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Effect of Anode Prelithiation on LEO Cycling Life of Graphene Batteries

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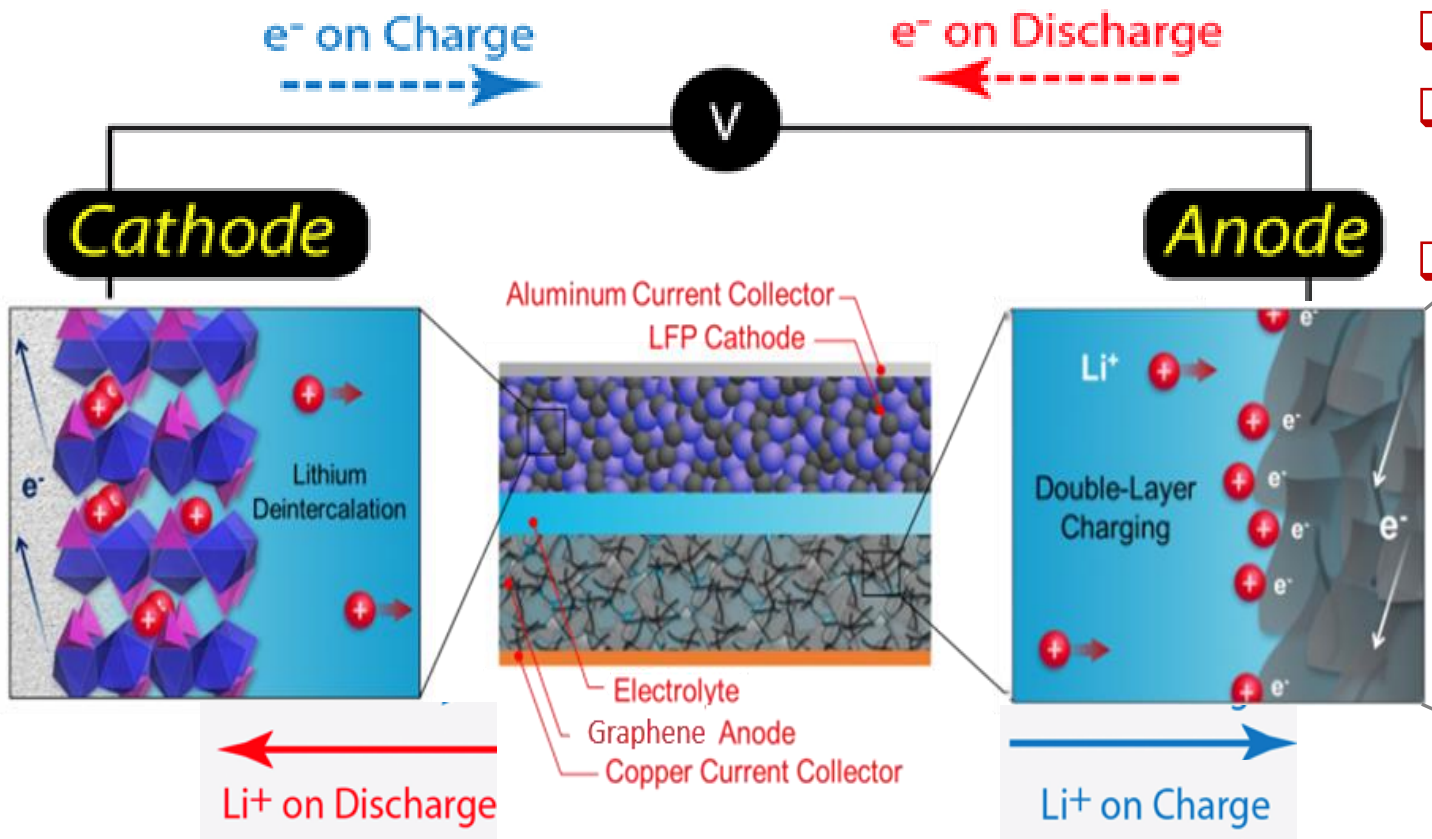
Batteries for Space Applications

- ❑ Critical role in satellite operation
- ❑ Low-earth orbit (LEO) satellites: batteries supply power during eclipse

Space batteries requirements:

- ❑ *High energy density*
- ❑ *Charge and discharge rates:* LEO satellites circle the earth in 90 minutes; eclipsed for 35 minutes
- ❑ *Lifetime:* 5,000 cycles per year
- ❑ *Depth of Discharge (DOD):* limited to low levels to reduce stress
- ❑ *Weight:* typically 10 to 20% of the overall mass

Lithium-Ion Batteries (LIBs)



Benefits of Graphene

- ❑ Graphite is the main anode material in LIBs
- ❑ Graphene has 2X the capacity of graphite

Anode	Capacity, mAh/g
Graphite (LiC_6)	372
Graphene (LiC_3)	744

Graphene



Graphene Benefits

- ❑ Specific capacity $\sim 1264 \text{ mAh g}^{-1}$
- ❑ Record in-plane chemical diffusion coefficients for Li at room temperature $7 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$ vs. $10^{-7} - 10^{-6} \text{ cm}^2 \text{ s}^{-1}$ in graphite

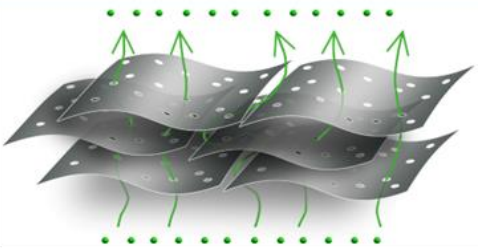
Technical Challenges

- Graphene re-stacks during electrode preparation
- Irreversible Li insertion, capacity loss with cycling
- Dendrite growth

