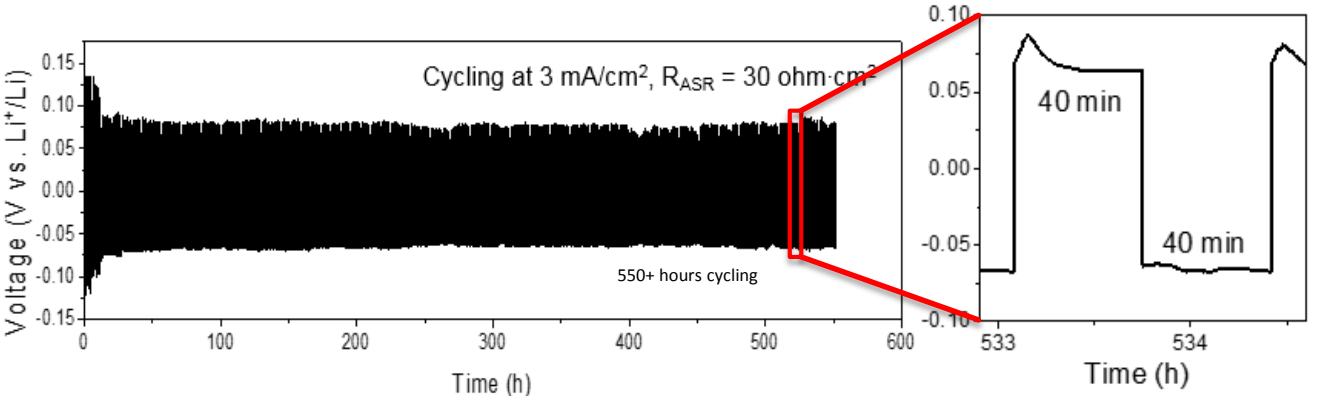


- Surface treatment dramatically reduces interfacial resistance



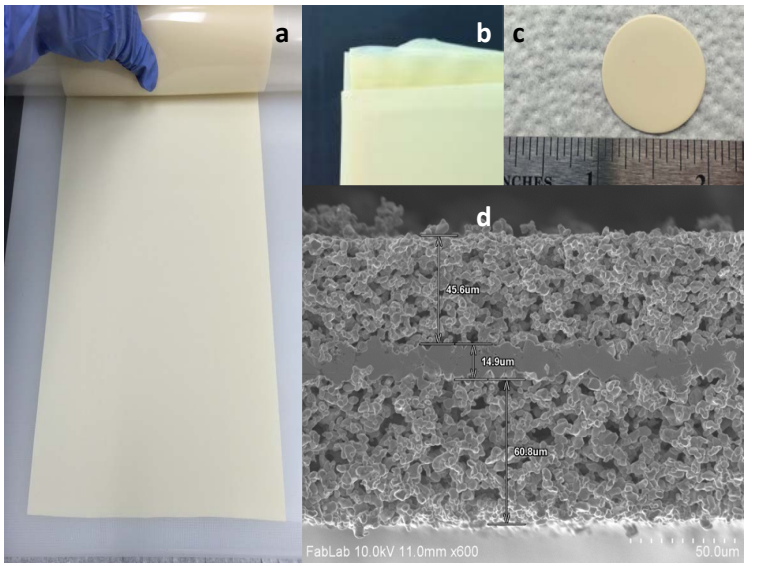


- Resulting in long term, high rate lithium cycling with no degradation
  - Demonstrated in multiple cells with planar, bilayer and trilayer configuration
  - Demonstrated reduction in overpotential due to negligible interfacial impedance

