

Polymer (Hybrid) Electrolytes for High Performance Lithium-based Cell Chemistries

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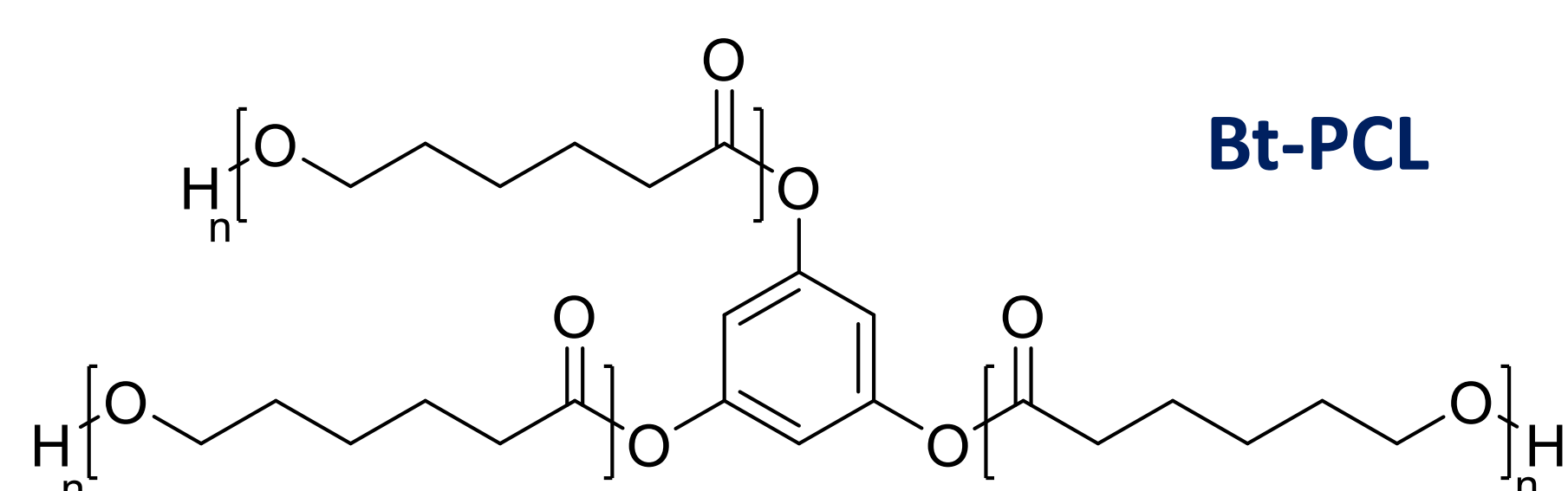
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Advantages of Si/C_x anodes

- Theoretical specific capacity up to 4200 mAh/g
- Faster charging
- Higher energy densities
- Long cycle life
- Lower carbon footprint compared to graphite
- Lower estimated production costs compared to graphite anodes

Synthesis of Bt-PCL polymer



- Star-based benzene trihydroxy-polycaprolactone
- Straightforward one pot reaction
- **Batch production** in a 2 L reactor (500 g product)
- Grafting of polymer: compatibility with higher voltage cathodes



Y.-H. Chen et al., *Advanced Functional Materials*, **2023**, 33, 2300501.

Matrix-supported polymer electrolyte

→ Boosting mechanical properties for Si/C_x anodes



Fabrication:

- Dry mixing & pre-melting @ 100 °C overnight
- Hot-pressing of polymer membranes (100 °C, 20 bar, 2 min)
- Let polymer membranes rest for at least 24 h
- Hot-pressing of 5 µm PP separator between polymer membranes
- UV curing for 15 min → ~180 µm thick hybrid membrane

Conclusion

- Proof-of-concept galvanostatic cycling with Bt-PCL SPE in NMC₆₂₂ || Si/C and NMC₉₀₆₄ || Si/C cells
- High silicon content of 90% due to compensation of volume expansion effects by SPE
- Enabling higher mass loadings of electrodes with SPEs due to temperature assisted solvent-free vacuum melt infiltration
- Safer-to-operate polymer-based batteries