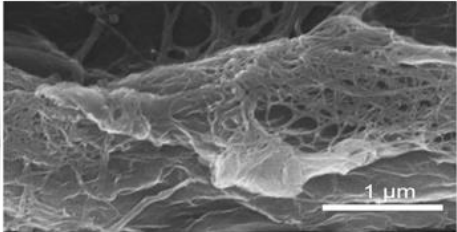


Graphene Materials for Next Generation LIBs

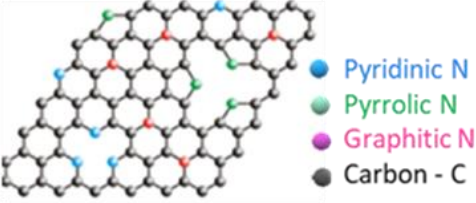
Graphene Materials



hybrid graphene - carbon nanotube

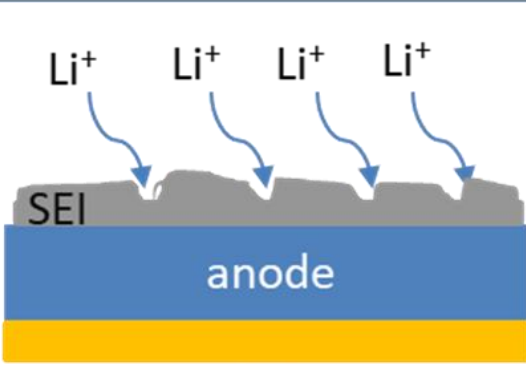


added lithiophilic sites



- Pyridinic N
- Pyrrolic N
- Graphitic N
- Carbon - C

Prelithiation

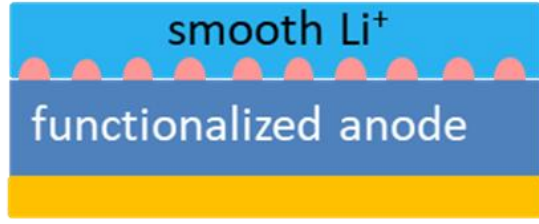


Li⁺ Li⁺ Li⁺ Li⁺

SEI

anode

dendrite free



smooth Li⁺

functionalized anode

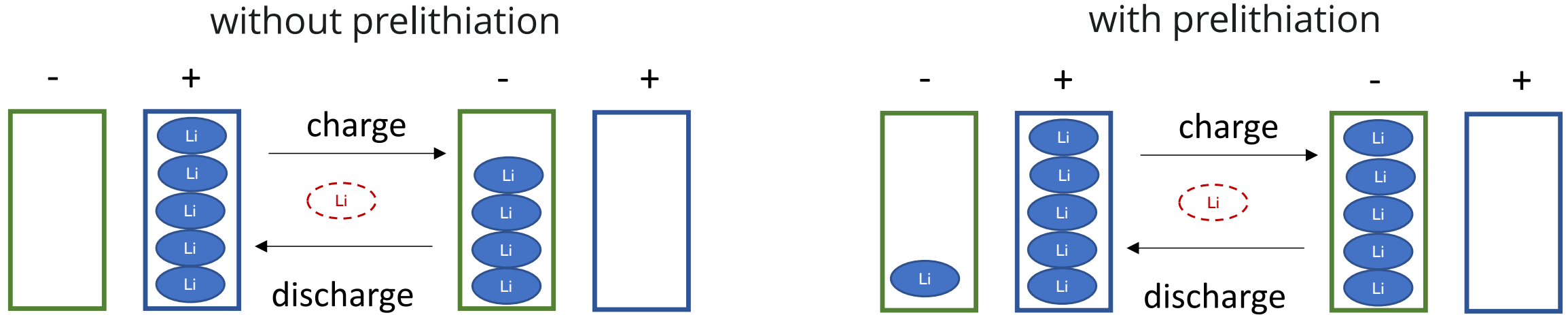
Next Generation Lithium Ion Battery



- high energy density
- light weight
- safe dendrite free
- high cycling stability
- long calendar life
- conformal design
- scalable manufacturing

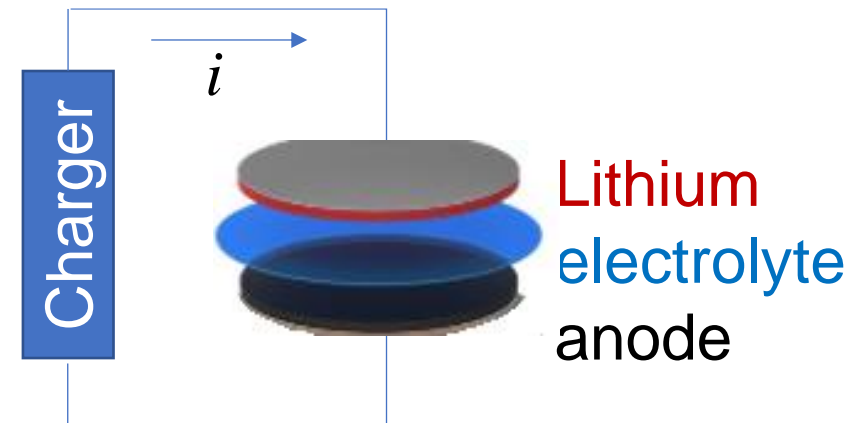
Prelithiation of Graphene Anodes

LIBs Operation



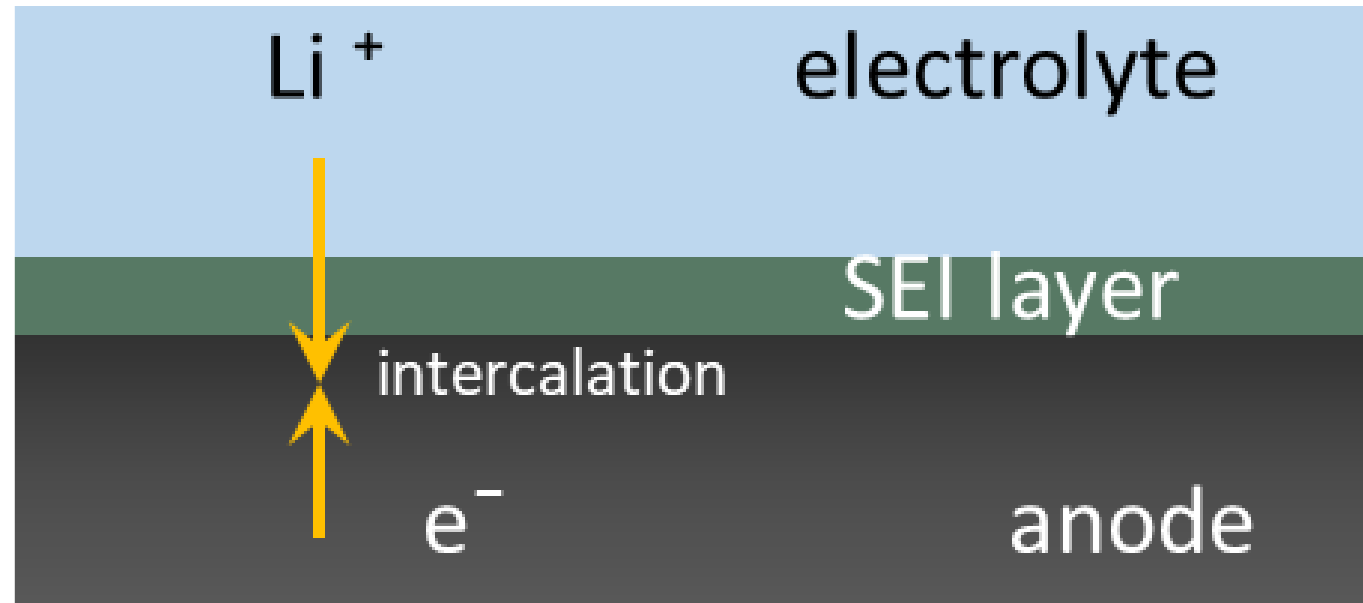
Electrochemical Prelithiation

- ❑ Compensates initial capacity loss
- ❑ Raises working voltage
- ❑ Decreases electrolyte consumption



