

Tunable Porosity and Structural Properties of Carbon Aerogels: Adapting Packing Density and Morphology of Dryfilm Cathodes for All-Solid-State Lithium-Sulfur Batteries

Hasti Rahimi^{a,b}, Magdalena Fiedler^{a,b}, Jessica Kröner^c, Marina Schwan^c, Barbara Milow^{c,d}, Felix Hippauf^b, Susanne Dörfler^{*b}, Stefan Kaskel^{a,b}

^aTU Dresden, Chair for Inorganic Chemistry I, Dresden, Germany

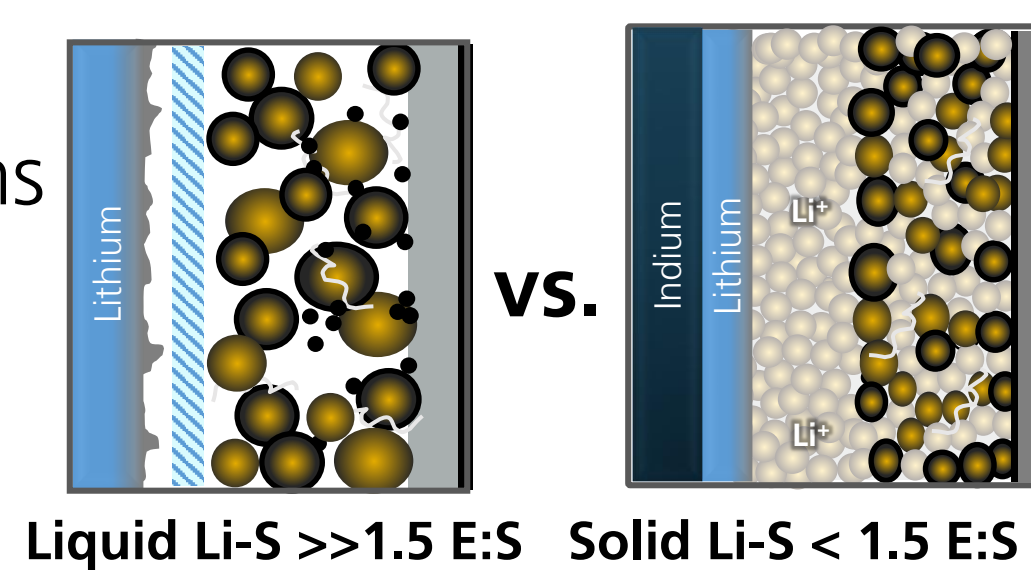
^bFraunhofer Institute for Material and Beam Technology IWS, Dresden, Germany

^cDeutsches Zentrum für Luft- und Raumfahrt e.V., Köln, Germany

^dUniversität zu Köln, Köln, Germany

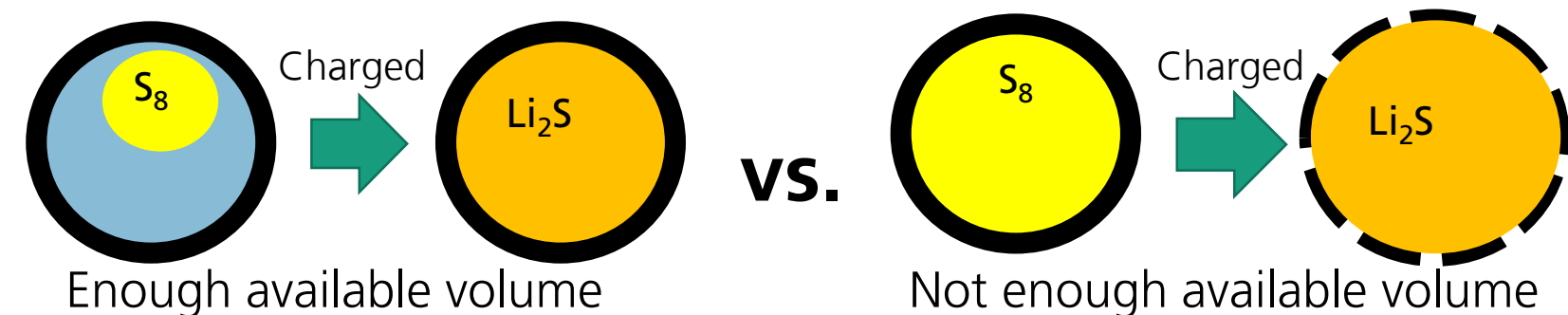
Motivation

- High gravimetric energy density possible > 500 Wh kg⁻¹
- Li⁺-ion selectivity → Prevention of polysulfide shuttle
- Adaptable micro- and meso-porosity and mechanical structure for carbon aerogels.
- Mesoporous carbon (C-Novel) outperforms microporous carbon but is cost intensive.



