

Synthesis of optimized cathode active materials for solid-state Na-batteries within the Franco-German project HIPOBAT

Christoph Roitzheim^a, Dina Fattakhova-Rohlfing^{a,b,c}, and Martin Finsterbusch^{a,b}

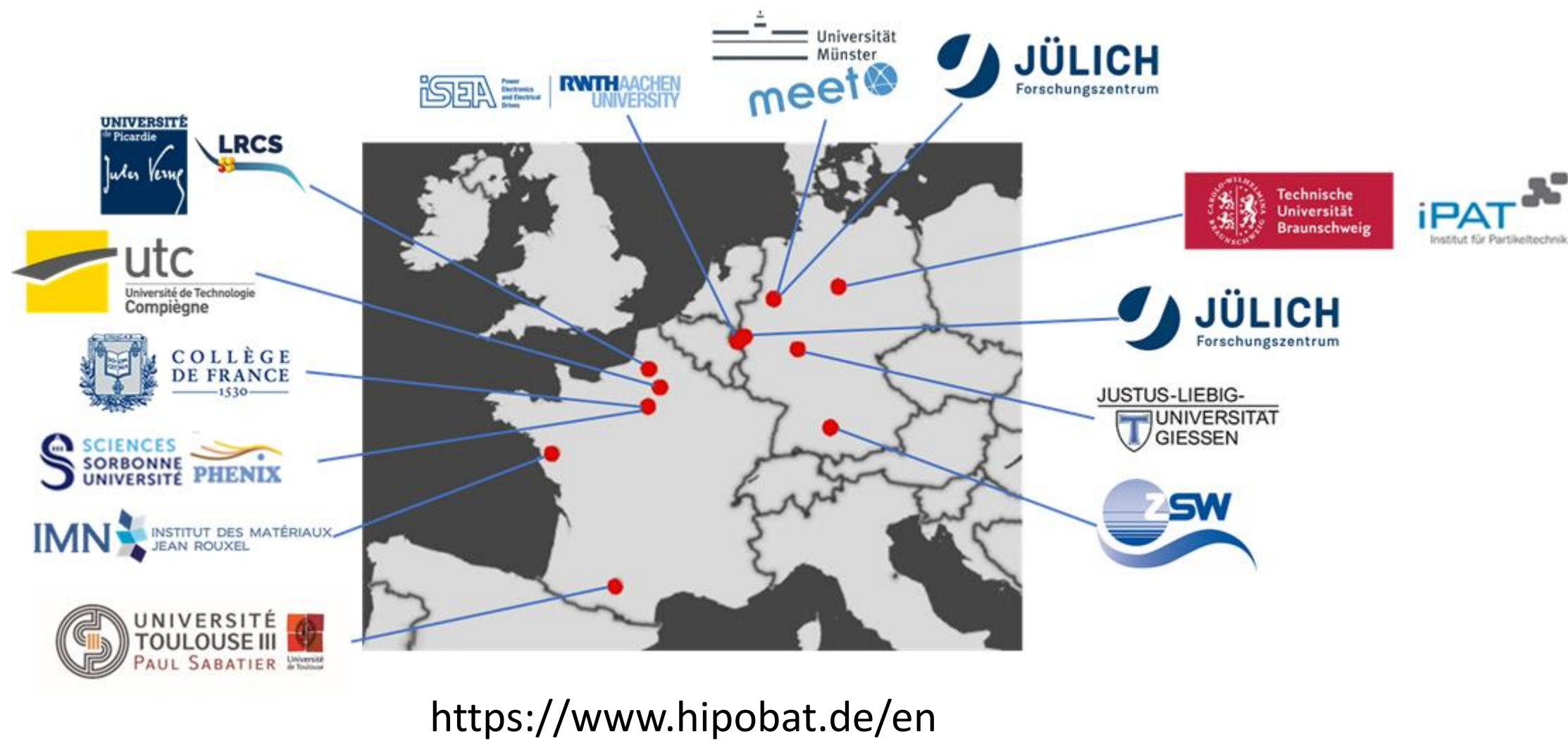
^aInstitute of Energy Materials and Devices – Materials Synthesis and Processing (IMD-2), Forschungszentrum Jülich GmbH, Wilhelm-Johnen-Str., 52425 Jülich, Germany

^bHelmholtz Institute Münster: Ionics in Energy Storage (IMD-4), Corrensstr. 46, 48149 Münster, Germany

^cFaculty of Engineering and Center for Nanointegration Duisburg-Essen (CENIDE), Universität Duisburg-Essen, Lotharstraße 1, 47057 Duisburg, Germany

