Towards the Practical Application of Lithium Sulfur Batteries: Electrochemistry and Chemistry

Jie Xiao, Jianming Zheng, Dongping Lv, Terence Lozano, Qiang Wang, Eric Walter, Yuyan Shao, Natalio T. Saenz, Ji-Guang Zhang, Wendy Bennett, Gordon Graff and Jun Liu

Pacific Northwest National Laboratory

9th US – CHINA
Electric Vehicle and Battery Technology WORKSHOP
Seattle, August 18-19, 2014

Pacific Northwest
NATIONAL LABORATORY

Proudly Operated by Battelle *Since 1965*

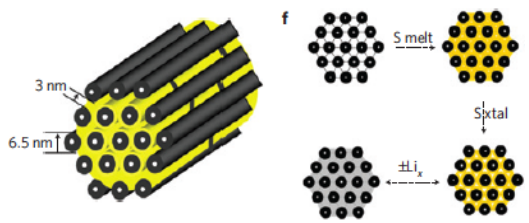
Outline

- Literature overview on Li-S batteries: challenges, approaches and problems
- Revisit Li-S system in a relevant scale
- Reaction mechanism in Li-S batteries: electrochemical/chemical reactions
- Summary

Pacific Northwest
NATIONAL LABORATORY
Proudly Operated by Battelle Since 1965

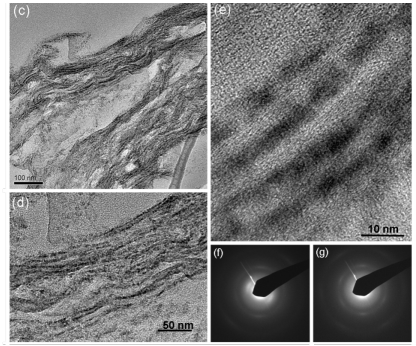
Literature Overview: Cathode

CMK-3/S composite



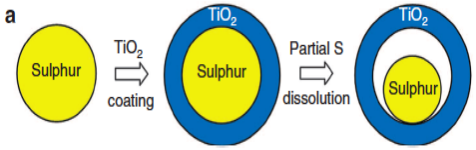
L.F. Nazar et al., Nat. Mater., 2009, 8, 500.

Nafion-coated graphene/S (PNNL)



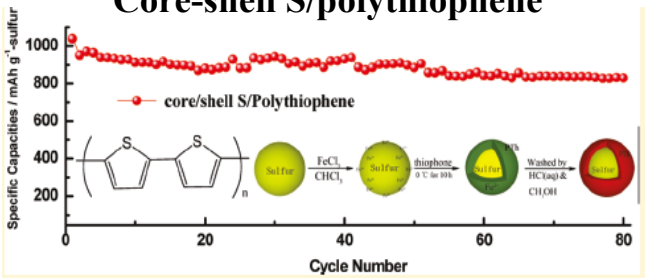
Y. Cao et al., Phys. Chem. Chem. Phys., 2011, 13,7660.

Yolk-shell S/TiO_2



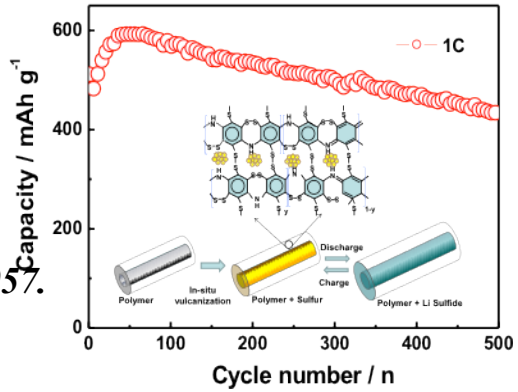
Y. Cui et al., Nat .Commun., 2013, DOI: 10.1038/ncomms2327

Core-shell S/polythiophene



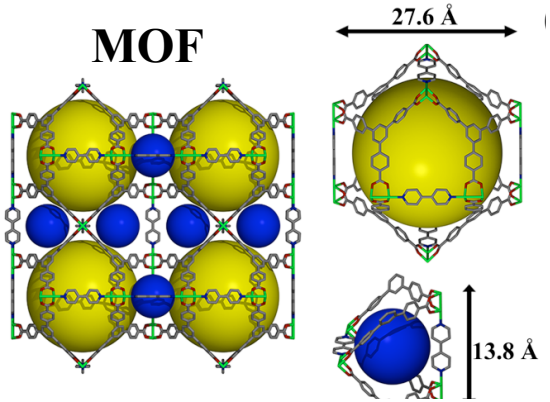
F. Wu et al,J.Phys.Chem.C,2011,115,6057.