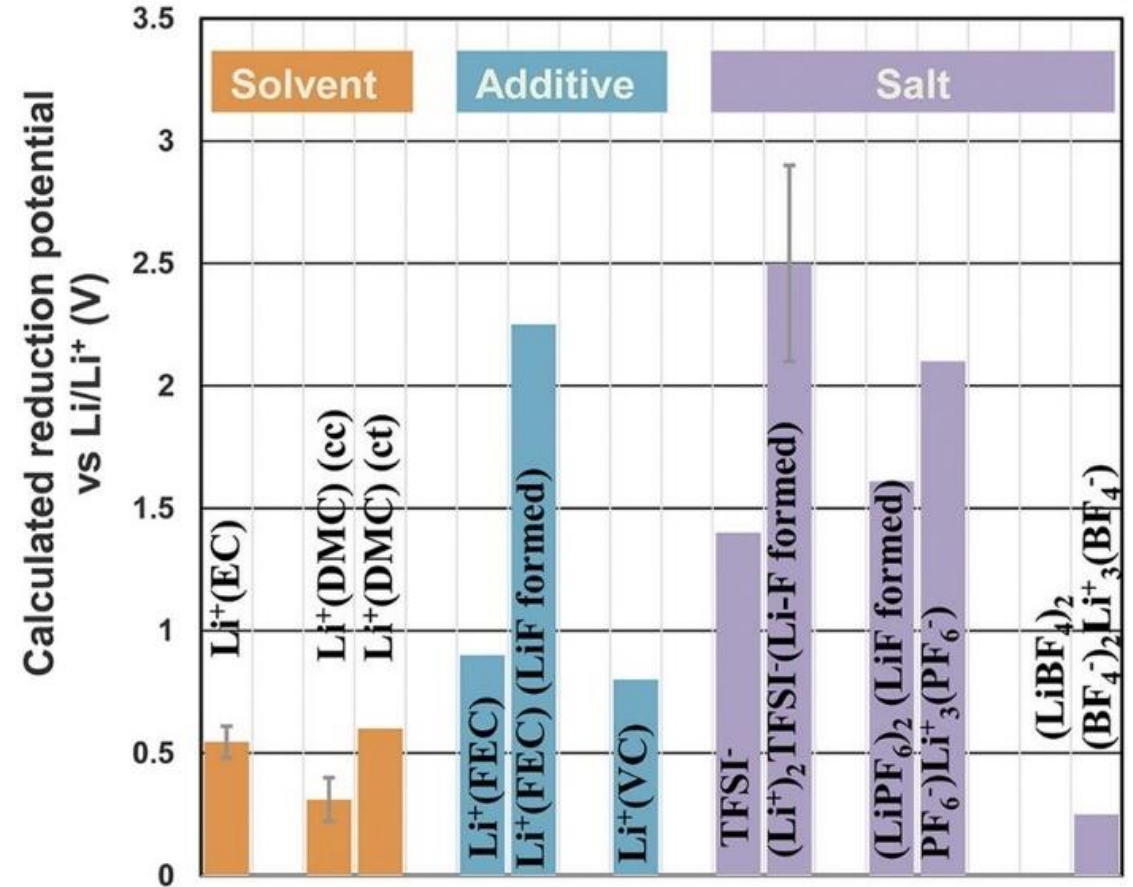
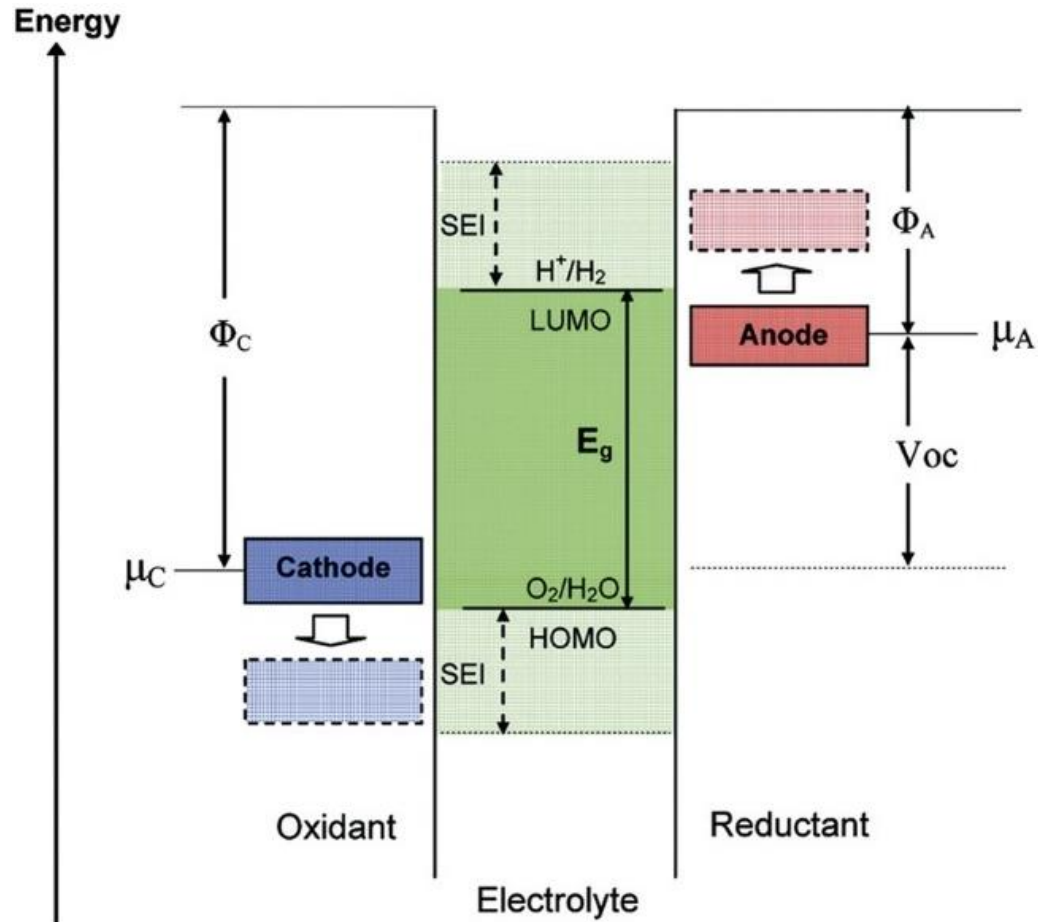
Role of Electrolyte in Prelithiation Process