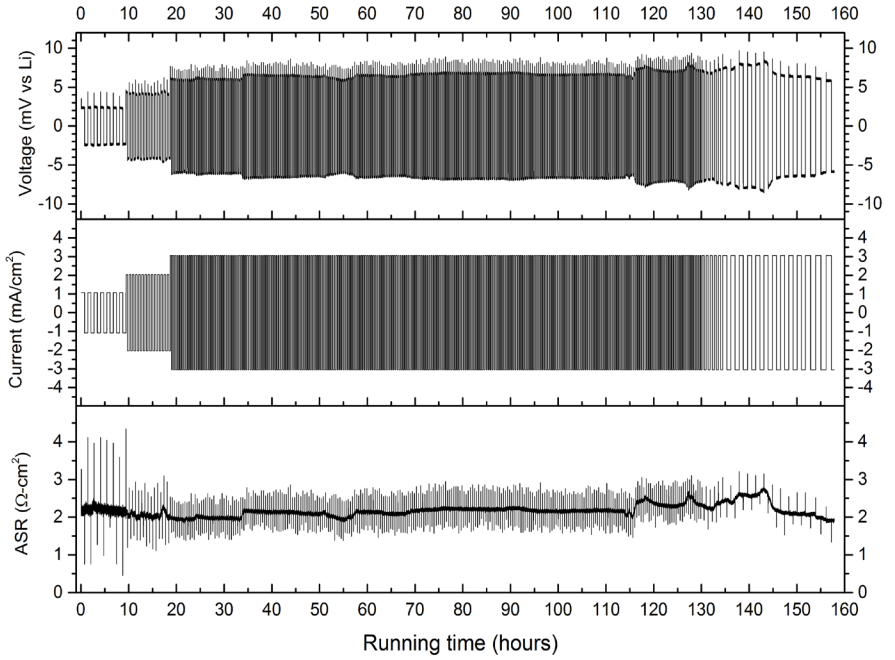


Overcoming Li/Garnet Interfacial Impedance

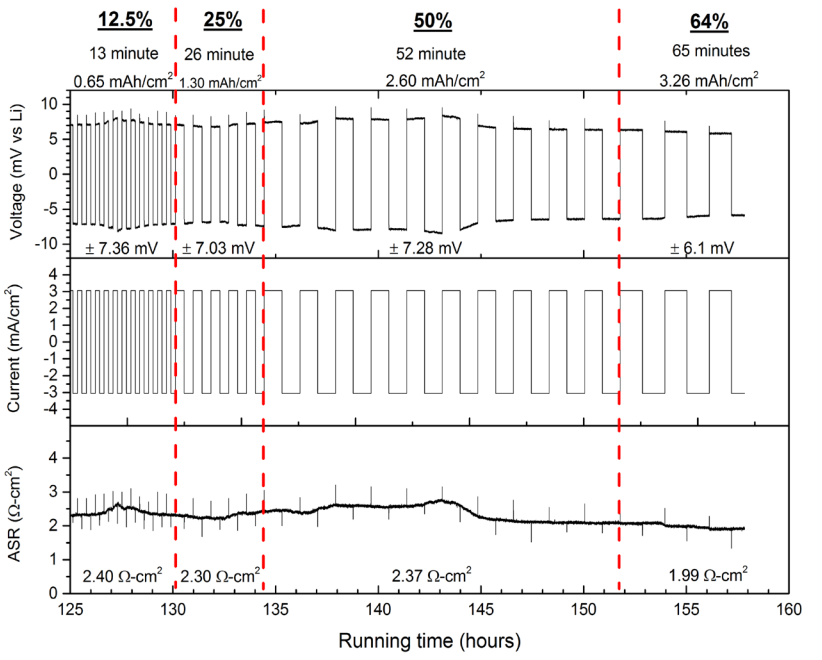
- Developed scalable and reproducible process to fabricate multilayer garnet structures with thin (15 µm) dense central layer on high surface area porous supports



