



Cathode engineering for limiting polysulfide shuttling in Lithium Sulfur batteries

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Presentation Overview

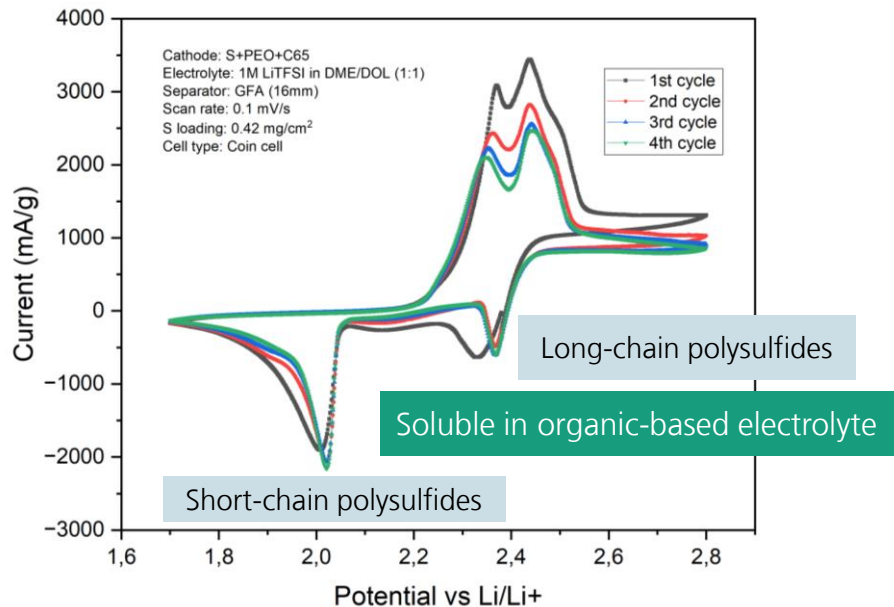
Outline

- 1. Switching Li–S from liquid chaos to solid control**
- 2. Our Approach**
- 3. Physico-chemical Characterization**
- 4. Electrochemical Results**
- 5. Conclusion & Outlook**

Switching Li-S from liquid chaos to solid control

Transition to solid phase polysulfide conversion

Discharge step: $S_8 \rightarrow Li_2S_8 \rightarrow Li_2S_6 \rightarrow Li_2S_4 \rightarrow Li_2S_2 \rightarrow Li_2S$



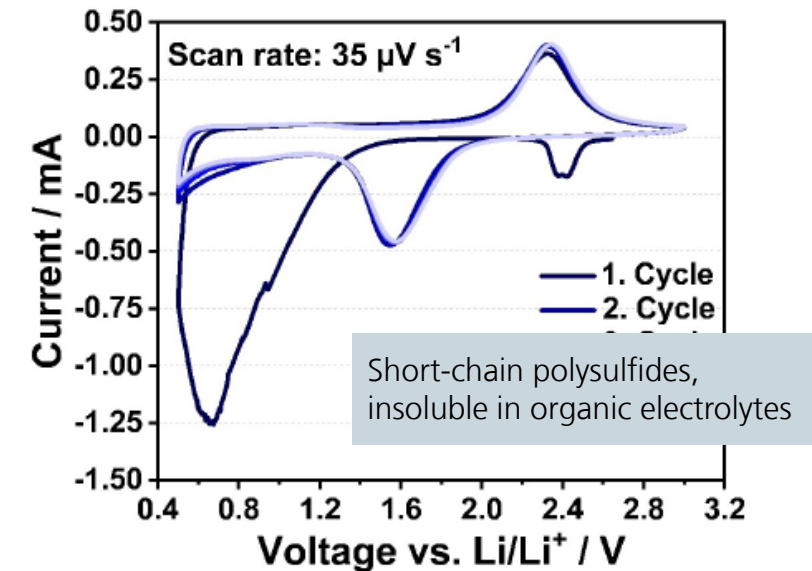
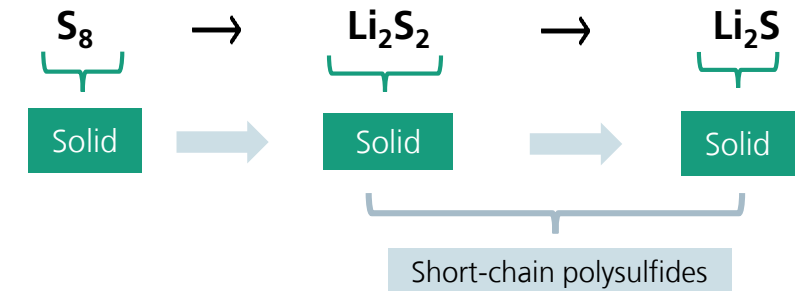
Polysulfide shuttling effect