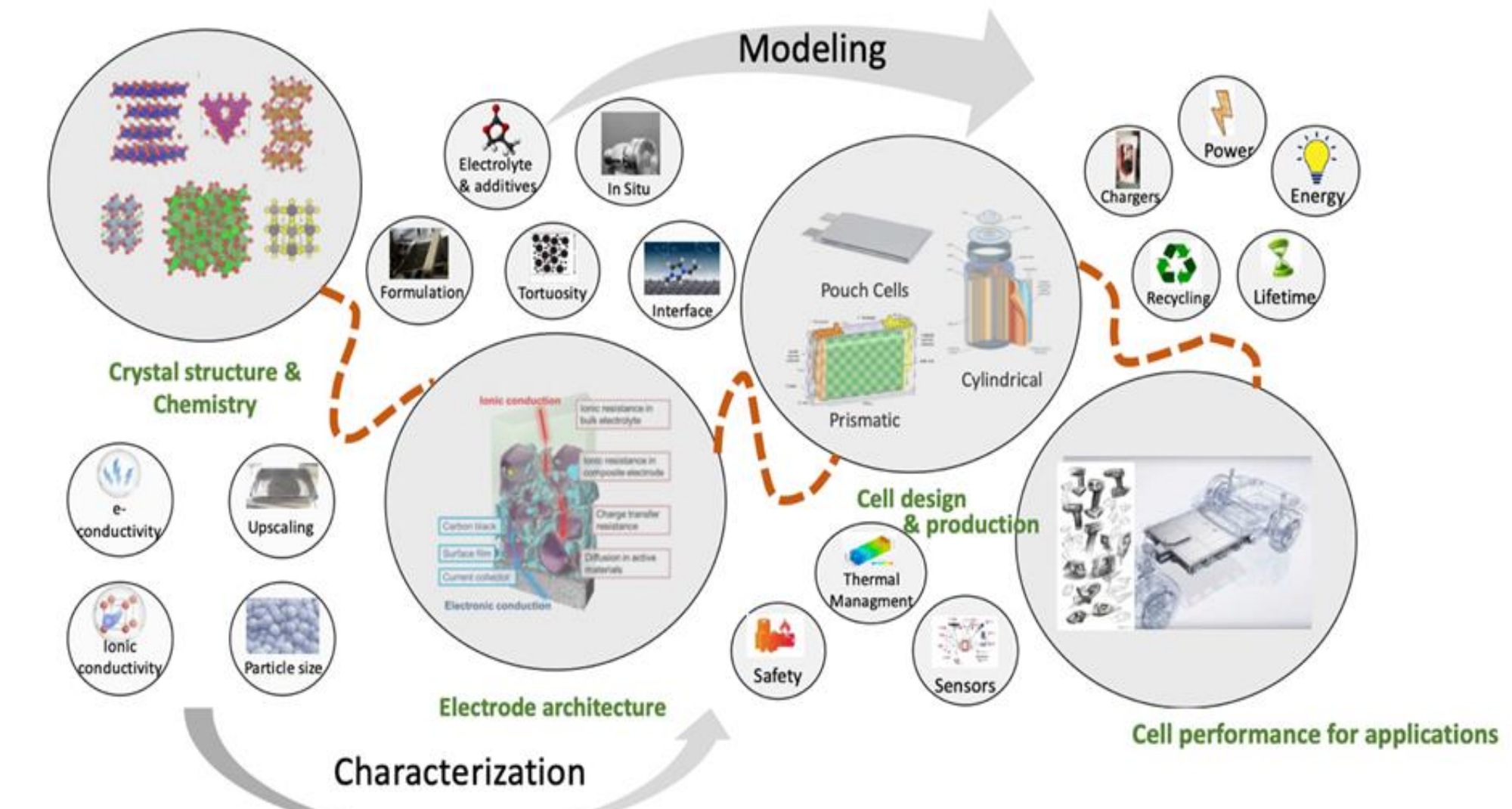
The HIPOBAT project

In the Franco-German project, six French and six German research groups have joined forces to develop high-power solid-state batteries (SSBs). The cells will be optimized towards fast charging, a long lifetime, and high energy density. The HIPOBAT project focuses on both lithium and sodium chemistries. Alongside cutting-edge scientific advances in the field of high-power batteries, the HIPOBAT project will establish an exchange program for early career researchers to promote the mobility between France and Germany to strengthen the Franco-German relationships in battery science.