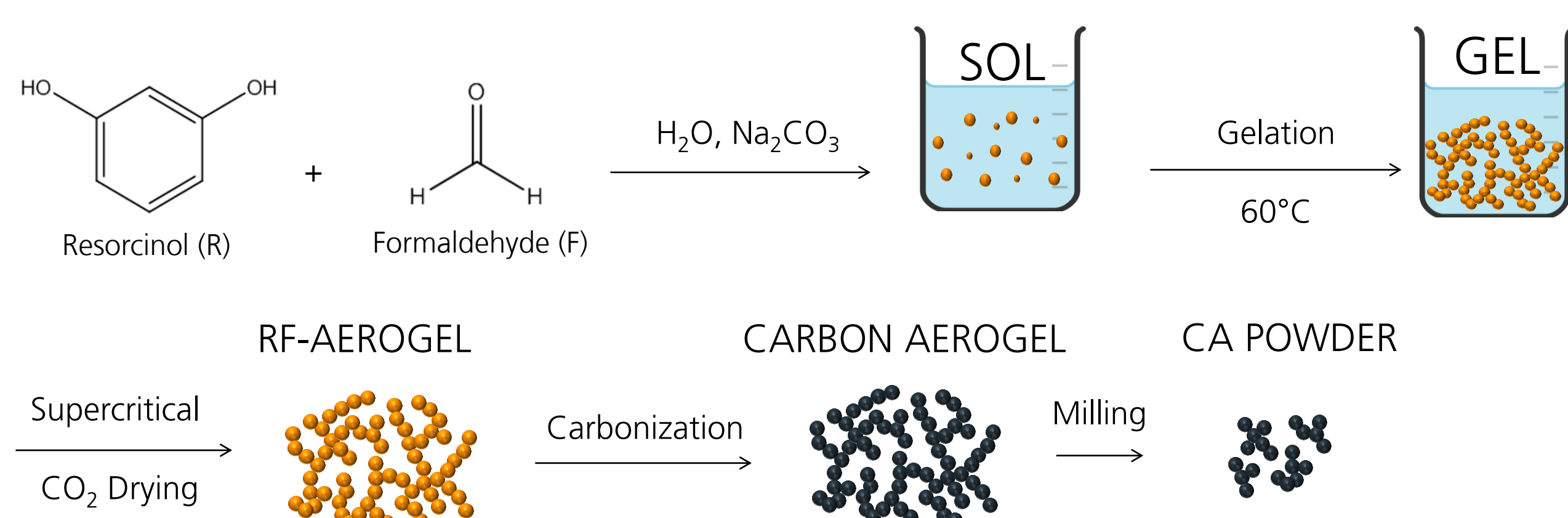
Challenges

- Formation of triple phase formation without high energy ball milling
- Volume expansion of Sulfur to Li₂S → Full active material utilization and mechanical stress