Pore
confinement in
microporous
carbon materials

Formation of cathode
electrolyte interface

Solid phase conversion of polysulfides



Kensy et al., Bat. and Supercaps, 4, 612-622 (2021)



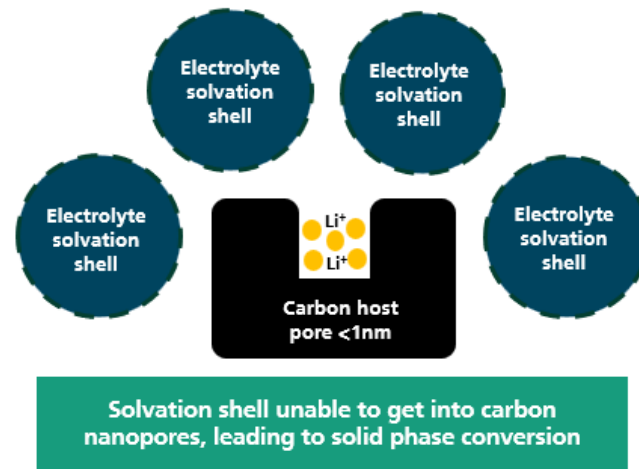
Our Approach



Our Approach

1

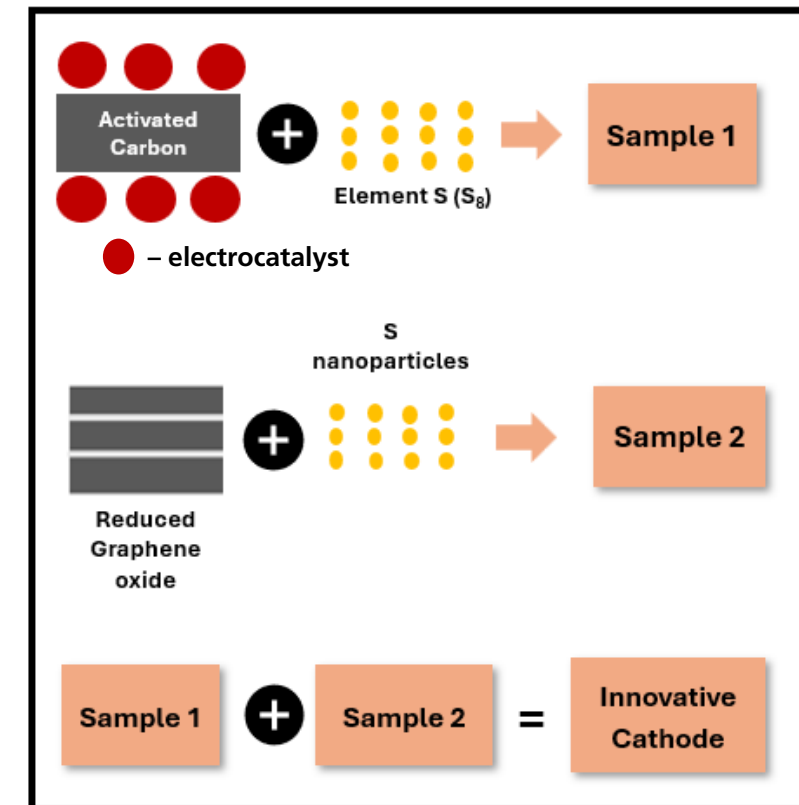
Immobilizing short-chain polysulfides in <1 nm carbon pores & cathode electrolyte interface formation for solid-phase conversion



2

Developing an electrocatalytic innovative carbon-sulfur + reduced graphene-sulfur cathode where (i) S and polysulfides are hosted to prevent shuttling, (ii) achieve high S loading and (iii) increase electronic conductivity