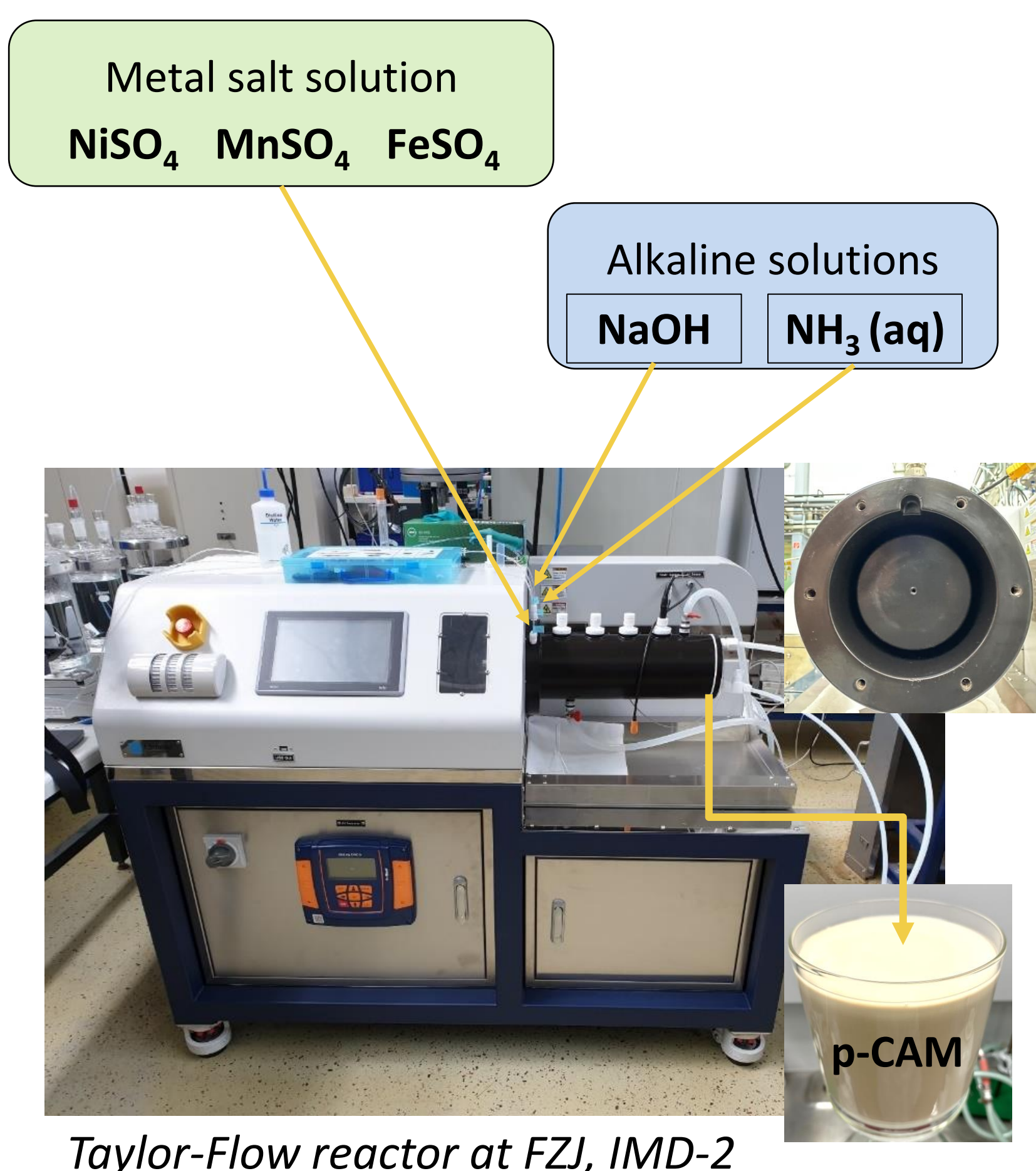


Research topics at Forschungszentrum Jülich (IMD-2)

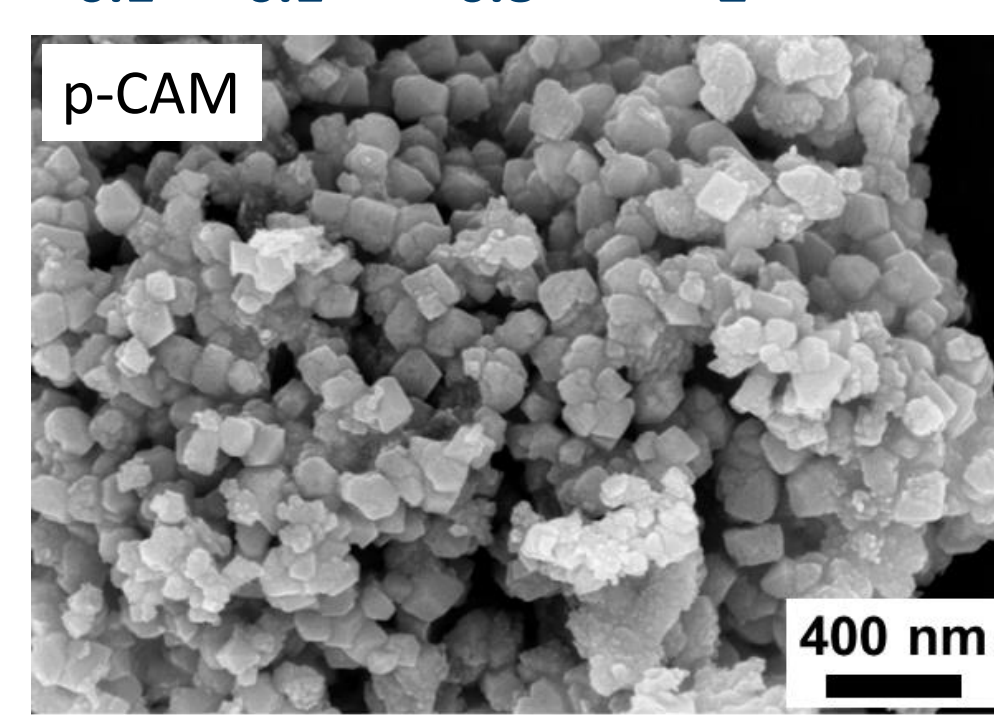
- Development of oxide ceramic and hybrid solid-state Na-batteries
- Synthesis and optimization of oxide solid-state electrolytes ($\text{Na}_{3.4}\text{Zr}_{2.0}\text{Si}_{2.4}\text{P}_{0.6}\text{O}_{12}$, NZSP)
- **Synthesis and optimization of layered cathode active materials (Na_xMnO_2)**
- Multiscale modeling of materials



