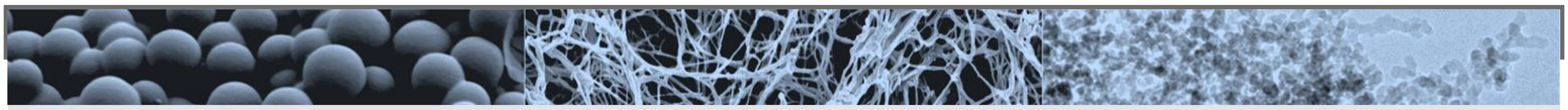
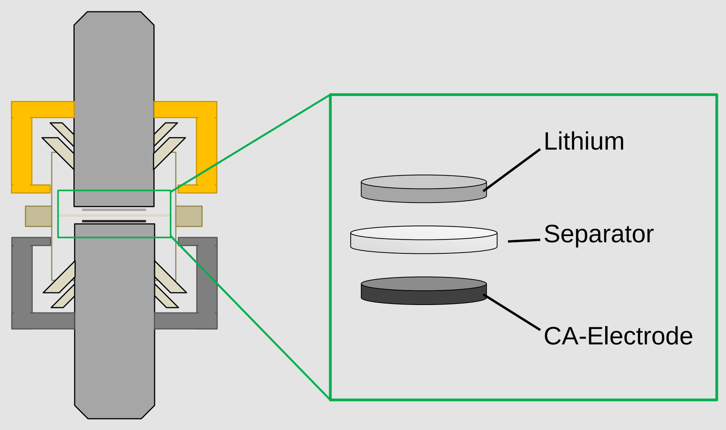
High sulfur content in microporous carbon aerogels for lithium-sulfur batteries

DLR

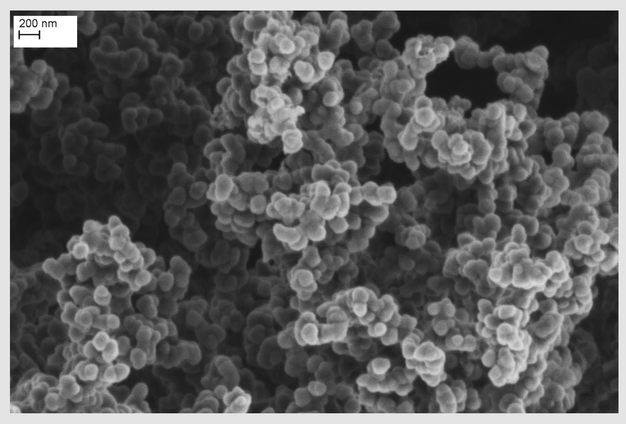


INTRODUCTION

Carbon aerogels (CAs) are highly promising materials as matrices for sulfur to act as cathodes in metal-sulfur batteries. Resulting from organic resorcinol-formaldehyde aerogels, CAs exhibit highly porous structures with porosities up to 97%, high surface areas of about 500-3500 m²/g, large pore volumes of about 0.1-2.5 cm³/g, and significant electrical conductivities of about 80-150 S/m.
Additional important advantages of CAs are their tunable porous structures and pore size distributions. Their microstructures are adjustable during the synthesis of organic aerogels and their subsequent carbonization. Moreover, the elastic compressibility of CAs prevents the formation of cracks as a result of changes to the sulfur volume.

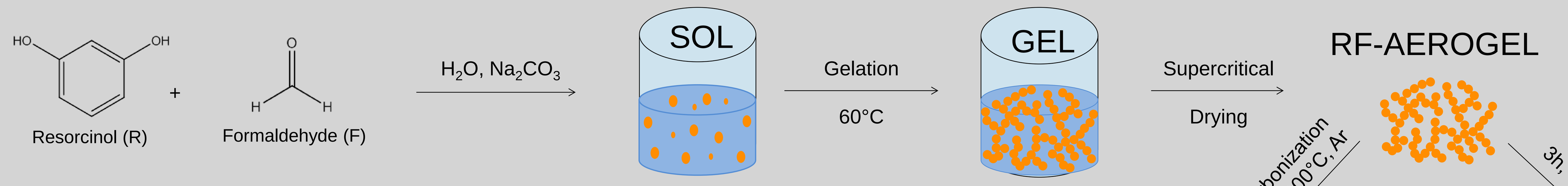


Testing battery cell



Microstructure of carbon aerogel

SYNTHESIS



Carbonization conditions:

Ar: 1 h, 1000°C, Argon atmosphere

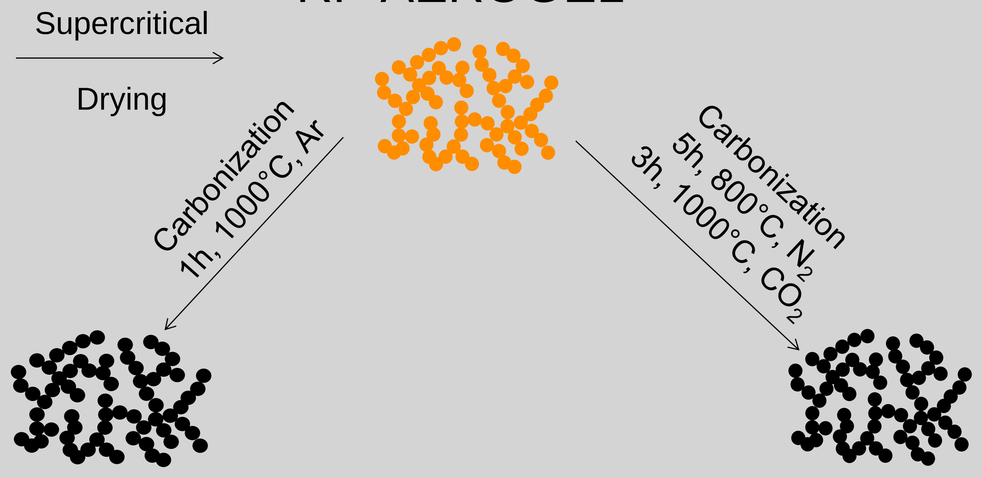
N_2/CO_2: 5 h, 800°C, N_2 atmosphere
3h, 1000°C, CO_2 atmosphere

Molar ratios:

Resorcinol/Formaldehyde = 0.5
Resorcinol/Water = 0.008
Resorcinol/Catalyst = 50

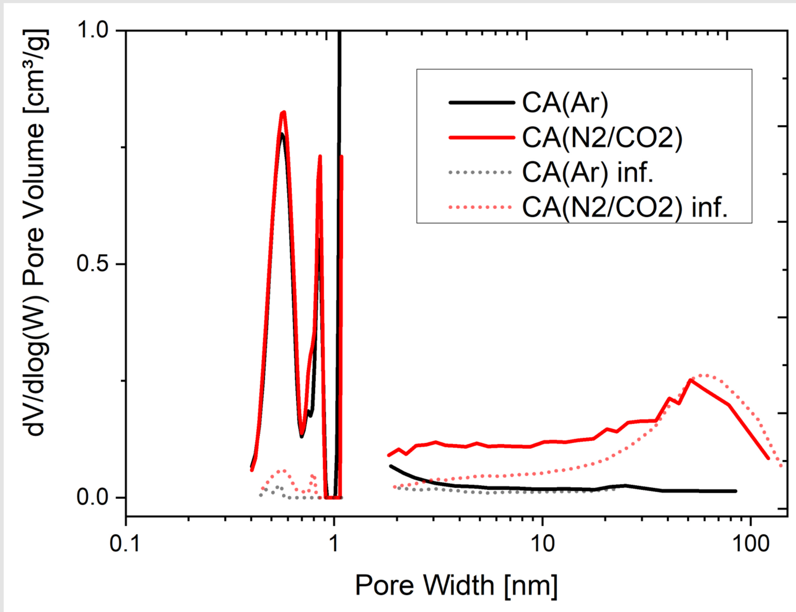
Reaction conditions:

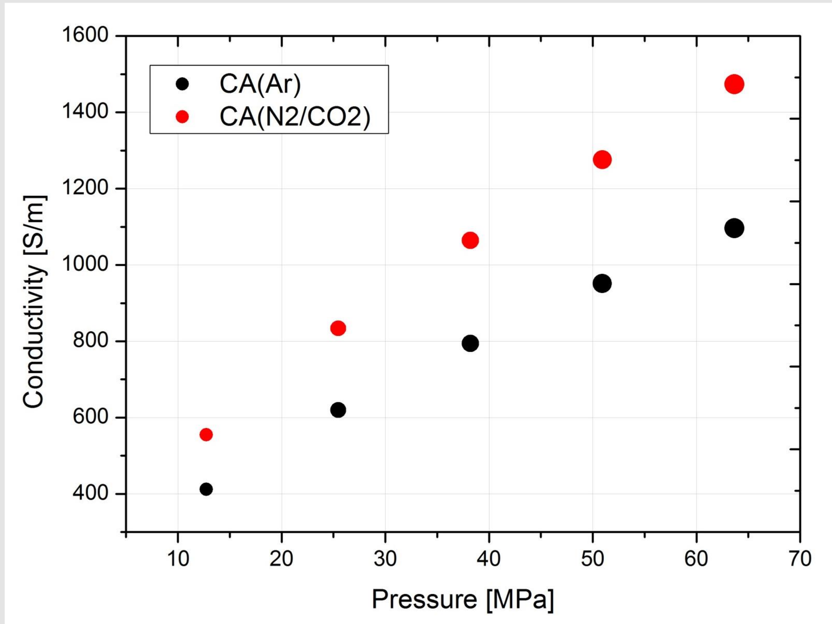
stirring time 60 min
at room temperature

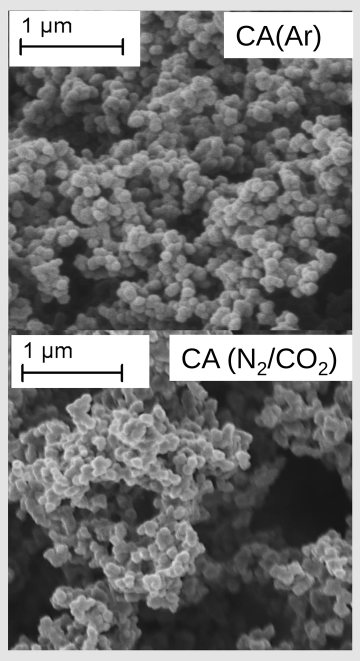


MICROSTRUCTURE

Properties	CA(Ar)	CA(N_2/CO_2)
Skel. density [g/cm³]	2.72	2.71
Bulk density [g/cm³]	0.04	0.07
Porosity [%]	98.6	97.3
Specific surface area [m²/g]	561	1912
Micropore volume [cm³/g]	0.18	0.60
Mesopore volume [cm³/g]	0.04	0.28







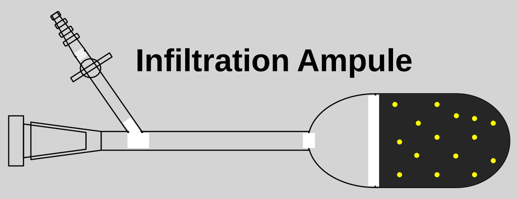
INFILTRATION

Two-step infiltration process:

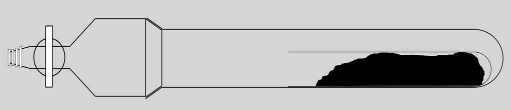
1. Gas Phase Infiltration
   - 600°C (6h) under vacuum in a sealed glass ampule
   - evaporated sulfur enters the micropores (< 2nm)

2. Evaporation of excessive sulfur from aerogel surface
   - 330°C (1.5h) under argon in glass tube with condensation trap
   - increases electrical conductivity of carbon
   - prevents undesired reactions with electrolyte