Solid Electrolyte Interface (SEI)



- Cyclability of the anodes is affected by the structure and composition of the formed SEI
- Electrolyte determines the formed SEI

Electrolyte Compositions for Graphene Batteries



Reduction order:
 EC > DMC > VC > FEC > EMC > DEC

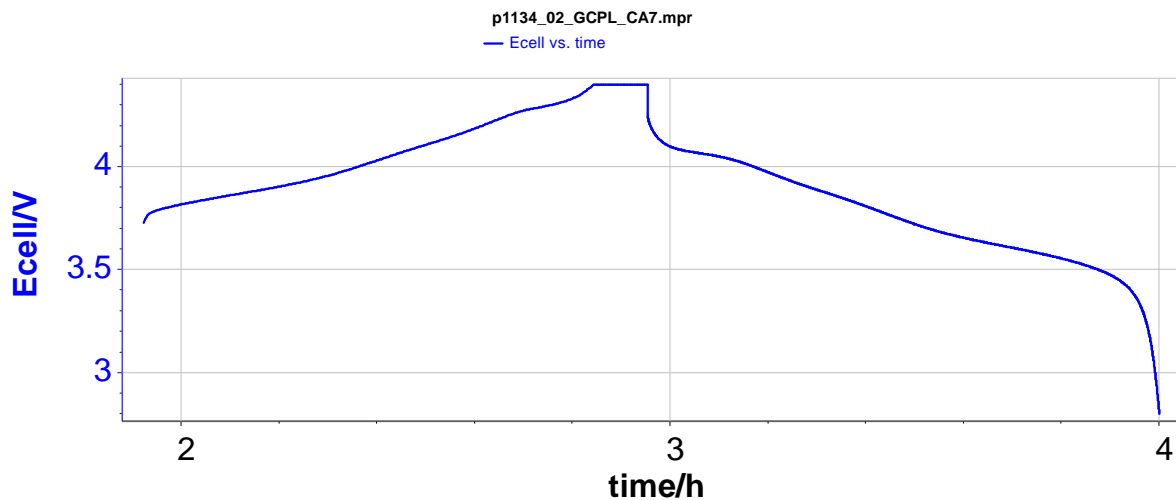
Goodenough and Kim, J. Power Sources 2011, 196, 6688.
 Delp et al., Electrochimica Acta 2016, 209, 498.

Electrolyte Compositions for Graphene Batteries

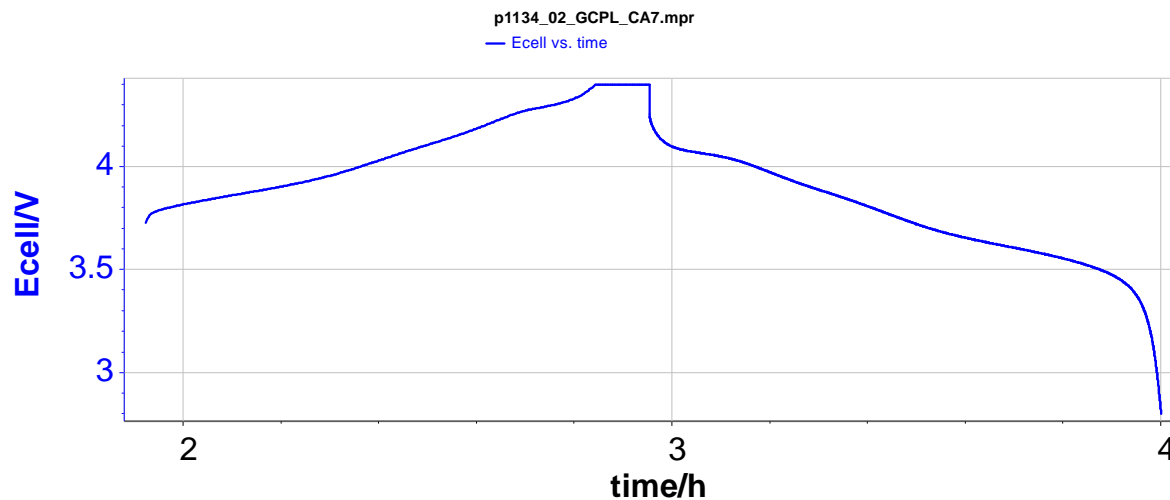
Electrolyte	LiTFSI	Vinylene (%)	Li salt (%)	FEC (%)
1				
2	1M			
3	1M	5%		
4	1M	5%	5%	
5	1M	5%	10%	
6	1M	5%	10%	5%