Cathode active material synthesis

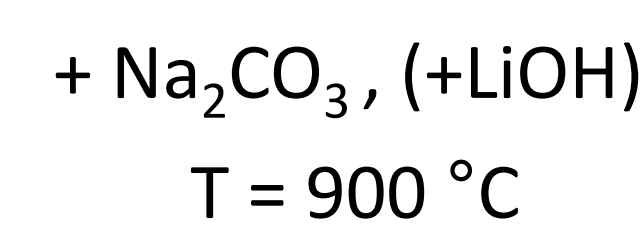


$\text{Ni}_{0.1}\text{Fe}_{0.1}\text{Mn}_{0.8}(\text{OH})_2$ (p-CAM)



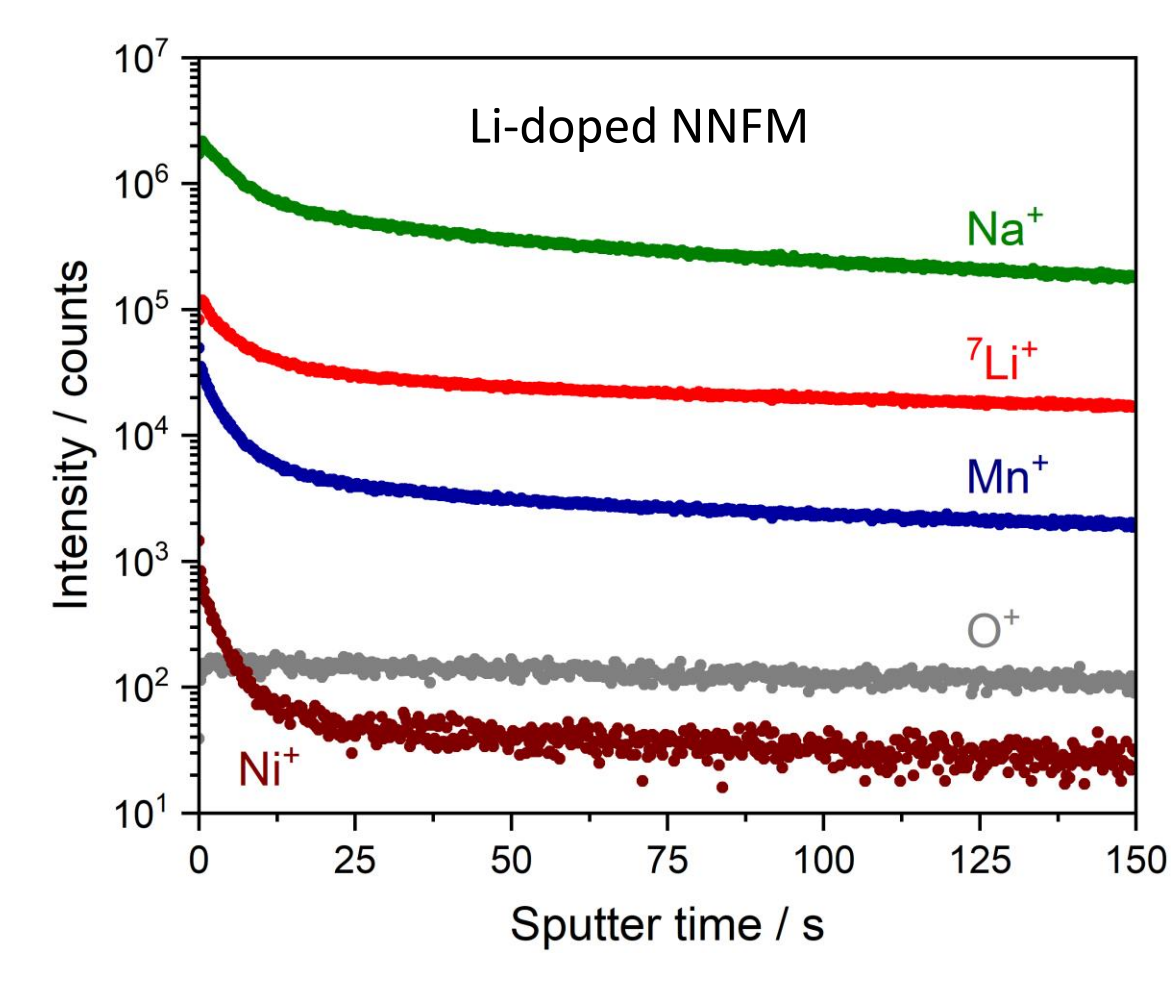
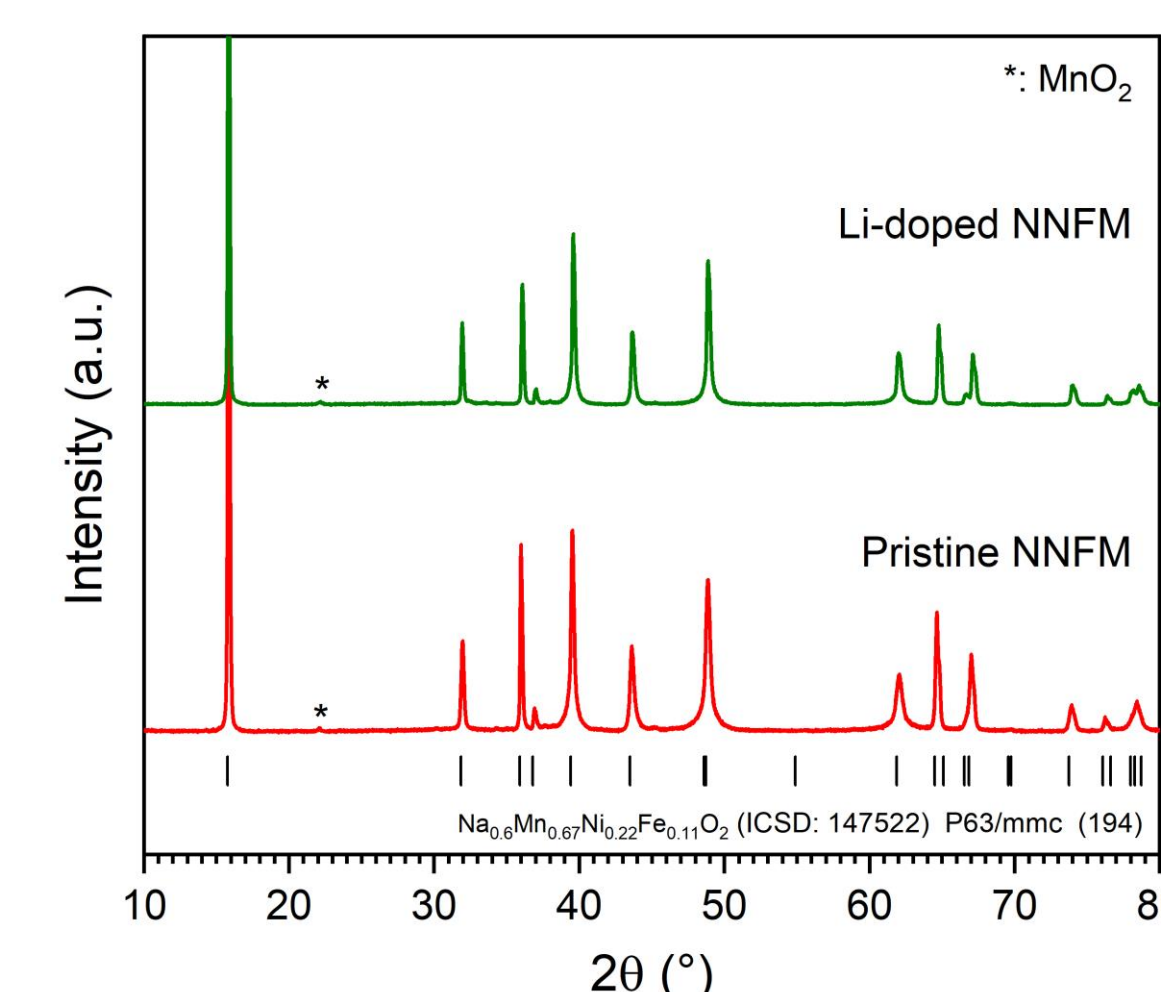