Overview of Research Direction



Our Approach

1

Varying carbon content in carbon + sulfur composite to attain efficient solid phase conversion

Screening and characterization of commercial carbon samples via BET analysis

Pore size distribution

Surface area

Melt infiltration of C/S composites – **C/S 55%:45%** and **C/S 60%:40%** – at 155°C for 21 hours; followed by ****TGA**

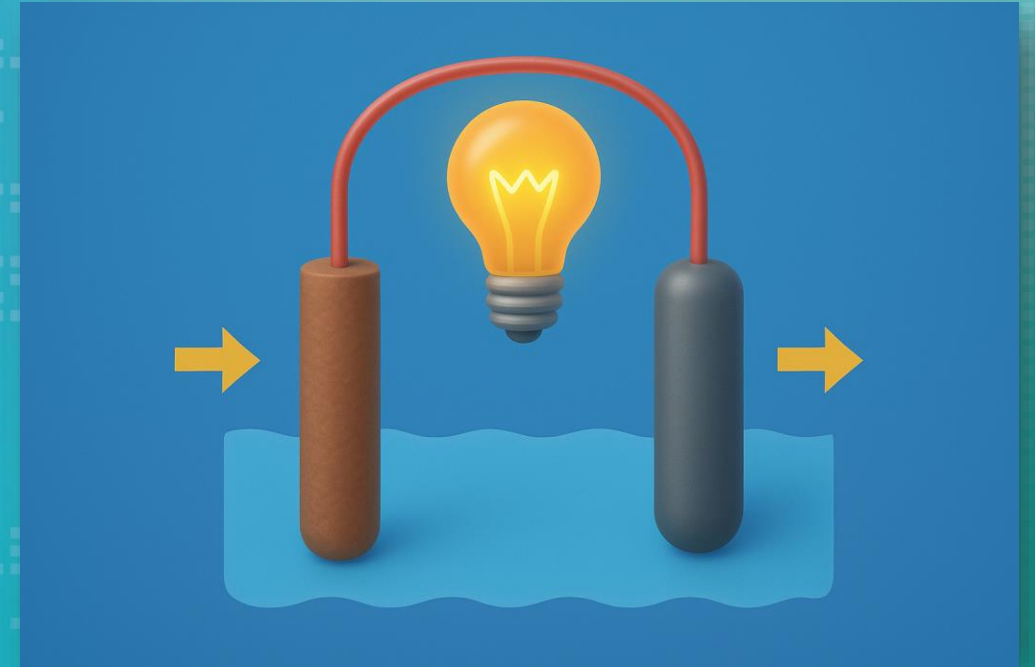
Effective S loading is ~52% for mesoporous carbon, microporous <50%
****Varying C content impacts packing density and interstitial spacing in electrode**

Electrode preparation: C/S comp. (90%) + PVDF (10%) + DMSO; followed by ****TGA**
****No C65**

Electrochemical performance assessment via CV, GCPL, Rate capability and EIS tests