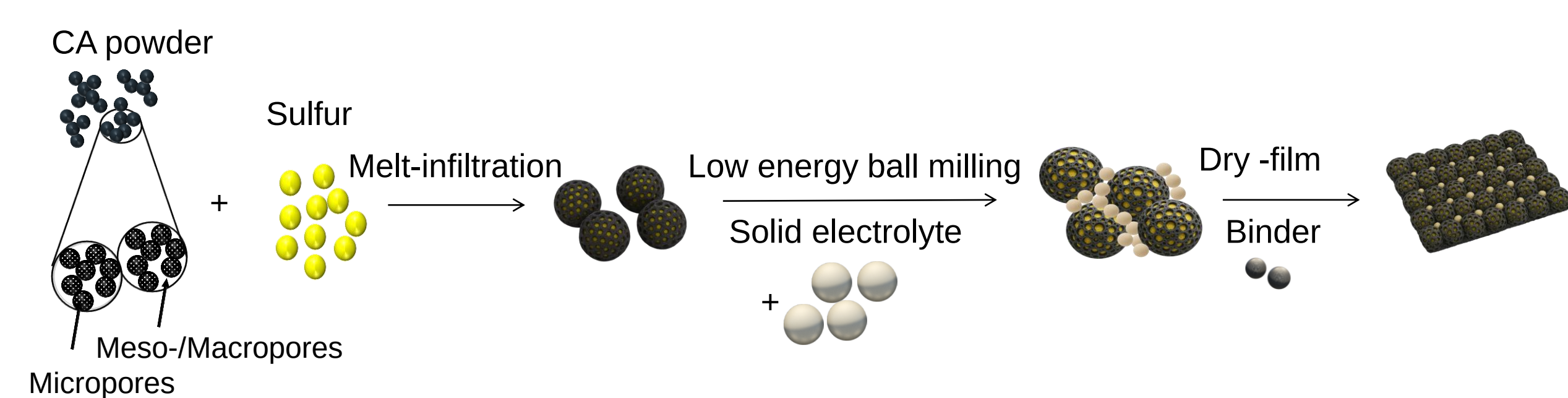
Method

- Synthesis of the carbon aerogels (CA)



- Melt infiltration of carbon and sulfur in 3 different ratios

1) $V_{\text{Li}_2\text{S}} = V_{\text{micro-} + \text{mesopore}}$ 2) 60% 3) $V_{\text{S}} = V_{\text{micro-} + \text{mesopore}}$



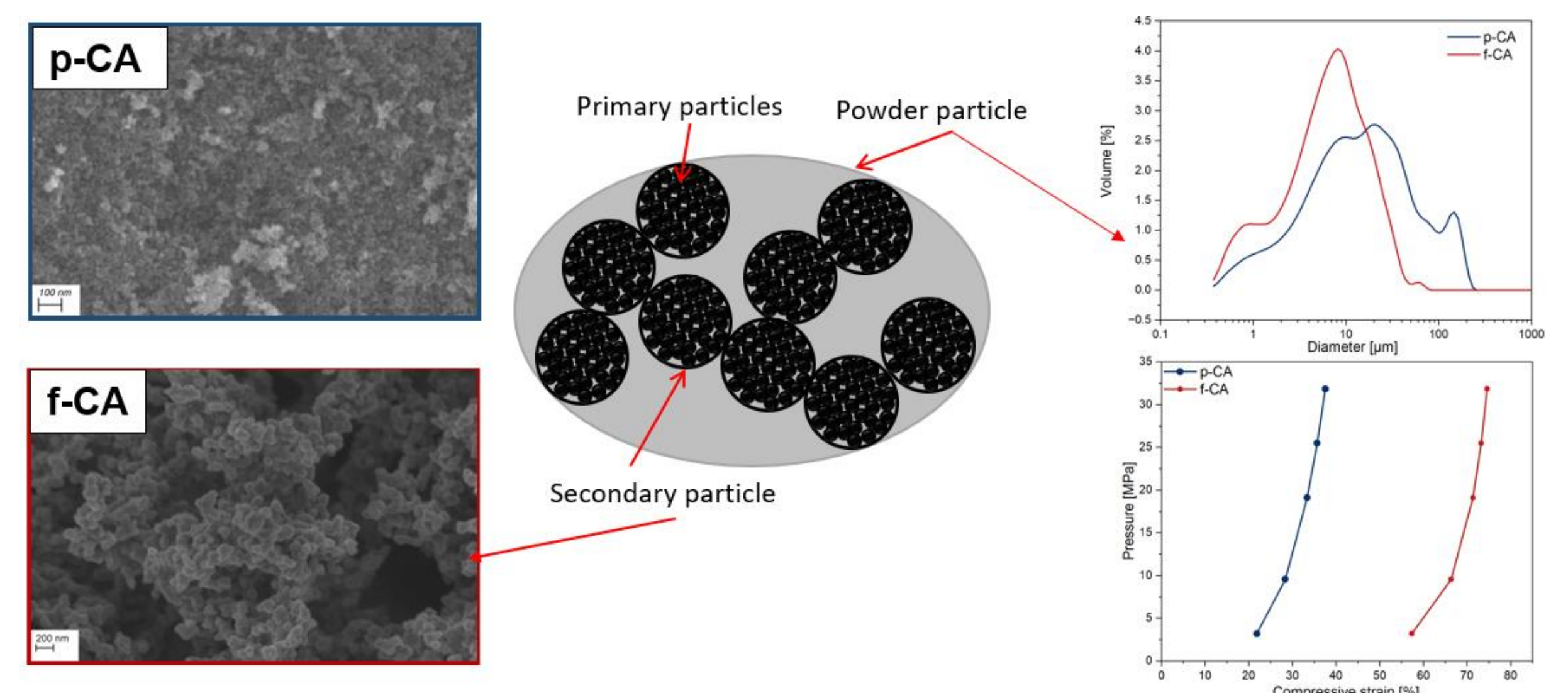
- Cell assembly in a pressurized lab cell with cathode of 1.5 mg_S cm⁻²; 2.5 mAh cm⁻² loading vs. In-Li alloy
- Evaluation of micro- and mesoporous carbon and their combination