Acknowledgement

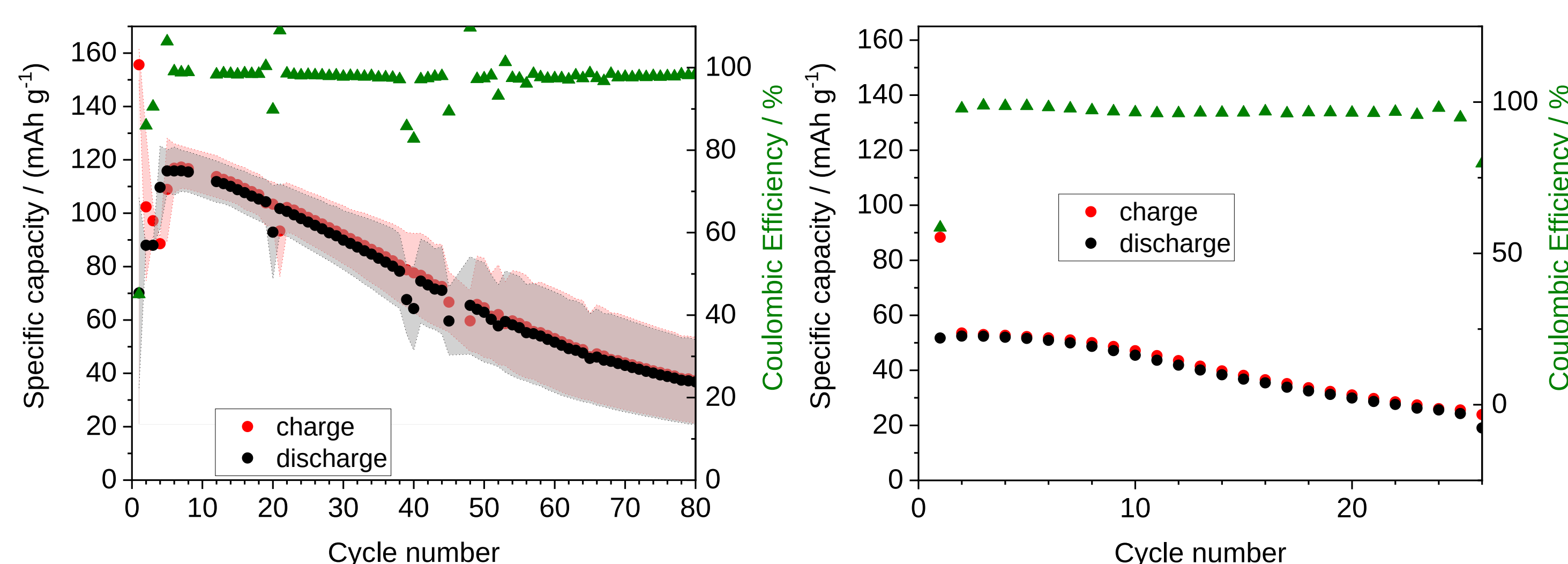
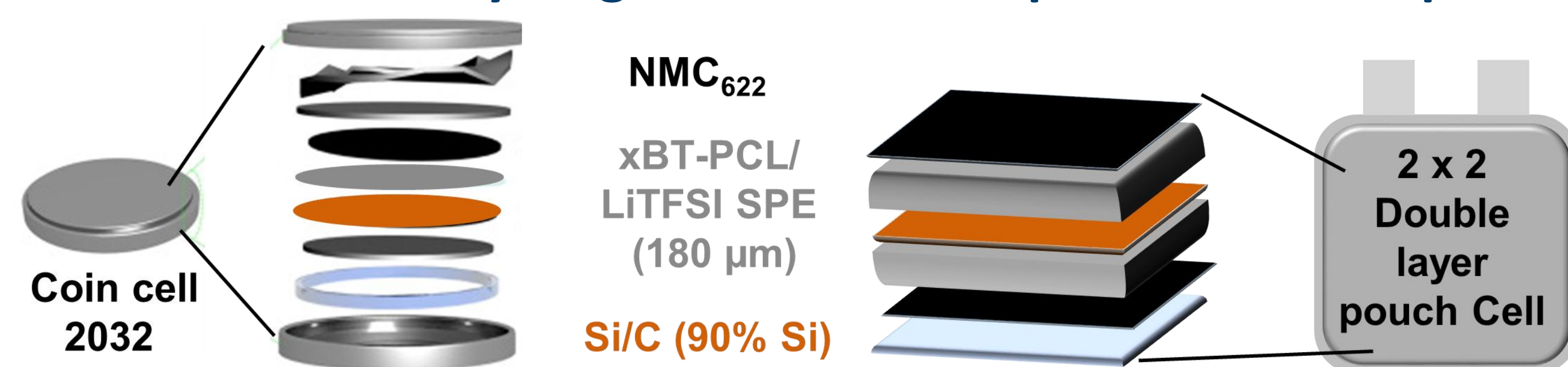