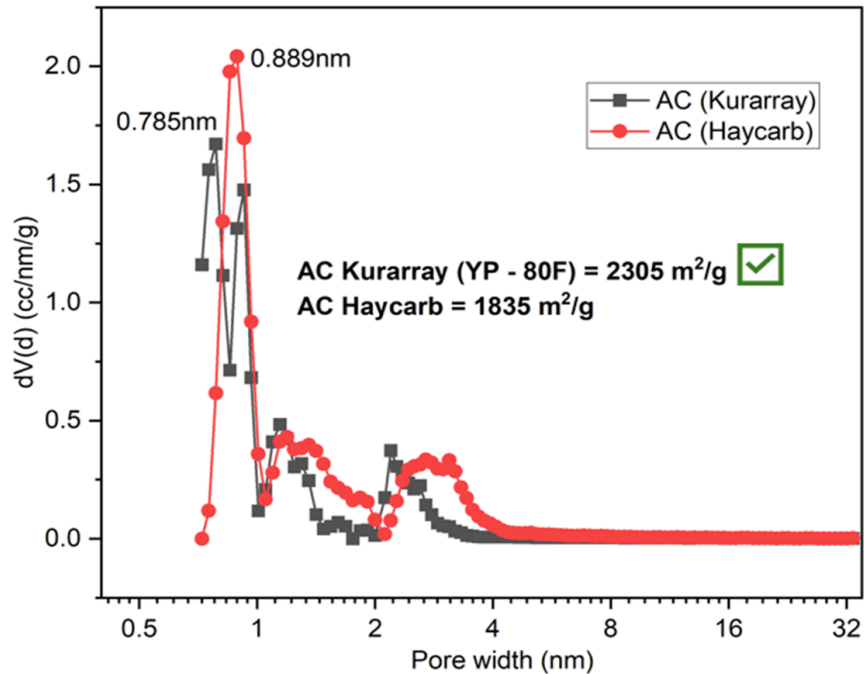


Physico-chemical characterization and Electrochemical Results



Physico-chemical Characterization

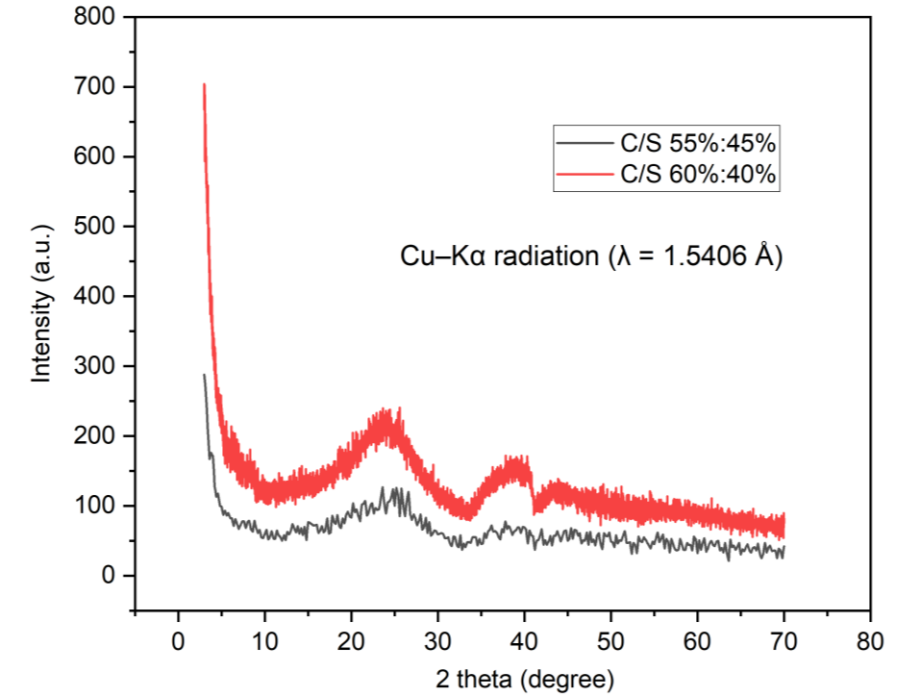
Key Findings



Pore size distribution and BET analysis of carbon samples

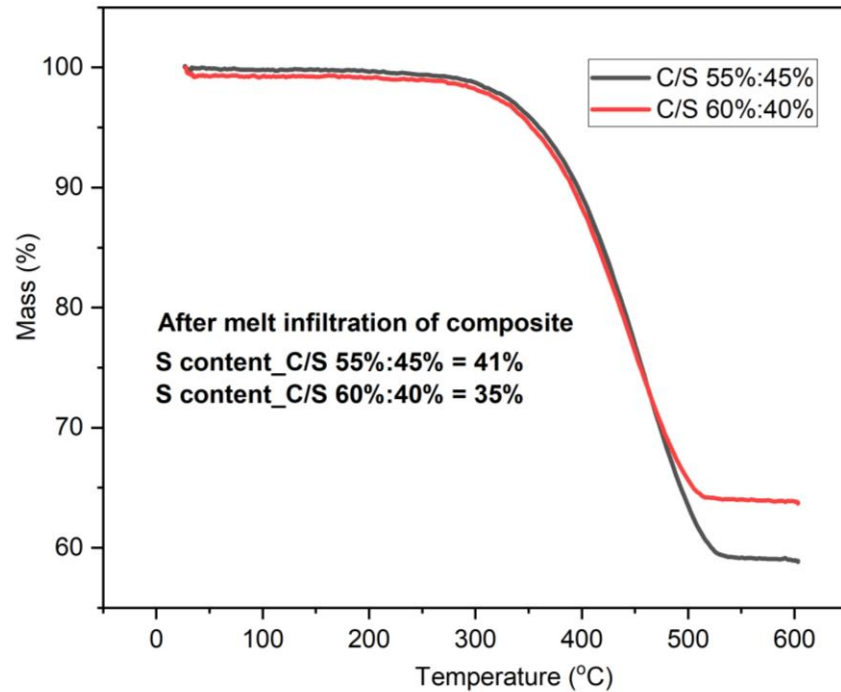
AC Kurarray with lower nanopores and higher surface area is selected as carbon host of choice