Conclusion

- The brittle mesoporous carbon (p-CA) showed higher active material (AM) utilization but low CE with a densified morphology.
- The flexible micro- and macroporous carbon (f-CA) demonstrated lower AM utilization but high CE with voids and cracks in morphology
- Addition of f-CA to p-CA increases packing density → Better ion and electron transportation → Better C-rate performance and capacity retention → beside porosity, the carbon flexibility is a crucial parameter for high S-utilization

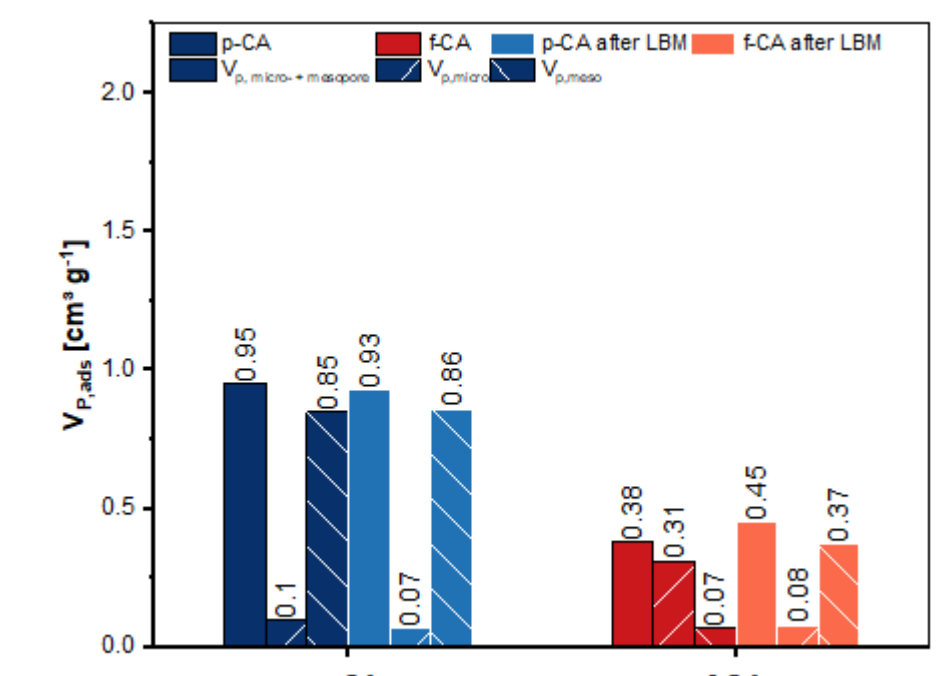
Results

Carbon characterization- before and after low energy ball milling



- Different microstructure and properties for p-CA (brittle) and f-CA (flexible) affects compression behaviour