- Demonstrated high current density 3mA/cm^2 with this trilayer structure



- and high Li capacity (3.26 mAh/cm^2) cycling with no degradation or shorting



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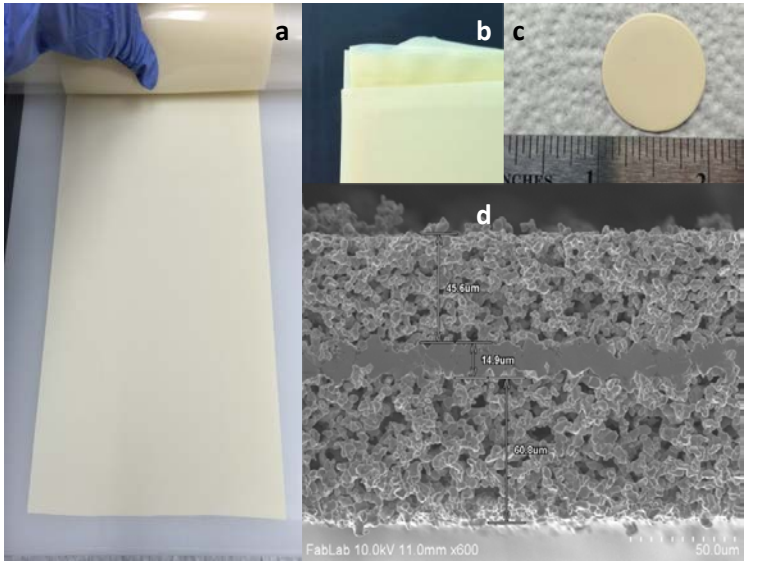
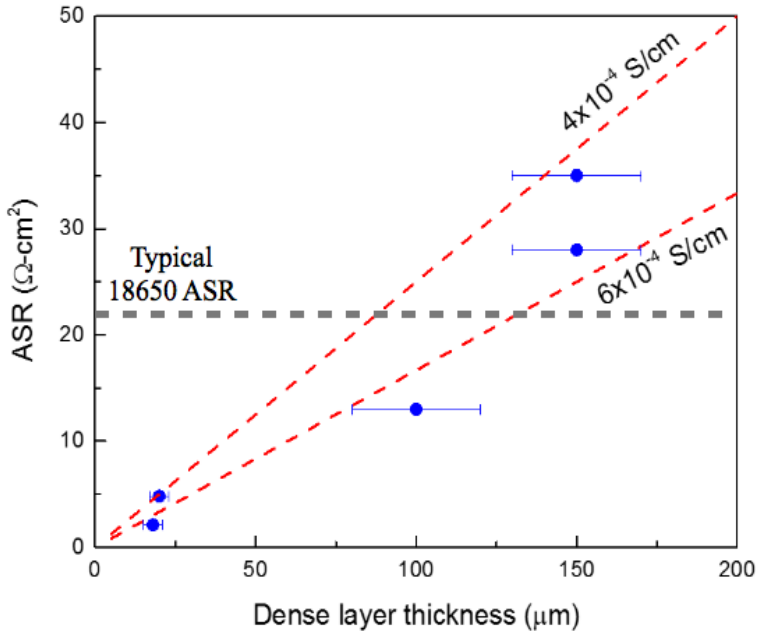
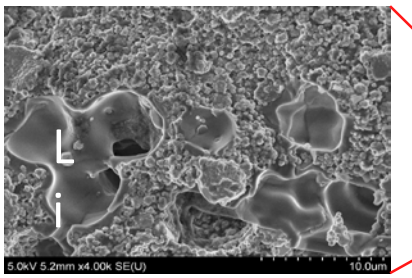
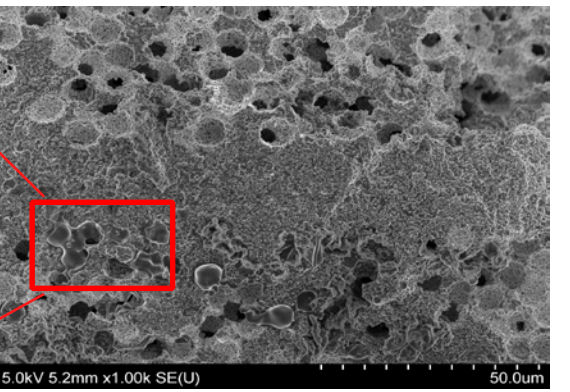


- Resulting in extremely low ASR (2 Ωcm^2) consistent with 15 µm thick garnet conductivity indicating no interfacial impedance



- and demonstrating surface treatment extends to garnet interior with Li metal filling of garnet pores