Effective melt infiltration with no elemental S₈ on the surface of AC



XRD of as-prepared carbon-sulfur composites

Physico-chemical Characterization



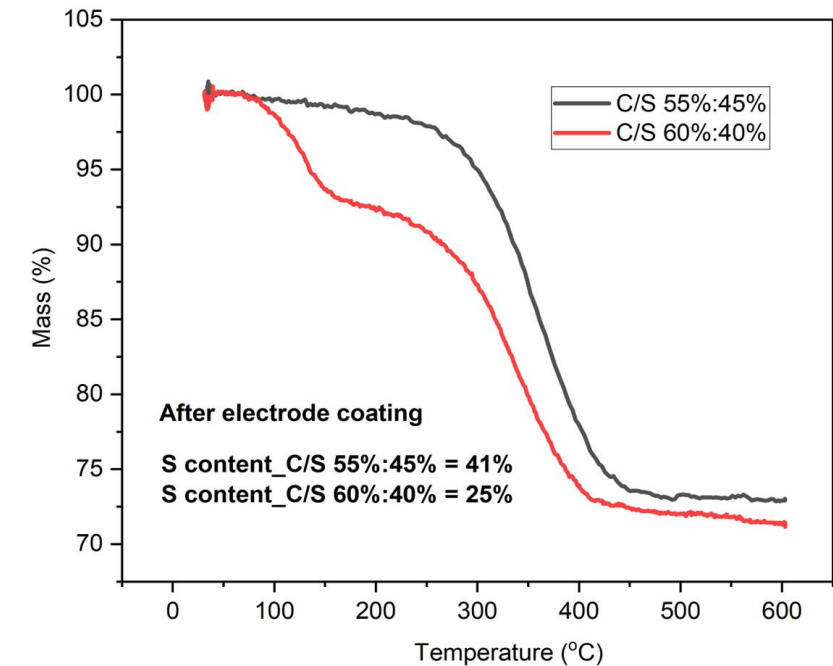
TGA of as-prepared carbon-sulfur composites

Key Findings

Limited packing density and higher interstitial spacing leads to limited S loading

Increased packing density of S limits S loss during melt infiltration

C/S 55%:45% = 8.9%
C/S 60%:40% = 37.5%



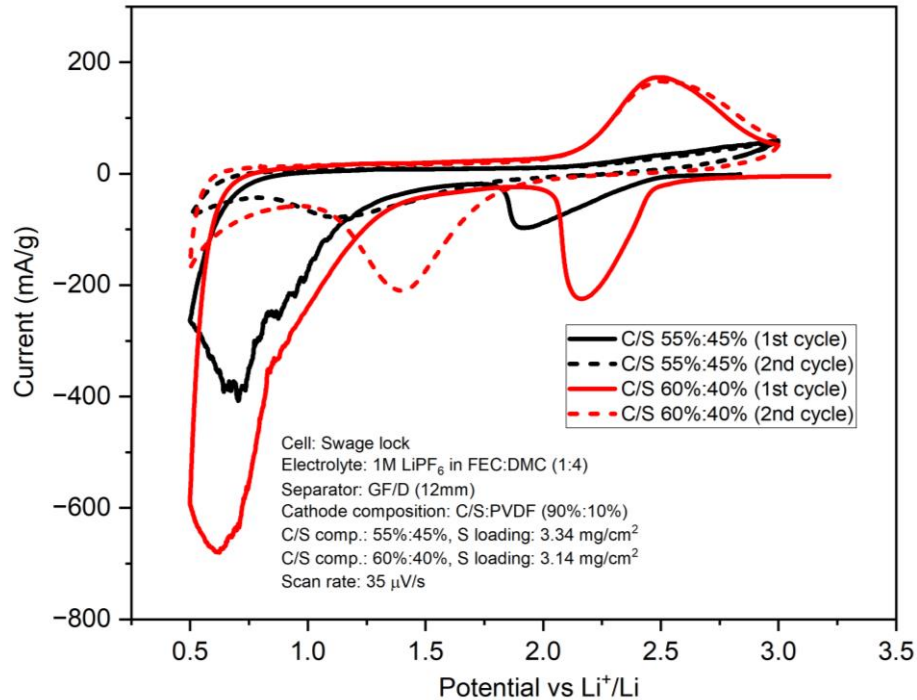
TGA of as-prepared carbon-sulfur electrodes