Self-breathing PANI/S(PNNL)



L. Xiao et al., Adv. Mater., 2012,24, 1176.

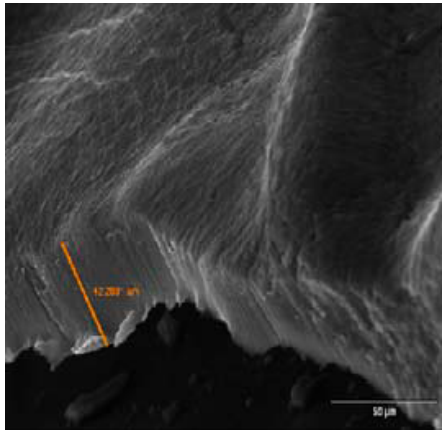
Lewis acid-base reactions (PNNL)



J. Zheng et al., Nano Lett. 2014, 14, 2345.

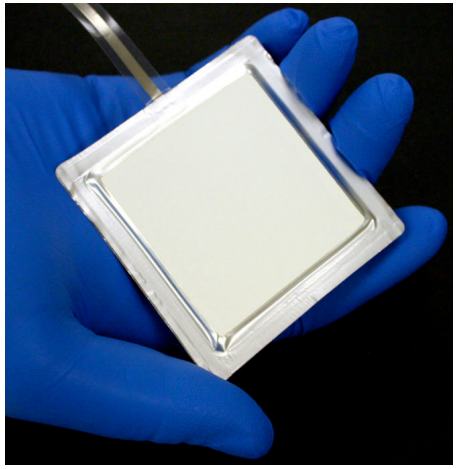
Literature Overview: Anode

Sion Power



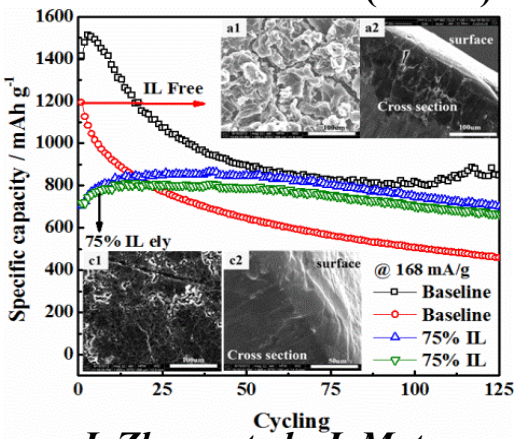
J. Affinito et al., ECS Transactions, 2010, 25,23.

Polyplus



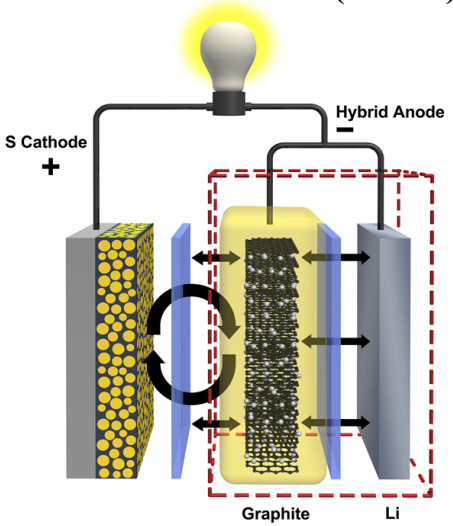
Courtesy of www.hybridcars.com

IL-enhanced SEI (PNNL)