Pouch Cells with Prelithiated Graphene Anodes

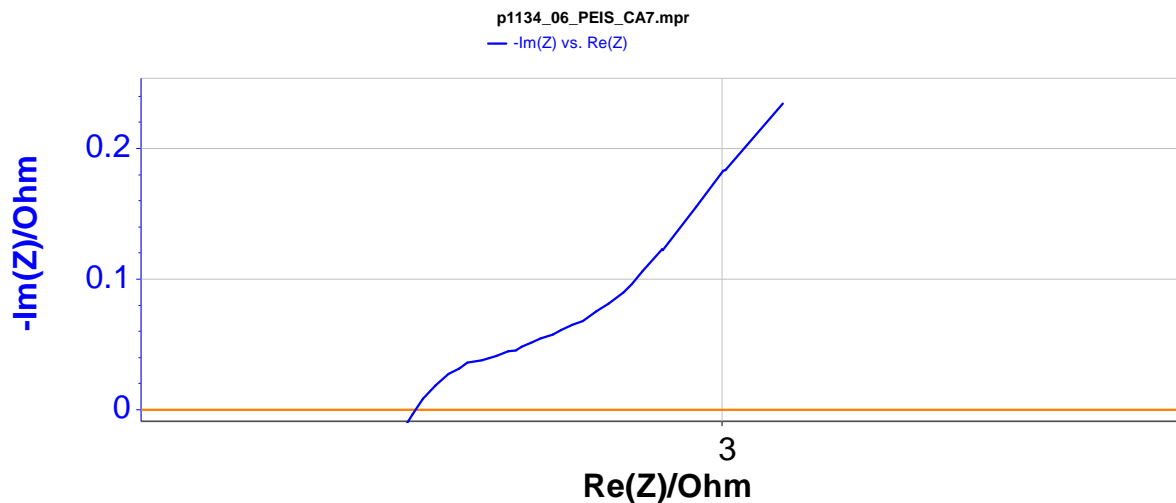
1C E = 300 Wh/kg



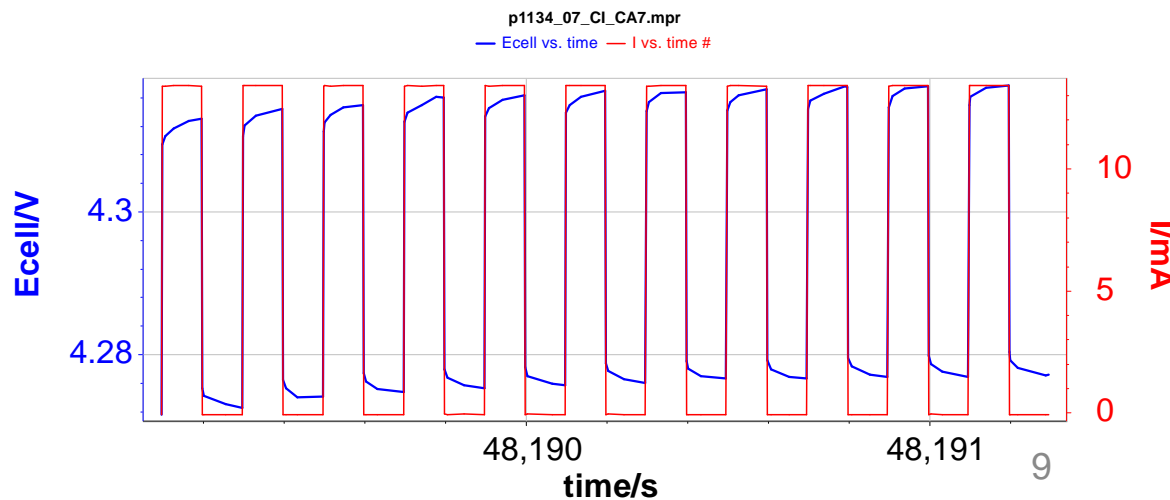
C/2.25 E = 318 Wh/kg C=33.4mAh



Impedance 2.25 Ohm

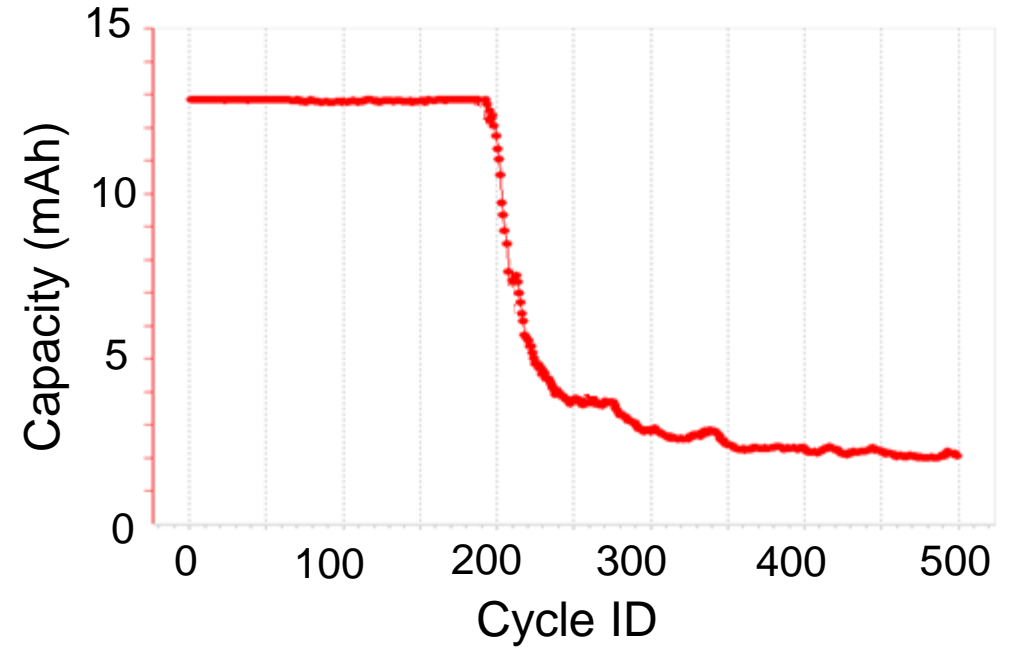
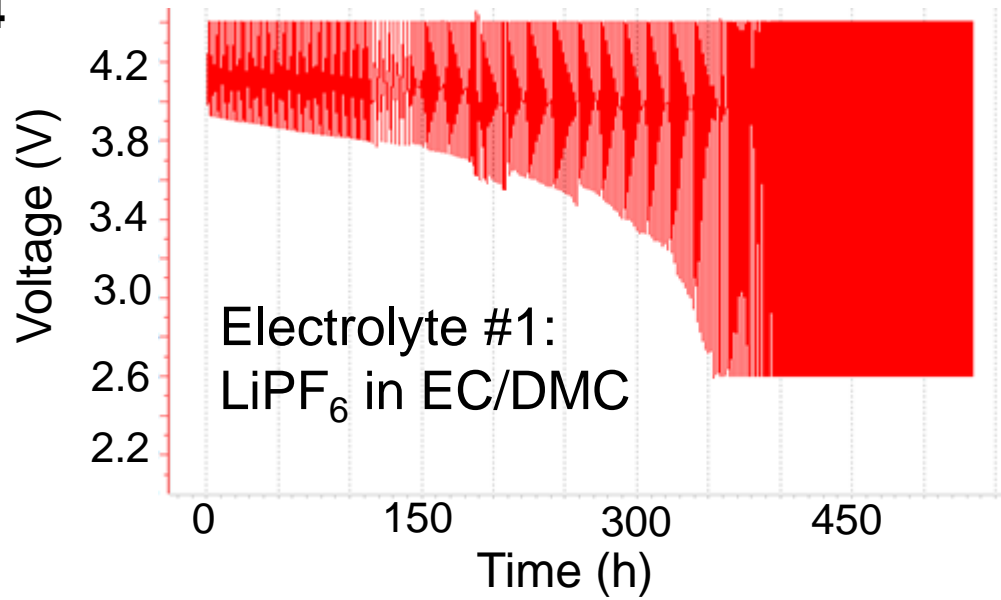