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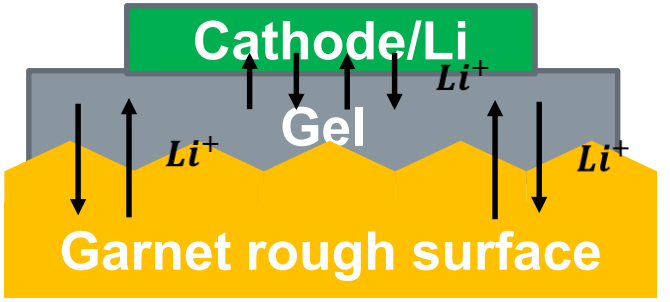
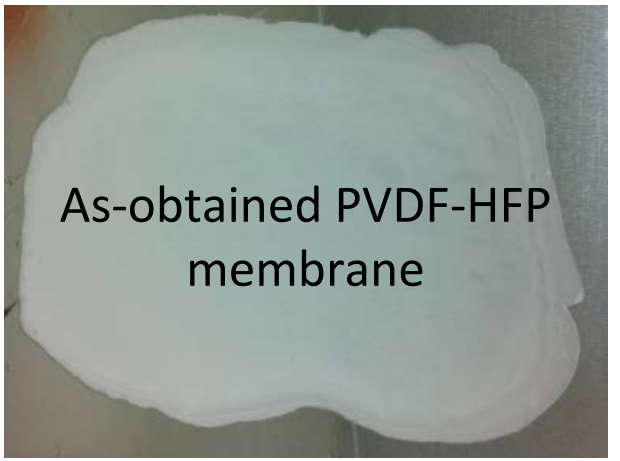
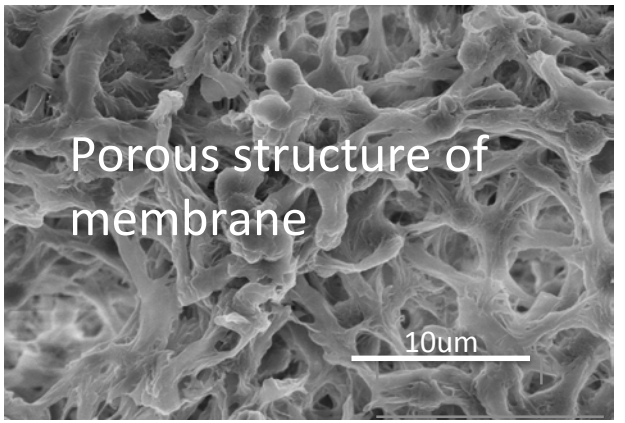
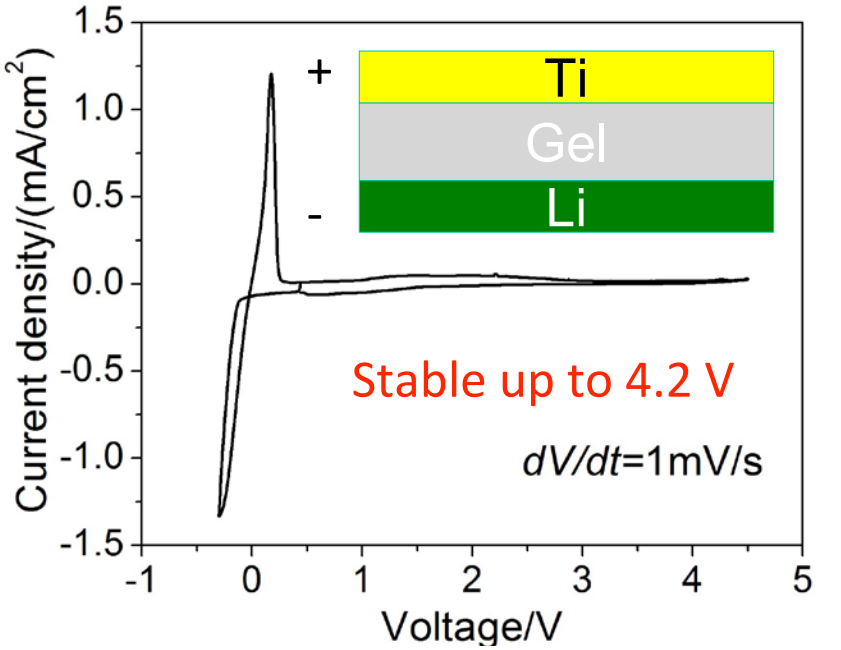
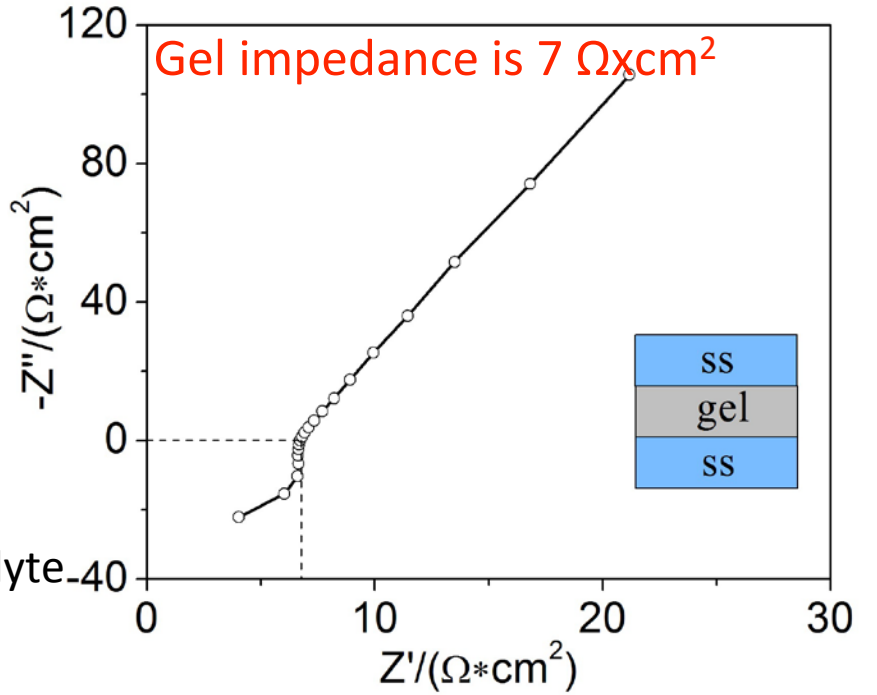
Overcoming Cathode/Garnet Interfacial Impedance