J. Zheng et al., J. Mater. Chem. A, 2013, 1, 8464.

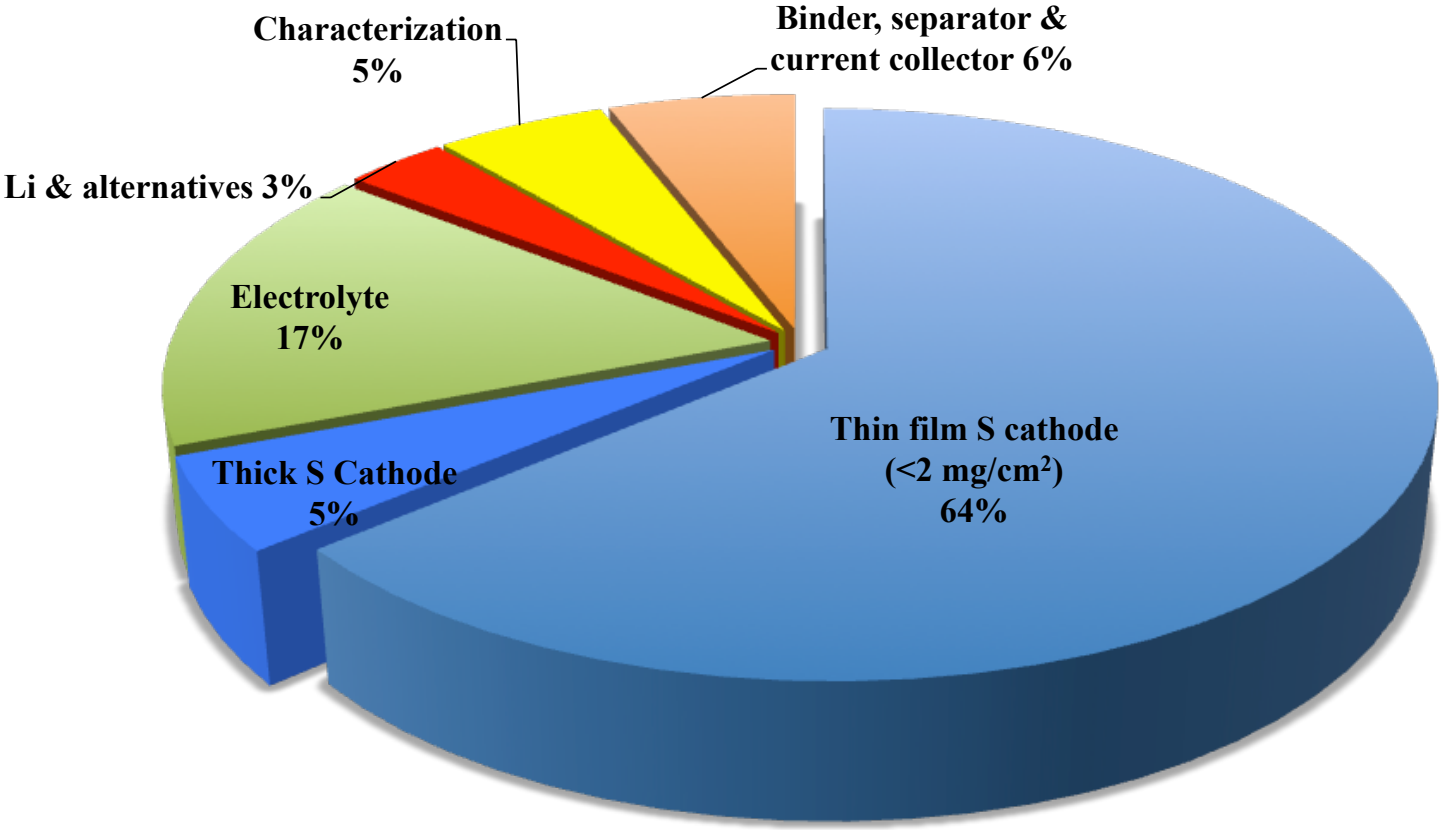
Artificial SEI (PNNL)