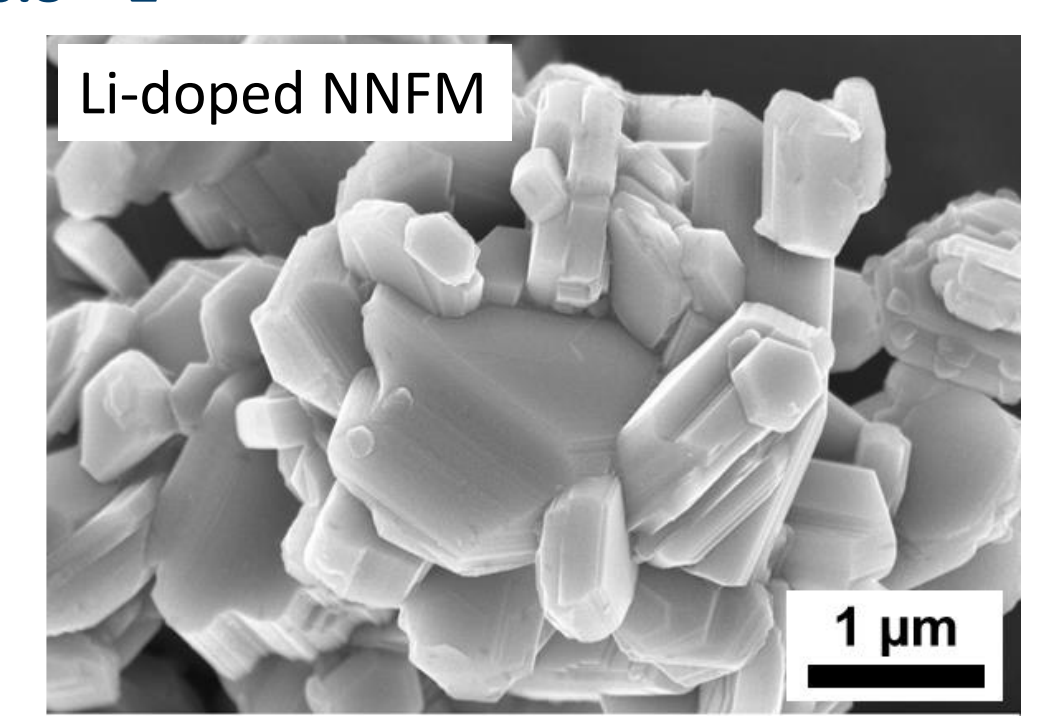
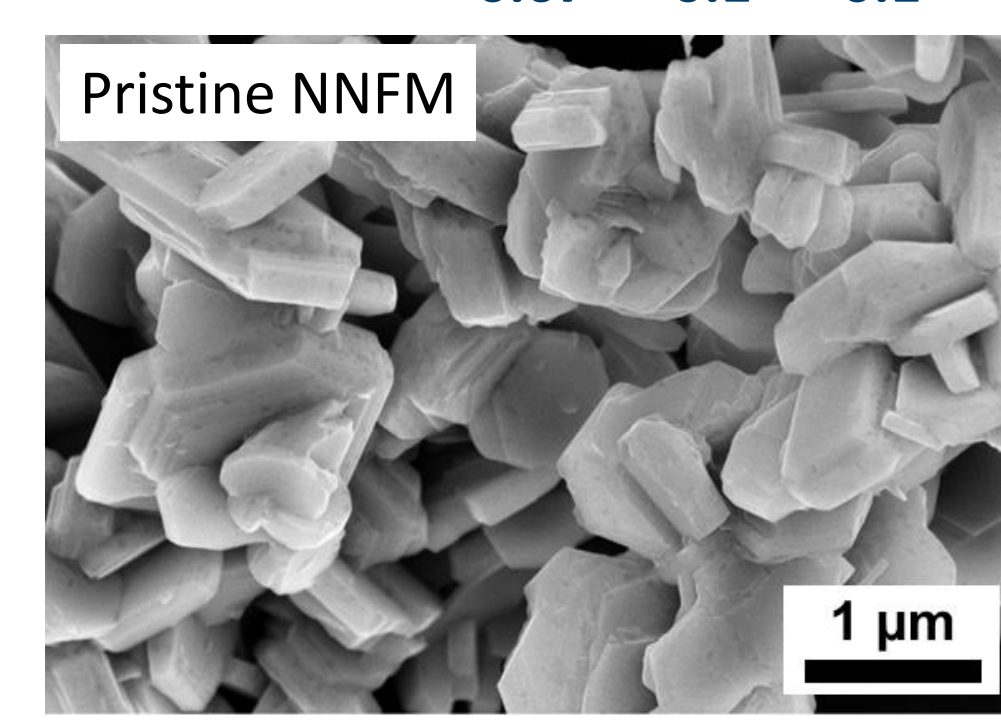
400 g dried p-CAM powder