DCR Rc=2.77Ohm

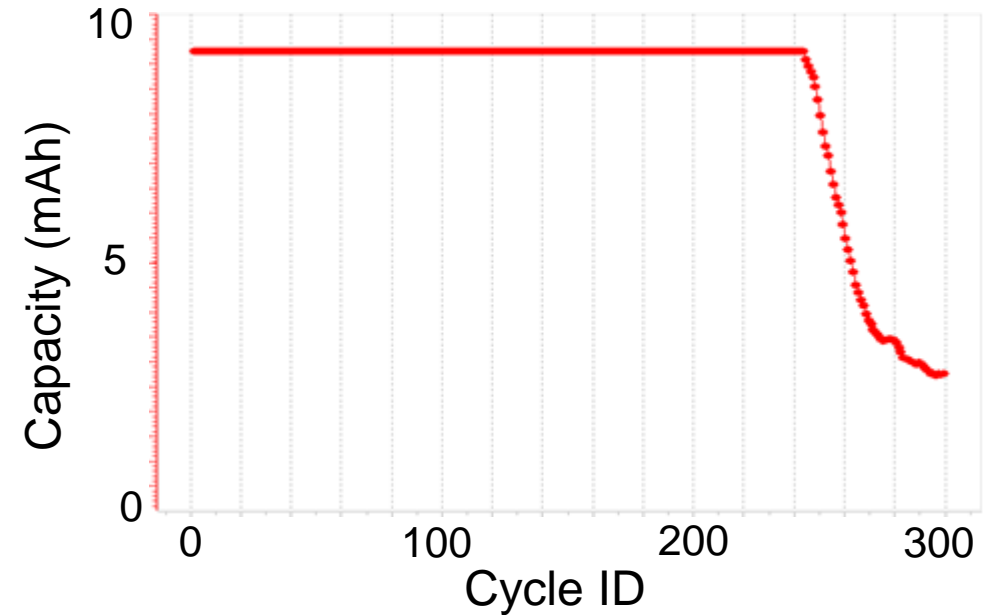
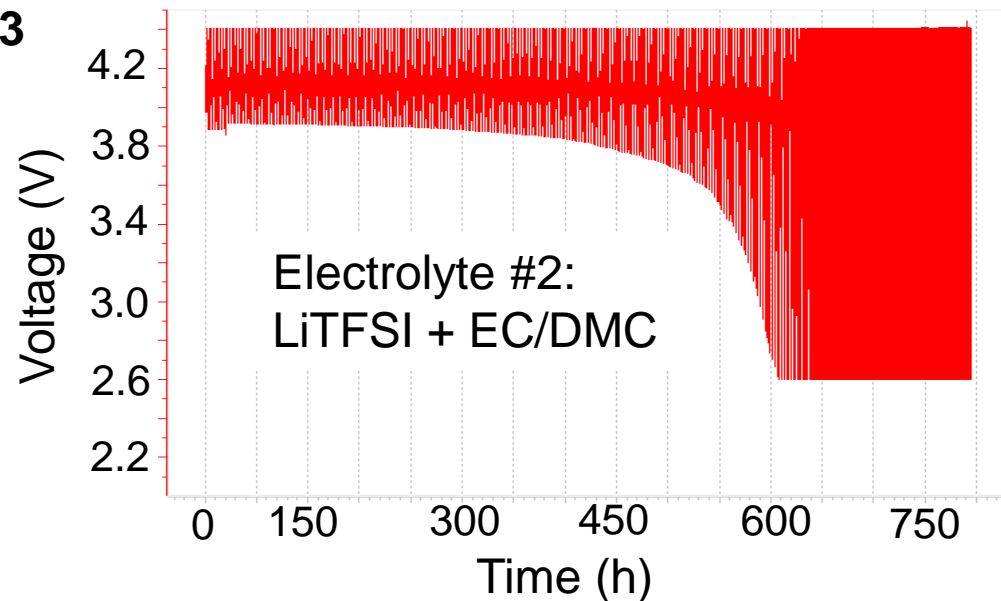


Effect of Electrolyte on LEO Cycling at 40% DoD

p1134

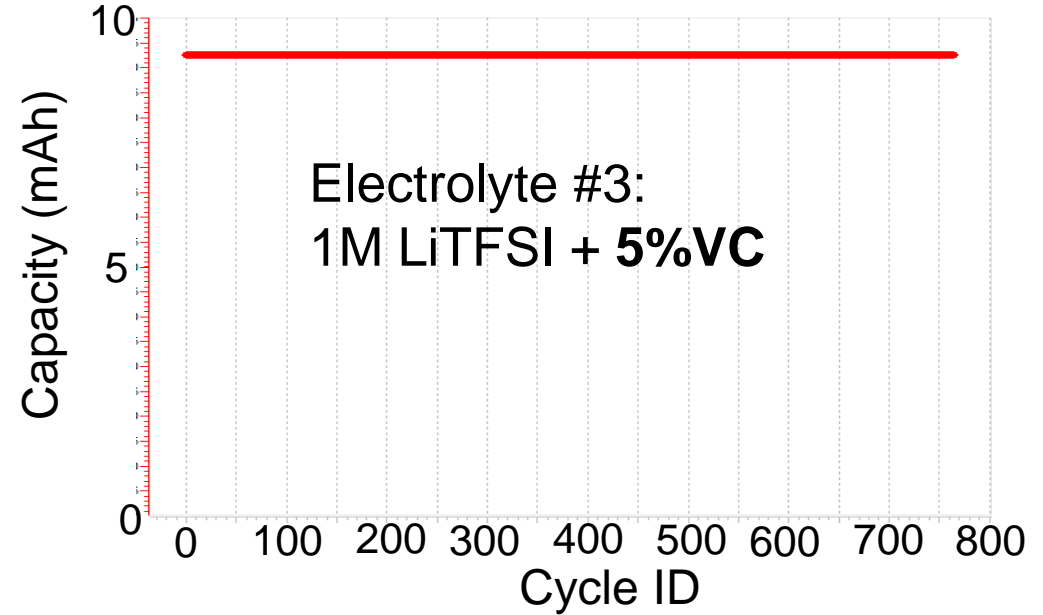
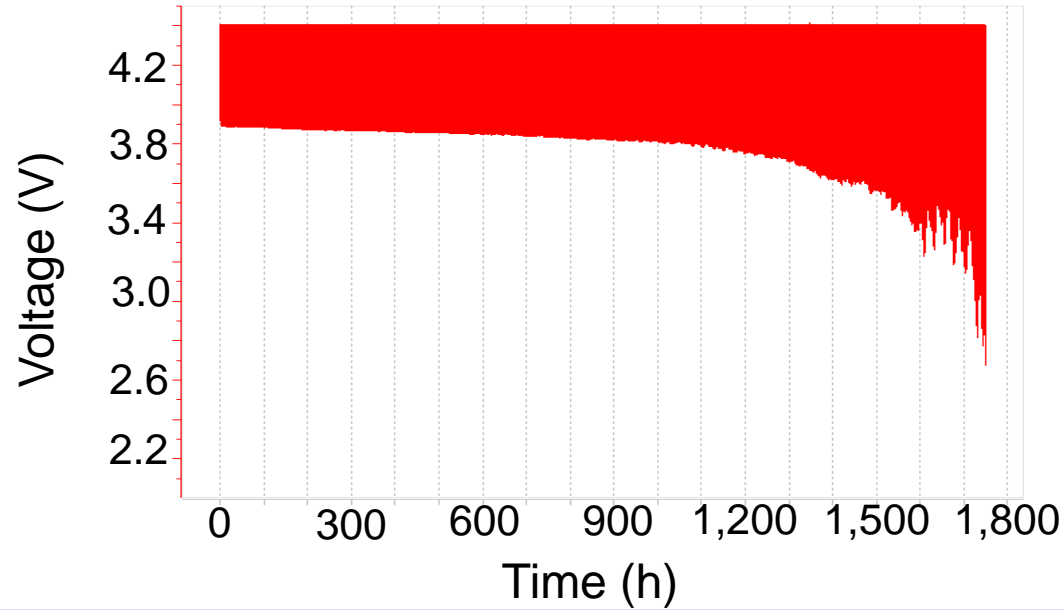