C.Huang et al., Nat. Commun. 2014, DOI: 10.1038/ncomms4015

Literature Distribution for Li-S

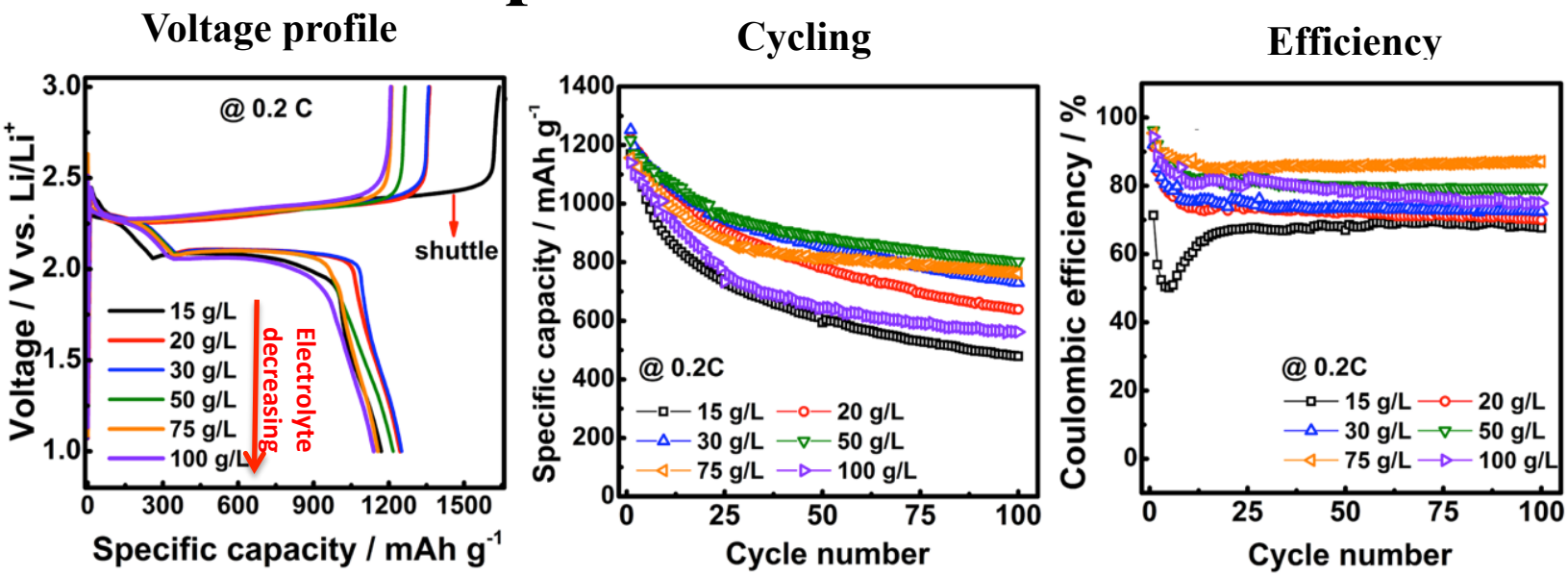
Updated in June, 2014



- Thin film electrodes: Good platform to understand nano-scale phenomena
  - Reproducibility?
  - Good baseline electrode?
  - Knowledge gathered still usable in thick electrode?