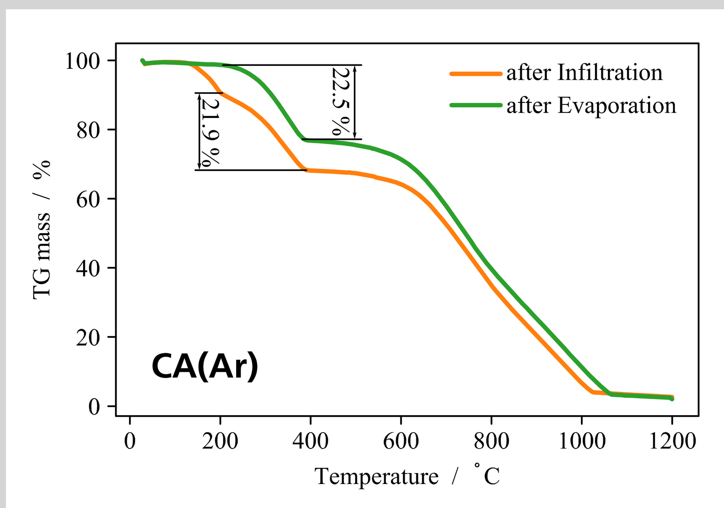
Infiltration Ampule

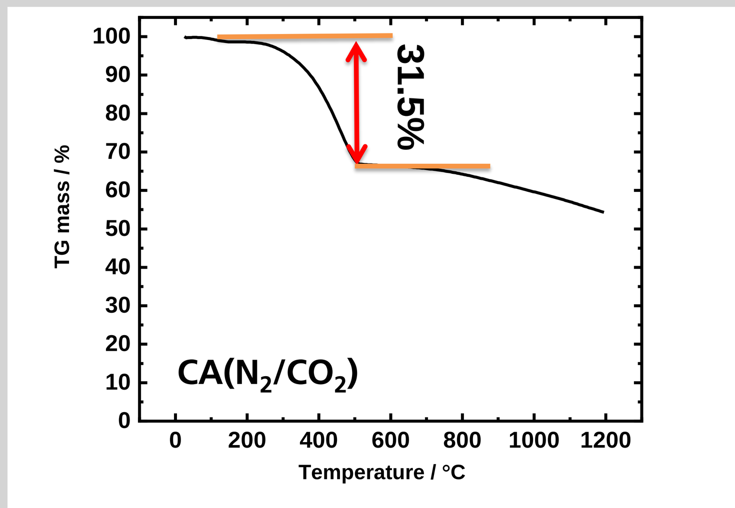


Evaporation Glass Tube



Thermogravimetric analysis





Higher microporosity leads to higher sulfur loading

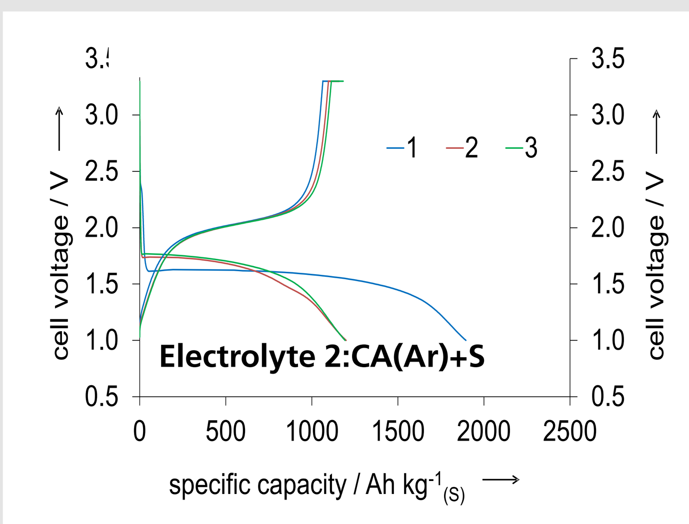
PERFORMANCE

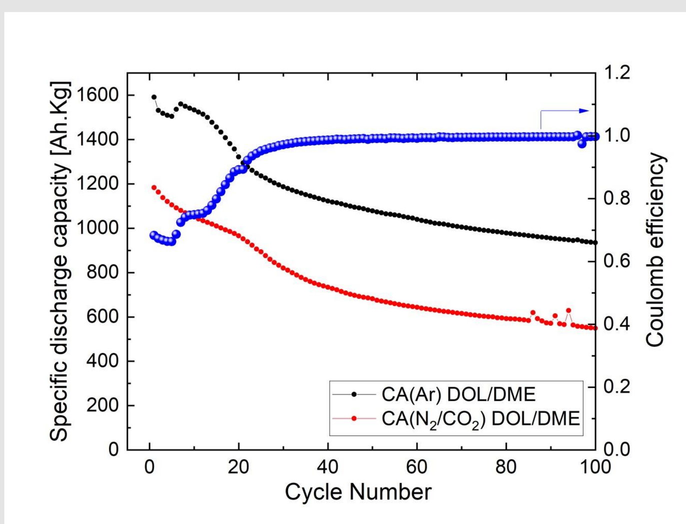
Cell Parameter:

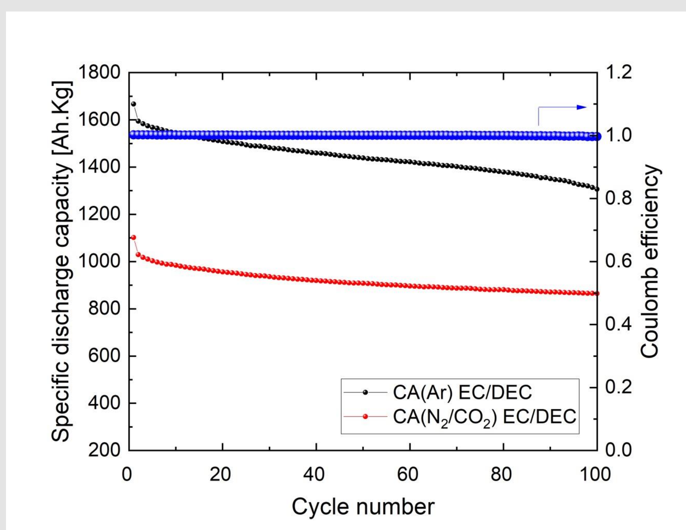
Cell type: EL-Cell
Electrolyte 1: 1M LiTFSI (lithium bis(trifluoromethylsulfonyl)imide in DOL/DME (1.3-dioxolane / dimethoxy ethane), 25 ℃
Electrolyte 2: 1M $LiPF_6$ (Lithiumhexafluorophosphat) in EC/DEC (ethylene carbonate / diethylene carbonate), 25 ℃
Sulfur-loading: 0.7–0.8 $mg_{(S)}/cm^2$

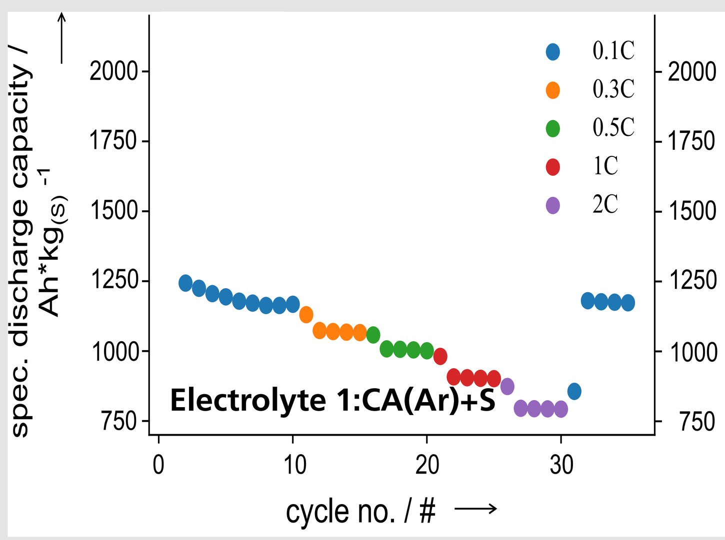
Results:

- Suppression of polysulfide shuttle effect using CA:S electrode
- Stable discharge capacity in both carbonate based electrolytes for CA:S
- High specific capacity and coulombic efficiency for CA:S cathode materials
- No irreversible losses in rate-capability test for CA:S cathode materials
- CA(N_2/CO_2):S exhibits lower discharge capacity due to presence of sulfur in mesopores









CONCLUSION

- Synthesis of the CAs with controlled and defined pore sizes and structures
- Increased surface area due to carbonization with N_2/CO_2
- Increased micropore volume in aerogel structure due to carbonization with N_2/CO_2
- Infiltration of short chain sulfur in the micropores through gas phase process
- Higher sulfur loading in CA (N_2/CO_2) due to higher micropore volume
- High cyclability of the Li-S cell via the encapsulation of sulfur in carbonate- and ether-based electrolytes

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