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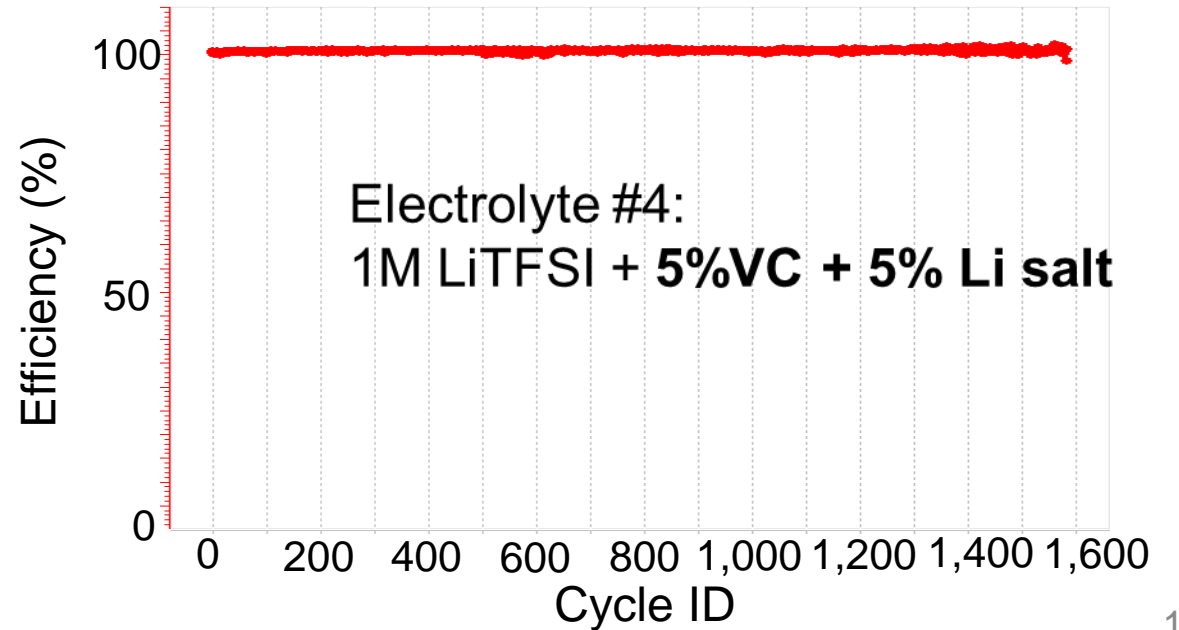
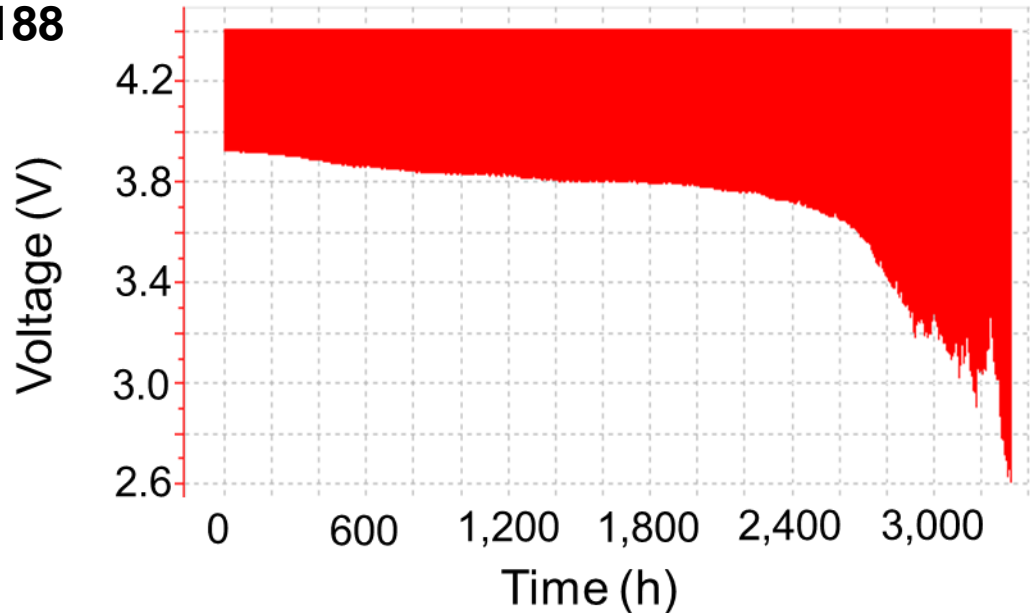