Pacific Northwest NATIONAL LABORATORY

Sulfur/Electrolyte Ratio is the Key for Reproducible Results



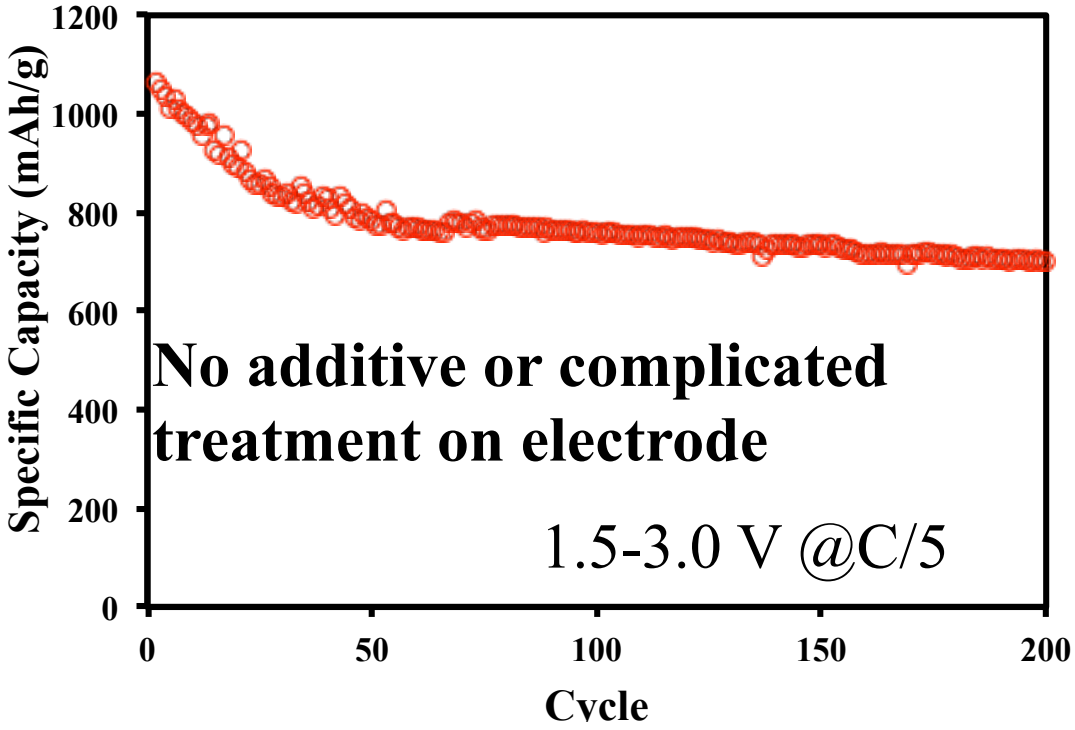
50 g/L = 1 mg S : 20 µL electrolyte

- Sulfur solubility, electrolyte viscosity and electrode wetting vary significantly in each cell.
- The optimized (balanced) ratio between sulfur/electrolyte is 50 g/L (for KB/S only).

Energies, ***2012***, *5, 5190. (ARL)*
J. Electrochem. Soc., ***2013***, *160, A2288. (PNNL)*
J. Power Sources, ***2014***, *249, 497. (Petr Novák)*

Pacific Northwest NATIONAL LABORATORY

S/KB Baseline Cathode

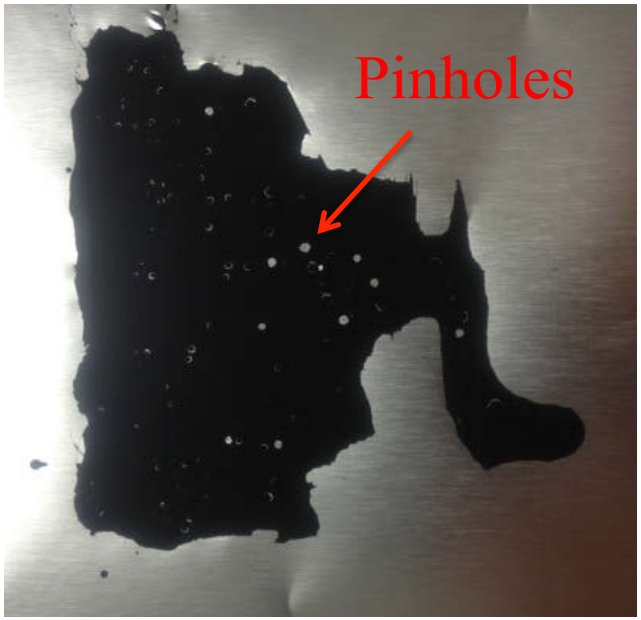


- A good S/KB baseline cathode is important.
  - 80% S in composite; >1 mg/cm^2 S loading for meaningful evaluation.
  - >700 mAh/g capacity after 200 cycles (>70% retention).

- Ensure the knowledge gained on baseline electrodes usable for thick electrodes as well.