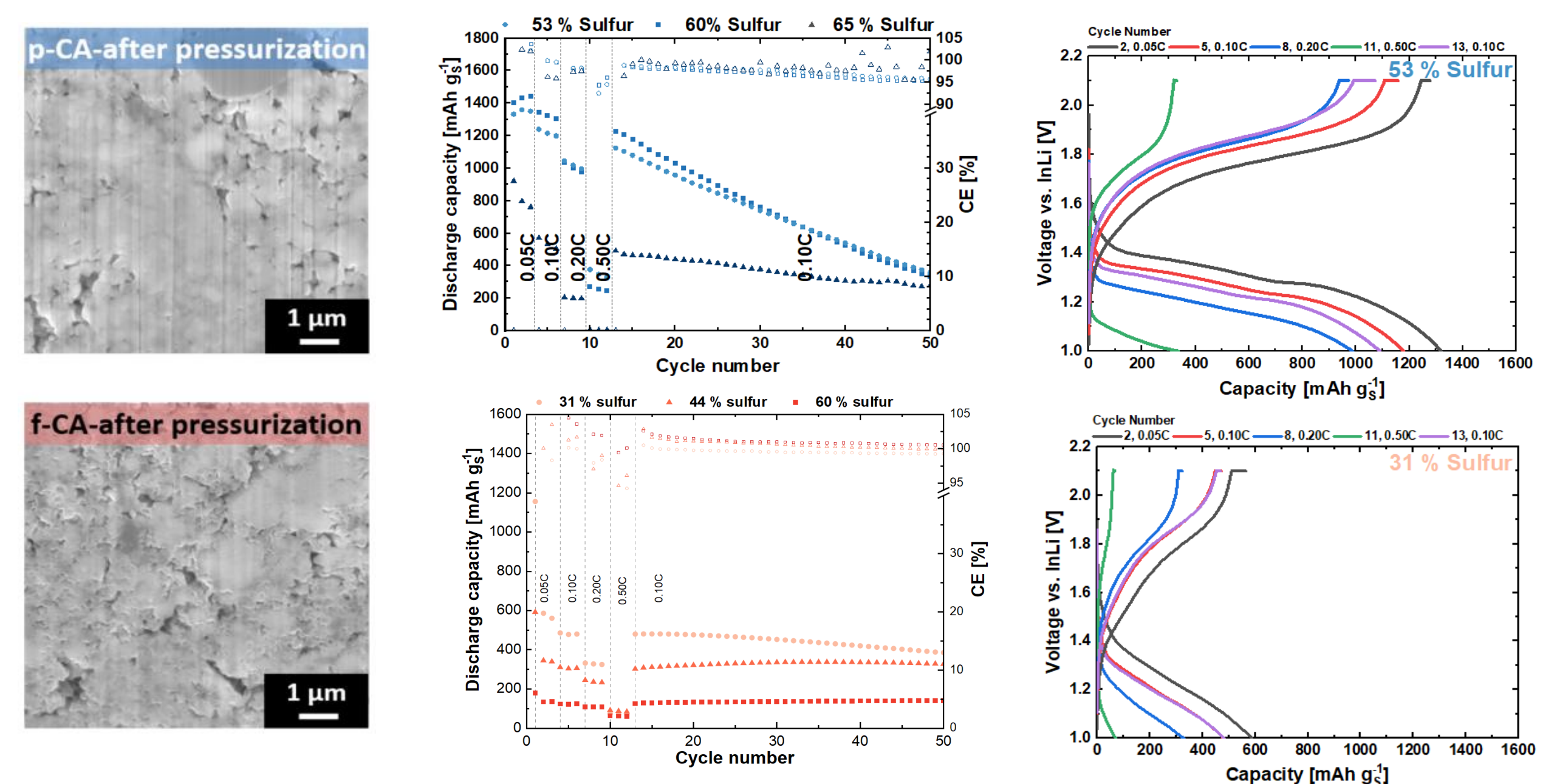
Properties	p-CA	f-CA
Porosity [%]	73	97
Specific surface area [m ² g ⁻¹]	693	836
Average primary particle size [nm]	4.2	2.7
Compressive strain at 1 kN [%]	22	57