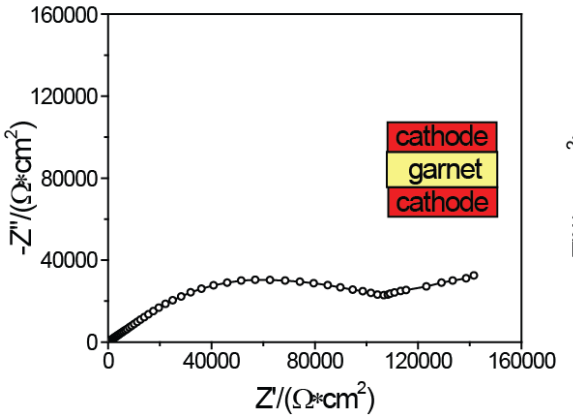


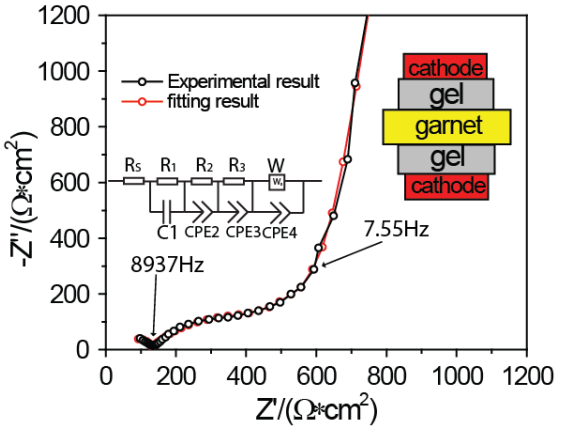


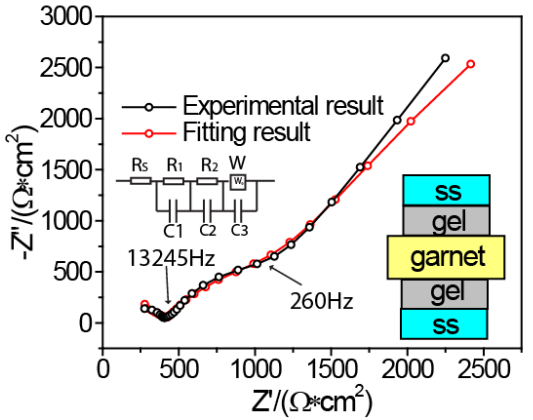


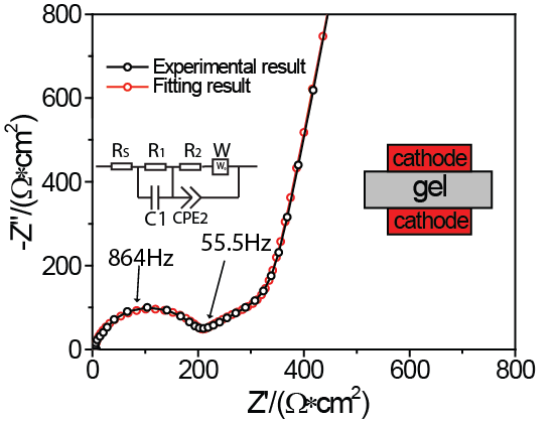
Gel: PVdF-HFP membrane + $LiPF_6$ in EC/DEC electrolyte