Electrochemical Results

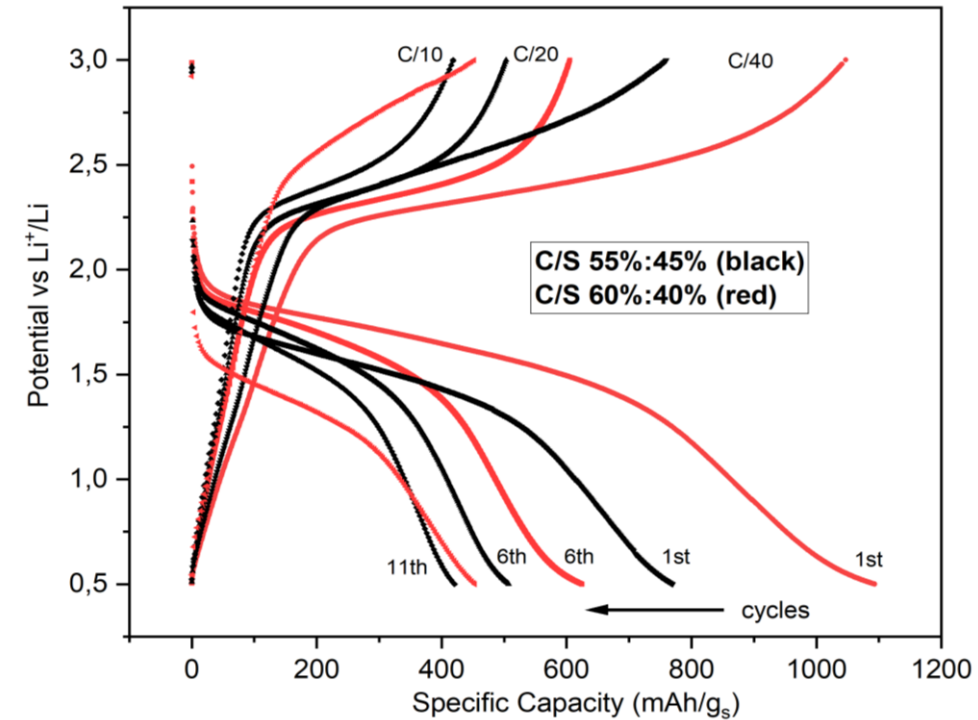
Key Findings

Formation of interphases such as solid electrolyte interface is prevalent with higher AC host content