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Electrochemical Characterization

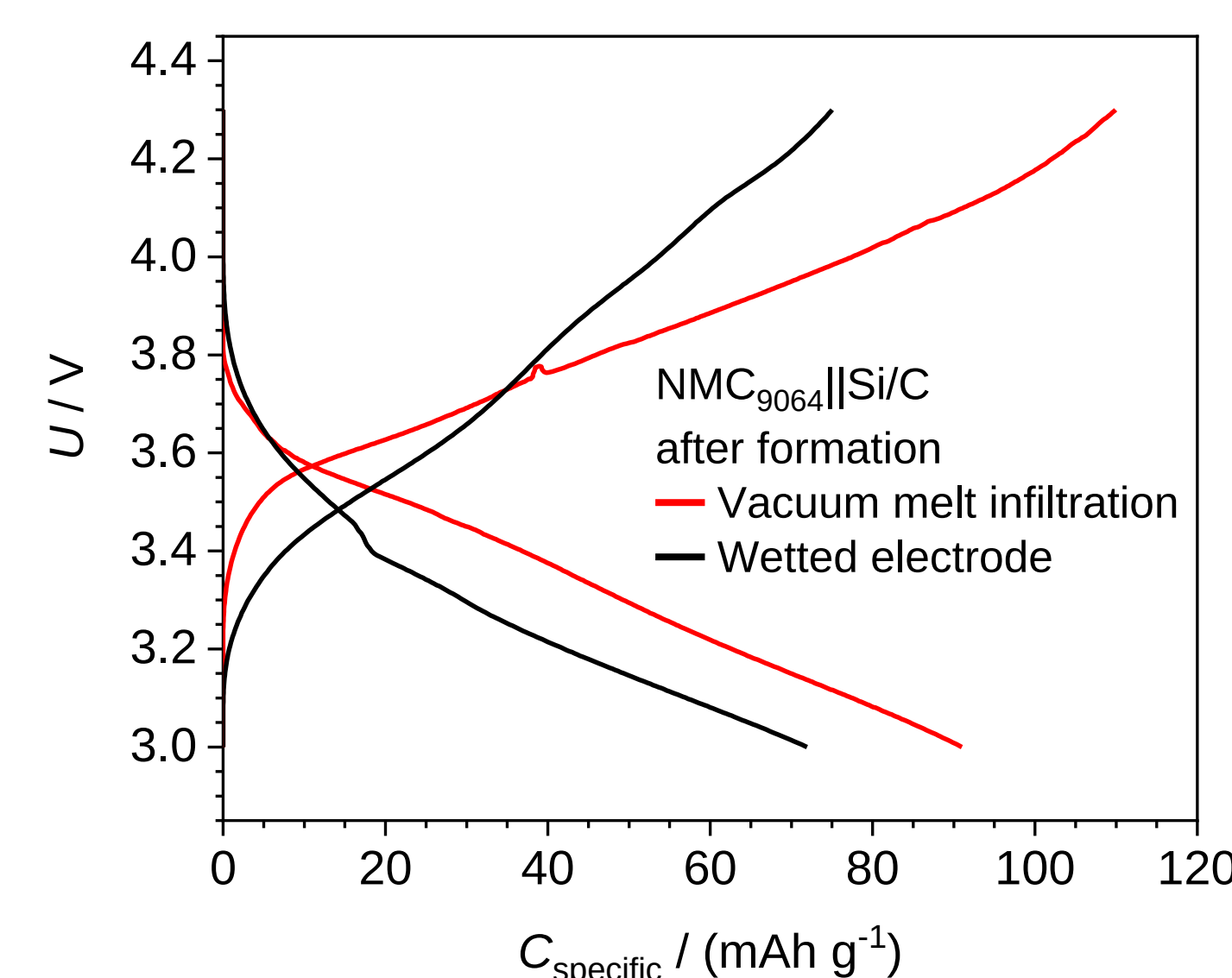
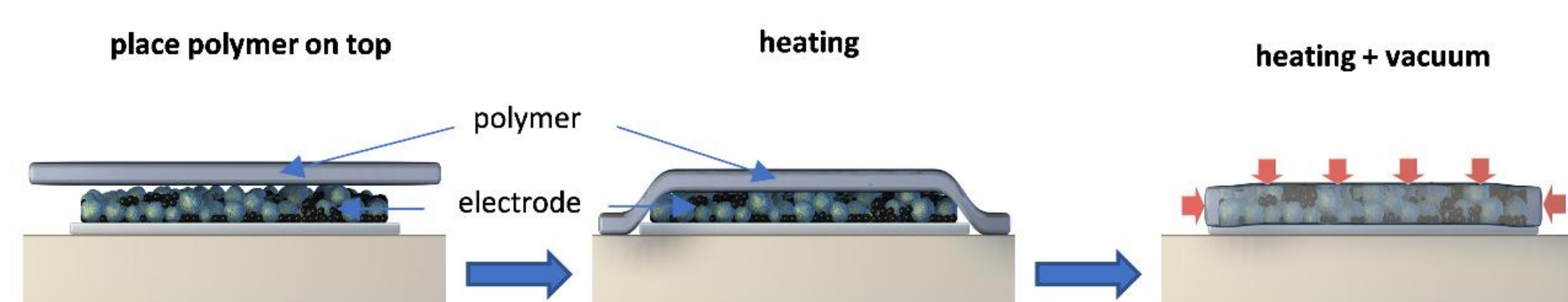
Galvanostatic cycling in coin cell and pouch cell set-up