**** PNNL unpublished results***

From Thin to Thick Electrodes



After modification

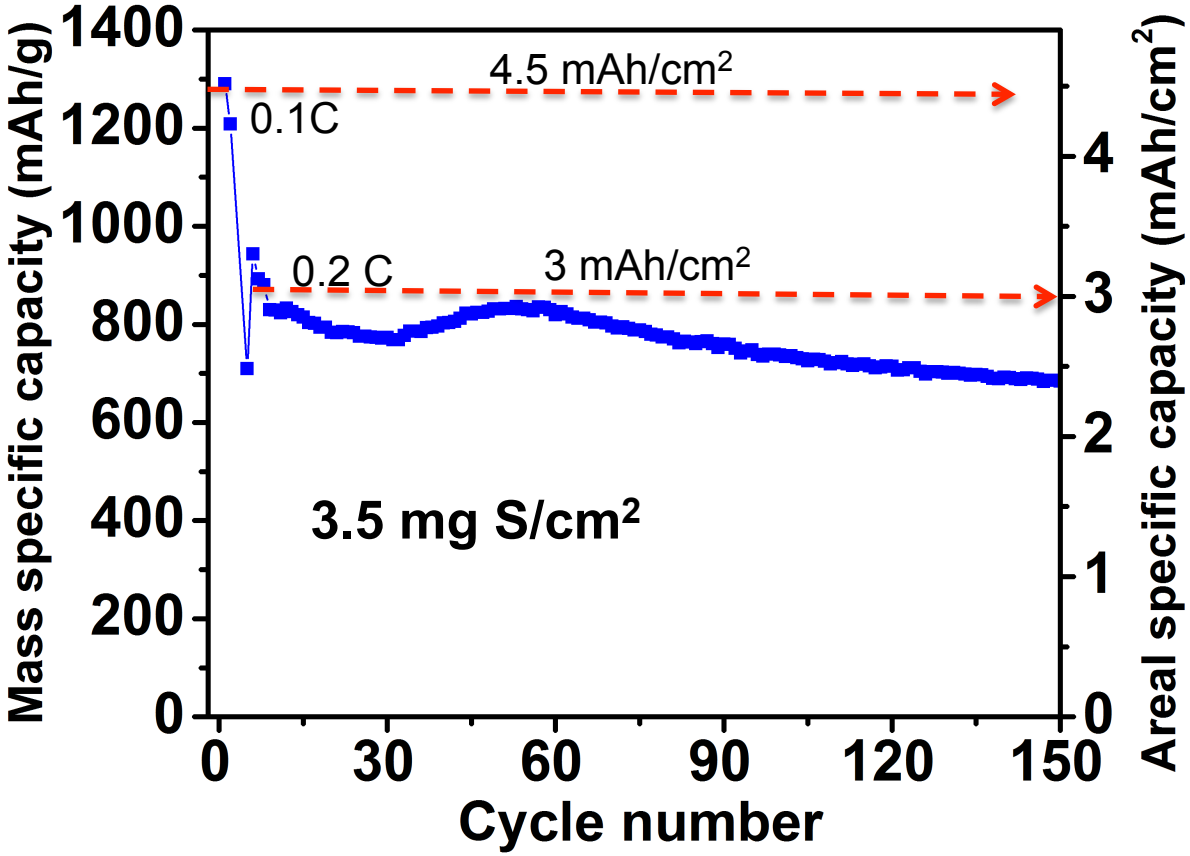
Binder: CMC+SBR

- Nano-sized particles are difficult to form thick and uniform coatings for large-area without pinholes.
- Adjustable sulfur loading: 2-8 mg/cm^2

**** PNNL patent application filed***

Pacific Northwest
NATIONAL LABORATORY

Thick Sulfur Cathodes Cycles Well



1C = 1000 mA/g
= 3.5 mA/cm²

- Stable cycling (150 cycles) was observed from thick electrodes.
- Rate capability and electrode wetting become poor.
- Lithium anode corrosion becomes the limiting factor.

PNNL unpublished results- Do not distribute

Pacific Northwest
NATIONAL LABORATORY