- Mechanical properties were effected in different manners with LBM

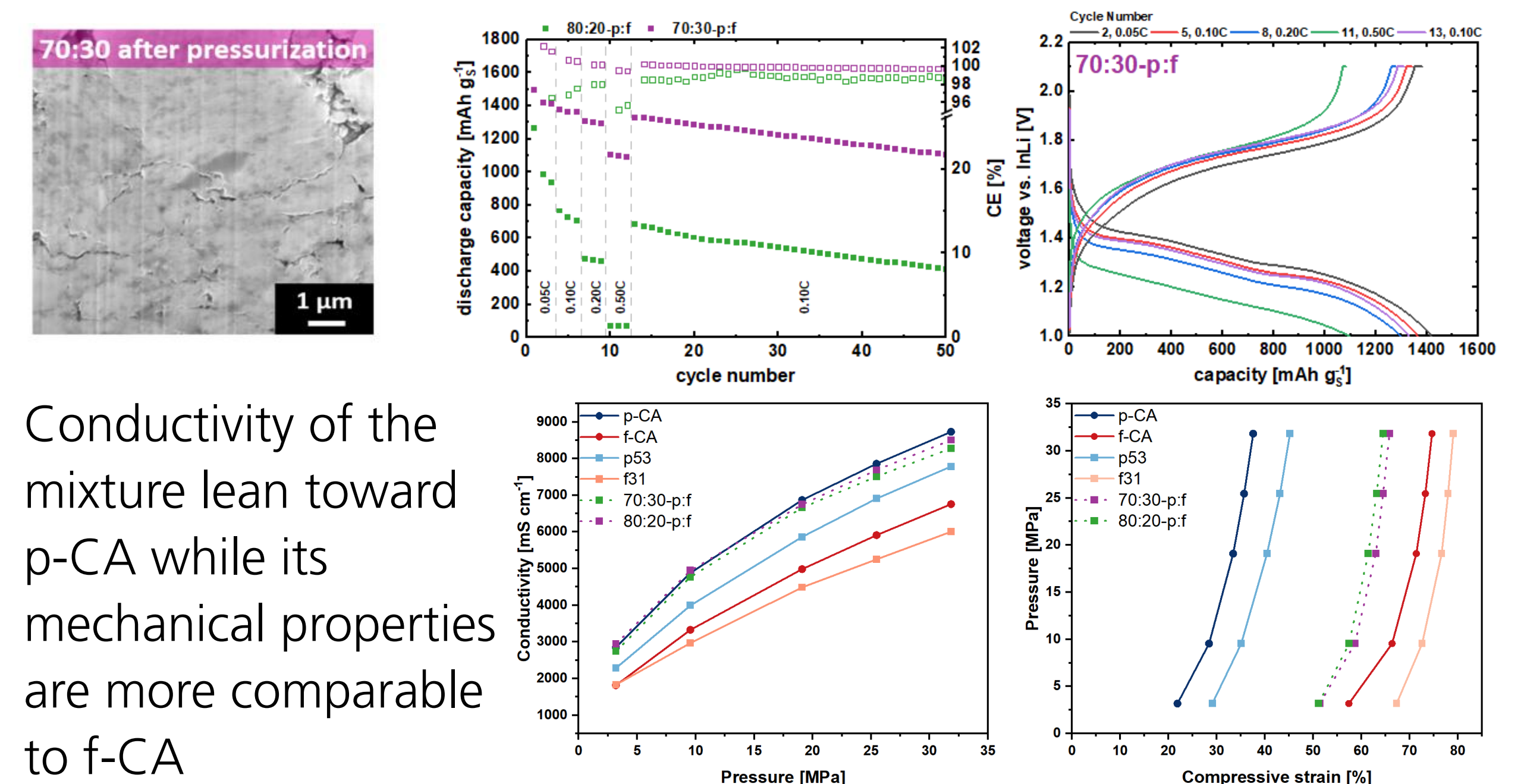
Electrochemical characterization

- p-CA shows higher sulfur utilization but lower CE; whilst f-CA has better capacity retention and CE but low sulfur utilization.
- Insufficient free volume in p-CA and less densified structure for f-CA



Mechanical and electrochemical characterization of with the CAs combination

- The sulfur utilization and capacity retention and morphology enhanced significantly by combining micro- and mesoporous carbons



- Conductivity of the mixture lean toward p-CA while its mechanical properties are more comparable to f-CA