Calcination, sodiation, (doping)



30 g calcined NNFM powder in an Al_2O_3 crucible

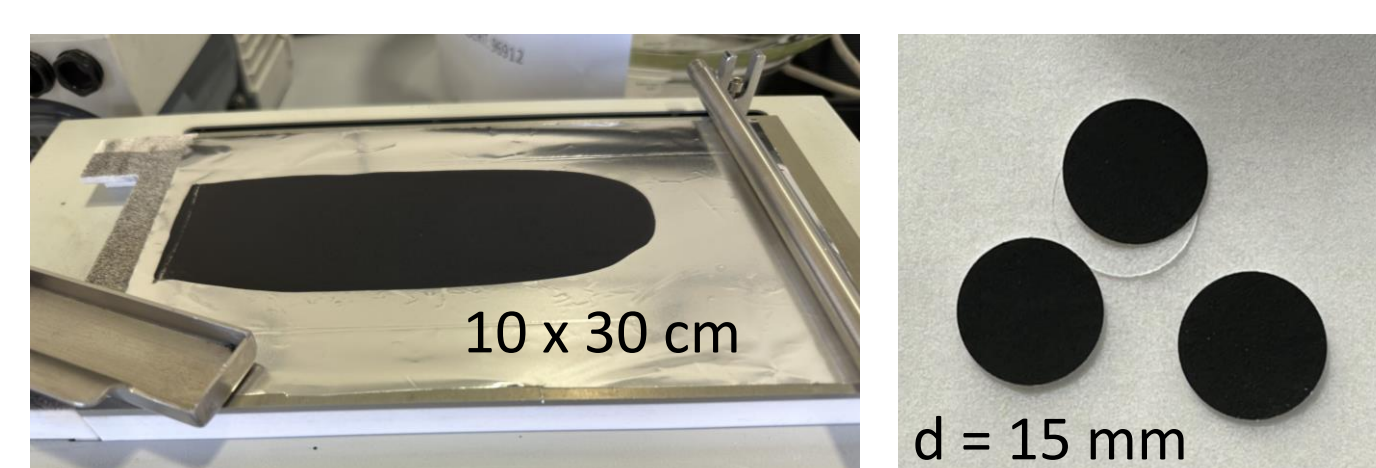
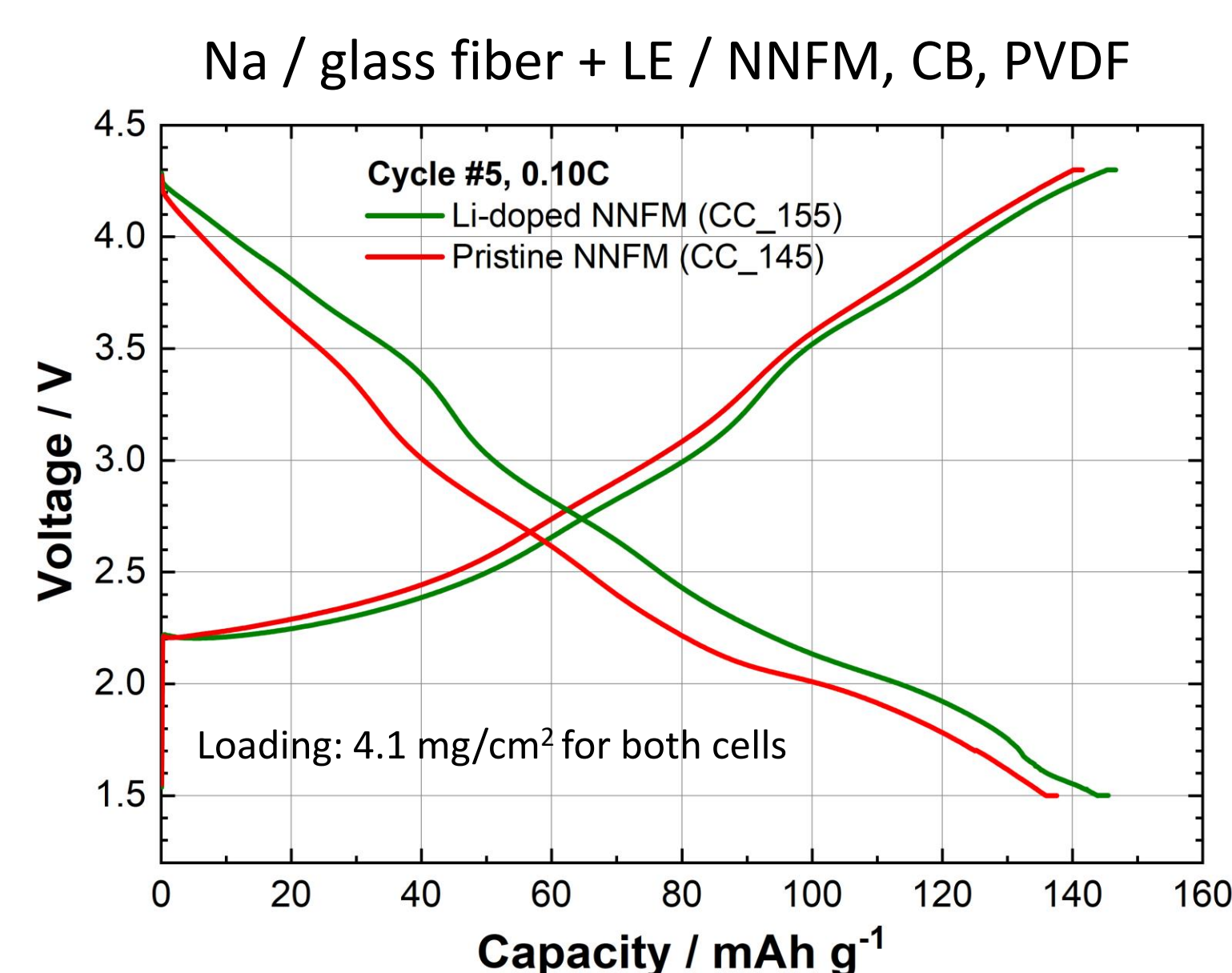
P2-type $\text{Na}_{0.67}\text{Ni}_{0.1}\text{Fe}_{0.1}\text{Mn}_{0.8}\text{O}_2$ (NNFM)