- **Proof-of-concept** galvanostatic cycling with xBt-PCL/LiTFSI in coin cells
- Lower pressure in pouch cells* leads to lower specific capacity
- Cell capacity currently reached: 0.008 Ah; volumetric energy density: 59 Wh L⁻¹; gravimetric energy density 56 Wh L⁻¹
- Challenge: Contact difficulties at electrode-electrolyte interphases at higher mass loadings

Cell setup	NMC ₆₂₂ or NMC ₉₀₆₄ Si/C (90% Si)
Electrolyte	xBT-PCL/LiTFSI [O:Li] = 5:1 Hybrid in 5 µm PP separator + 12 µL (coin cell) / 20 µL (pouch cell) 1 m LiTFSI in PC
θ / °C	40
q_{Cathode} / (mAh cm ⁻²)	1 or 1.5
Cycling procedure	CC+CV (charge), CC (discharge) with C/10
E / V	3.0 – 4.3
$C_{\text{practical}}$ / (mAh g ⁻¹)	160 or 200

Temperature assisted solvent-free vacuum melt infiltration



- Melt infiltration under vacuum conditions of non-crosslinked Bt-PCL SPE
- Method **boosts** accessible electrode **mass loadings** and **electrochemical performance** by optimizing contacts

P. Roering, M. Winter, G. Brunklaus et al., **2024**, *ACS Appl. Mater. Interfaces*, 16, 17, 21932.
A. Buchheit, F. Scharf, S. Krämer et al., **2025**, *Energy Storage Materials*, 83, 104670.