Effect of Electrolyte on LEO Cycling of Graphene Batteries

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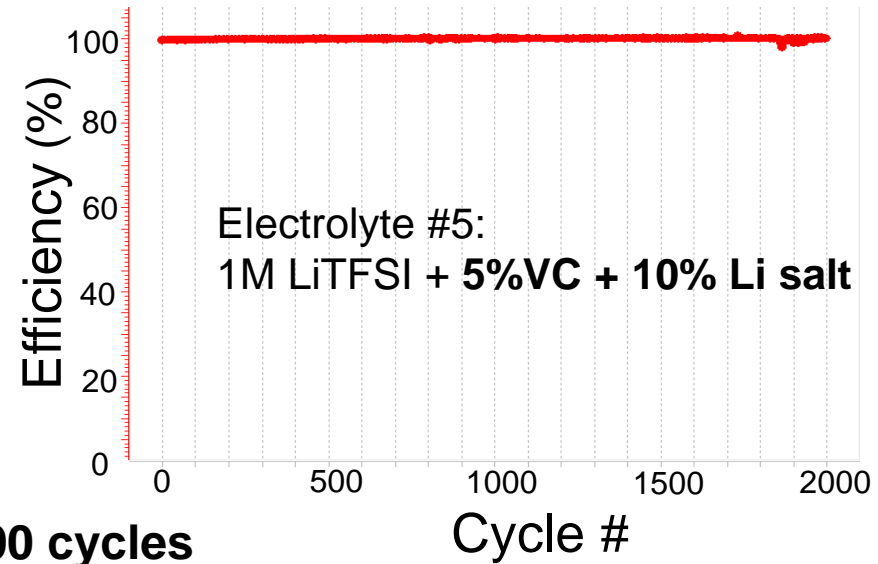
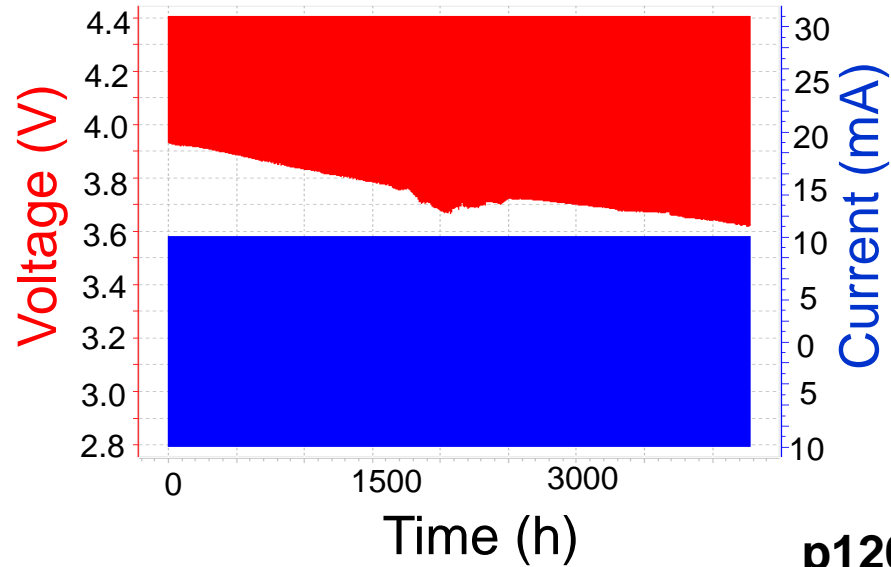


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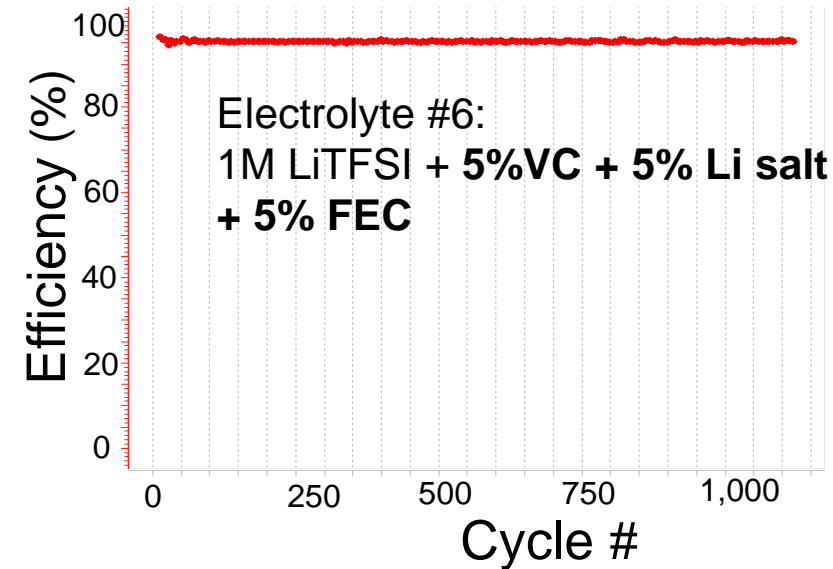
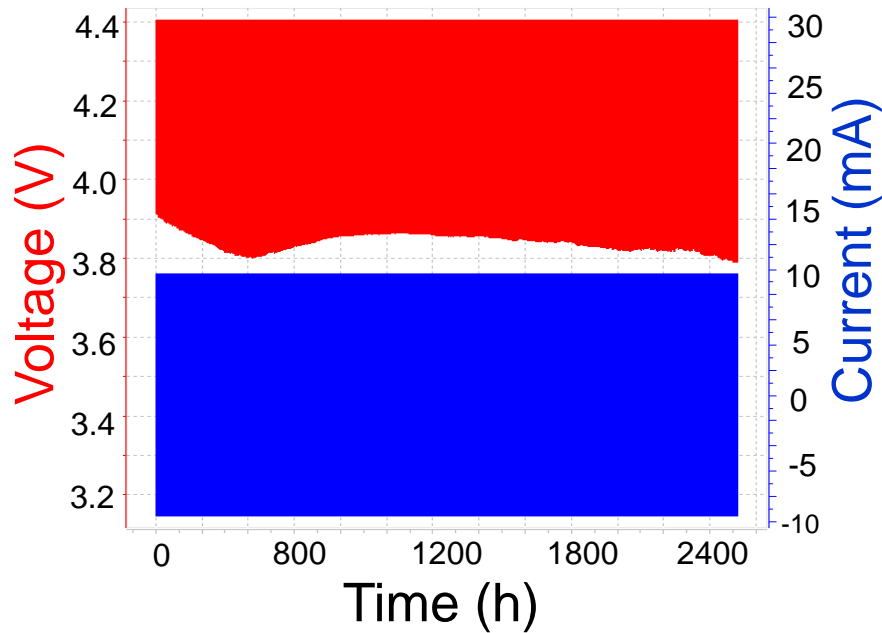


Effect of Electrolyte on LEO Cycling of Graphene Batteries


p1191 2,000 cycles



p1204 1,500 cycles



Effect of Electrolyte on LEO Cycling Stability

Electrolyte	LiTFSI	Vinylene (VC) %	Li salt %	FEC %	Cells	LEO Cycles	Cycling Stability
1					p1134	189	
2	1M				p1163	245	
3	1M	5%			p1168	765	
4	1M	5%	5%		p1188	1585	
5	1M	5%	10%		p1191	2000+	
6	1M	5%	10%	5%	p1204	1500+	

Li salt in combination of additives such as VC and FEC form stable SEI increasing the cycling stability of the graphene-based anodes.



Summary

- Graphene-based batteries with high energy density > 350 Wh/kg and Coulombic efficiency of $> 95\%$ using prelithiated graphene anodes
- The composition of the electrolyte plays critical role in the stability of the battery cycling performance
- A combination of Li salts and carbonate additives can be used to form a high-performance stable SEI and regenerate the prelithiated graphene surface during LEO cycling
- The graphene batteries have stable performance for more than 2000 cycles at 40% DOD with capacity retention of 100 % and stable resistance



Acknowledgement

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TPOC Alec Jackson