Overcoming Cathode/Garnet Interfacial Impedance







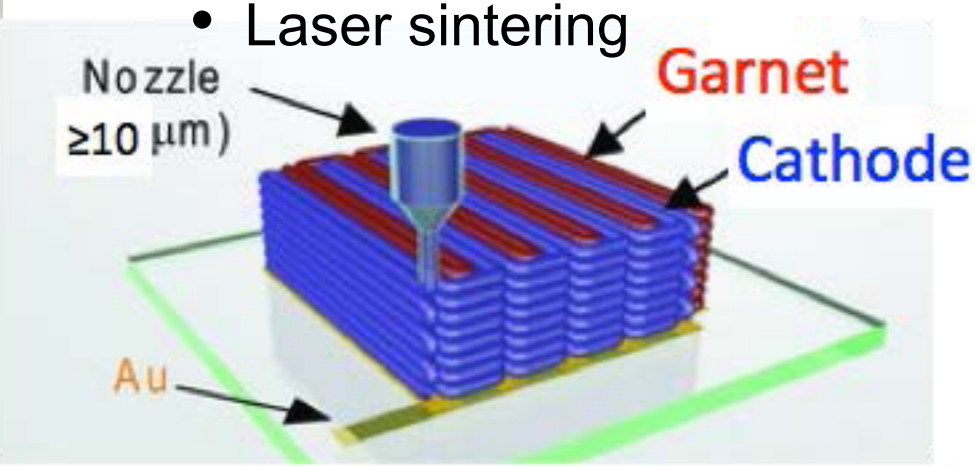


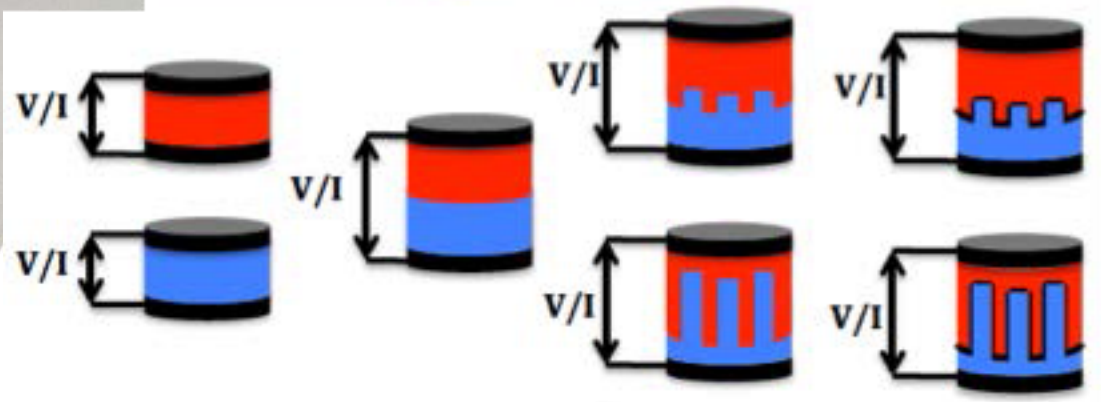
The gel electrolyte significantly reduces the interfacial impedance between the $LiFePO_4$ cathode and garnet from $6x10^4$ to 350 Ohm cm^2

Half cell with/without gel	Component	Resistance (Ω*cm2)
Cell with gel	Garnet	150
	Cathode/gel interface and gel/garnet interface	350
	Total	850
Cell without gel	One interface	~$6X10^4$
	Total	~$1.3X10^5$