Higher carbon content initiates fast surface limited reduction



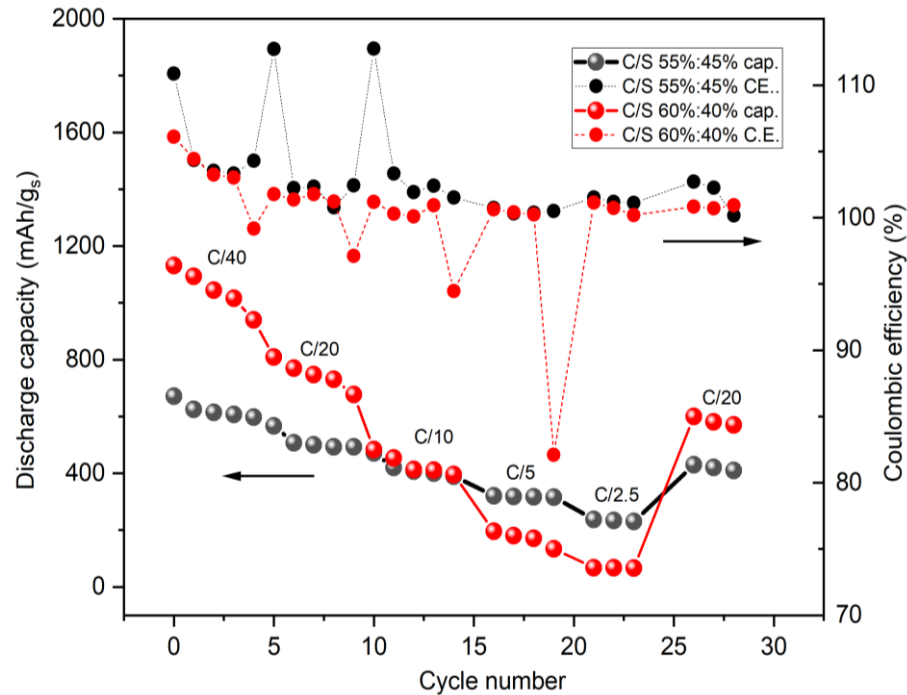
CV of as-prepared carbon-sulfur electrodes



GCPL of as-prepared carbon-sulfur electrodes

Electrochemical Results

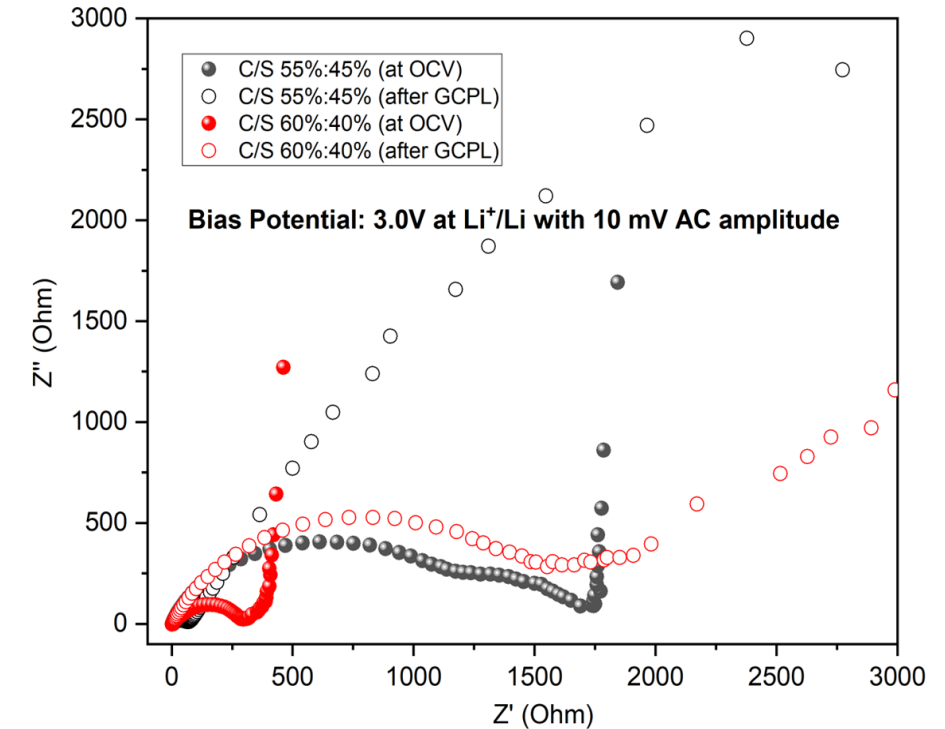
Key Findings



Rate capability of as-prepared carbon-sulfur electrodes

Drop in reaction kinetics at higher c-rates more significant for higher AC content

Higher charge transfer resistance and mass transport for higher AC content



EIS of as-prepared carbon-sulfur electrodes

Conclusion & Outlook

Conclusion



High carbon content (5% increase in C content & reduced interstitial packing density) without conductive carbon facilitates side reactions, significant CEI formation, and limits Li^+ diffusion during solid phase conversion in micropores of commercial activated carbon

Outlook

1

Development of composite carbon host to increase sulfur loading to range of 60 – 70%

2

Incorporate single atom electrocatalysts into host material to (i) increase the electronic conductivity and (ii) improve the polysulfide immobilization to the electrode surface

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Thank You for Your Attention!