Extending to 3D Interface Structures

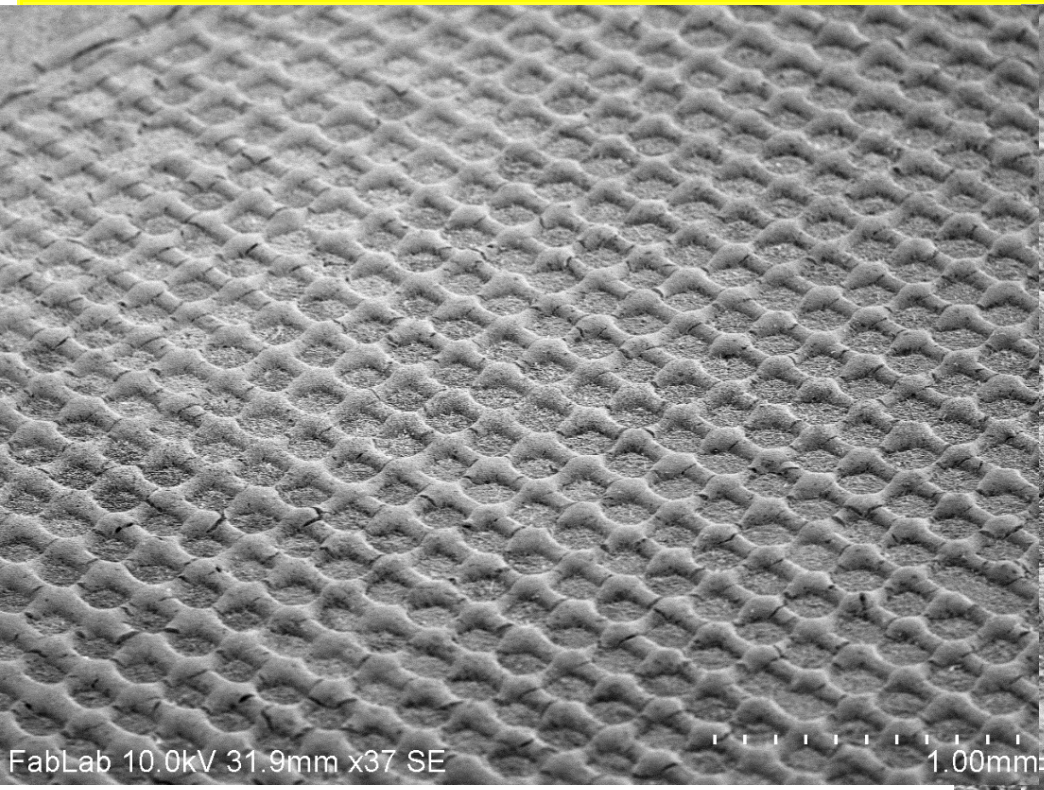
- Dual print heads
- Laser sintering

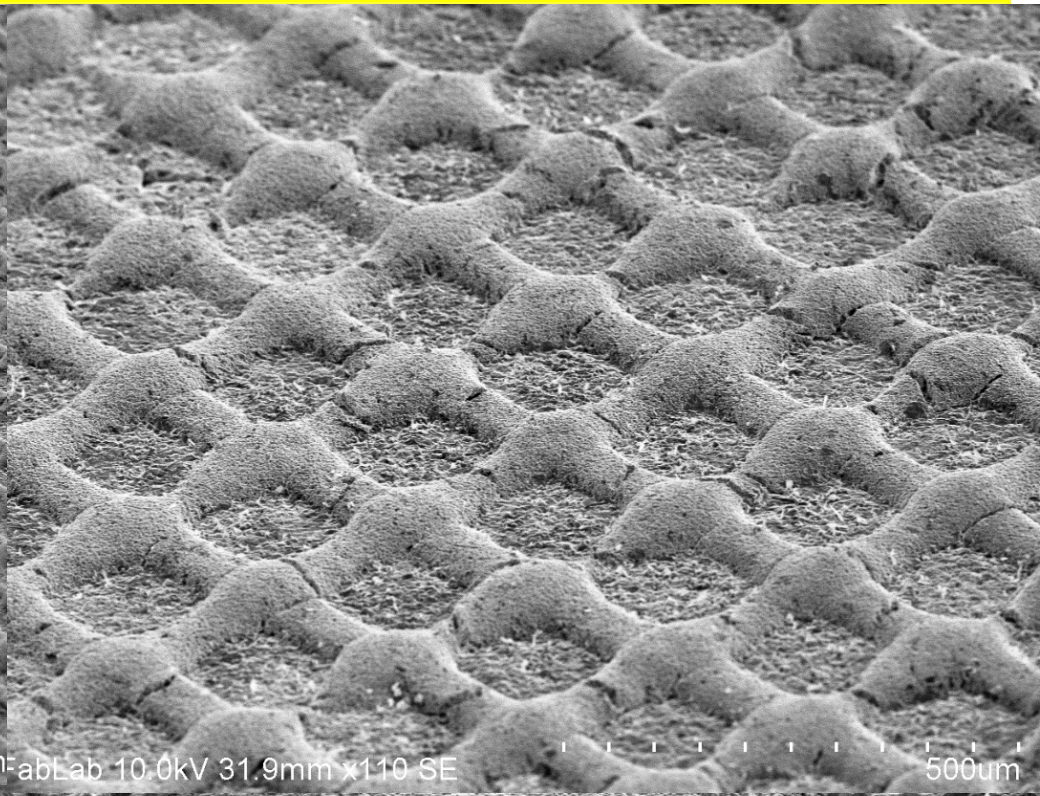




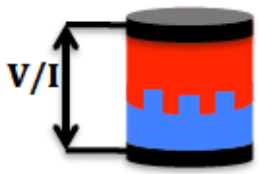
- Using to create controlled interface structures with varying geometries and aspect ratios
- Determining effect of interfacial layers between these structured interfaces

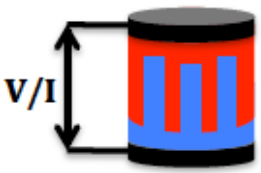
Extending to 3D Interface Structures

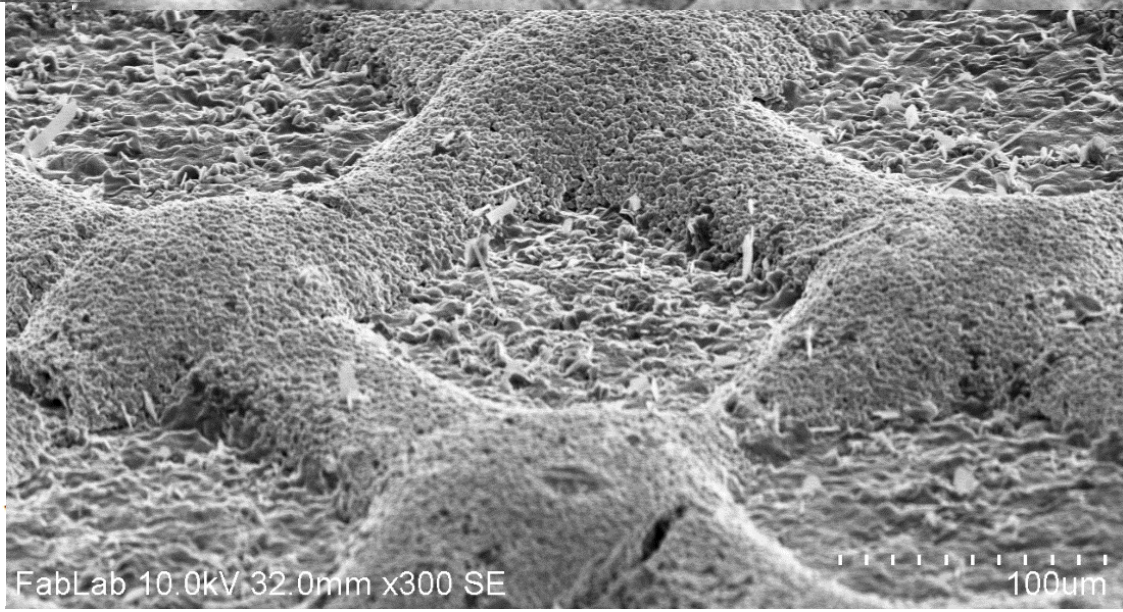




- Fabricating controlled garnet interface structures







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Conclusions/Summary

- Developing first principles computational approach to understand garnet/electrode stability and interfacial impedance
- Developed garnet surface treatment that allowed Li-metal wetting and overcame Li-metal/garnet interfacial impedance
- Developed stable high conductivity gel electrolytes and demonstrated their ability to significantly reduce cathode-garnet interfacial impedance
- Fabricating controlled 3D garnet structures to determine effect of interface architecture

Acknowledgement

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Tien Duong, Christopher Johnson, Yi Ding