→ Pristine and Li-doped NNFM show high phase purity and similar morphology

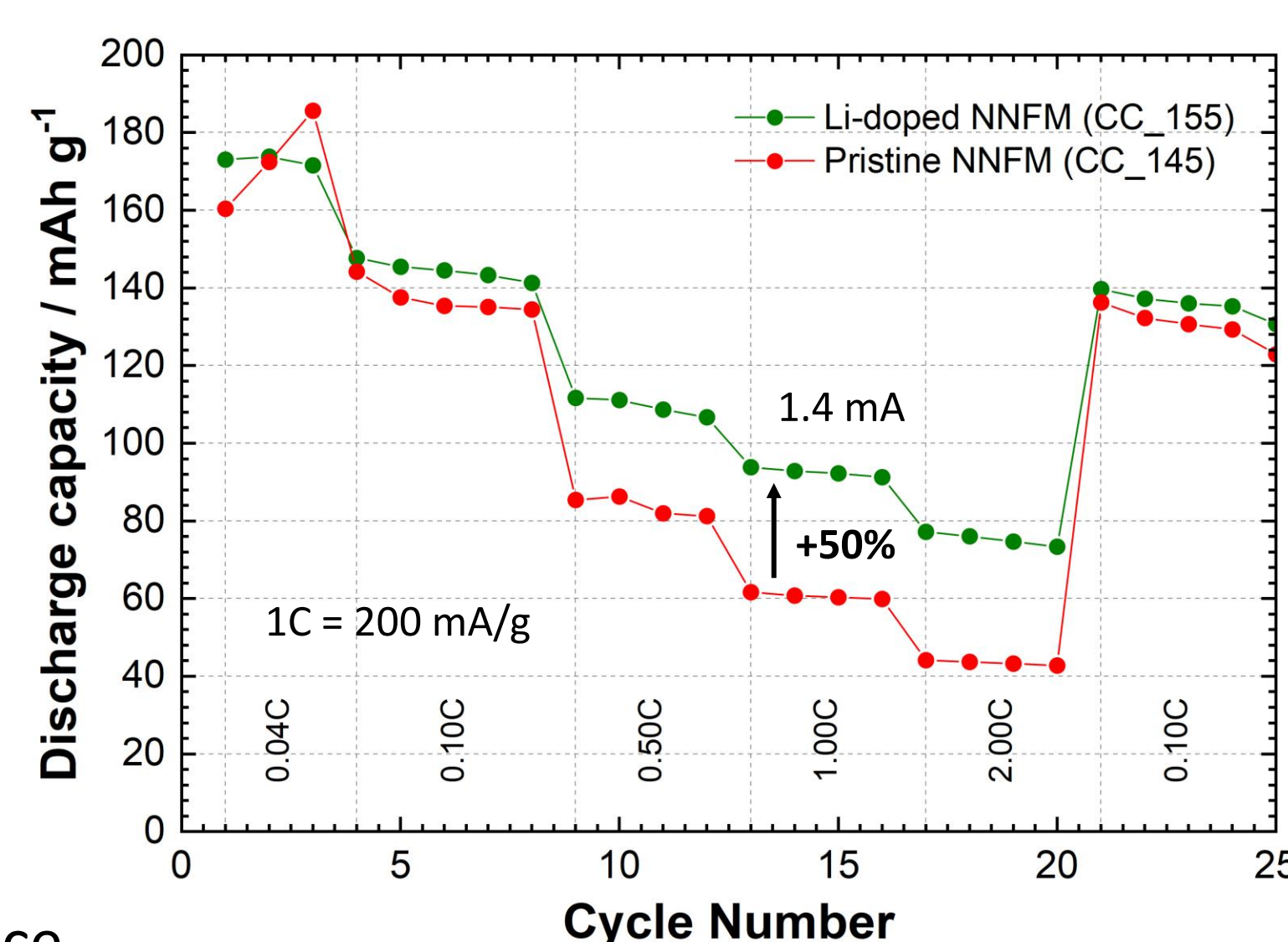
Conventional and solid-state Na-batteries

Conventional liquid electrolyte based batteries



Cathode fabrication

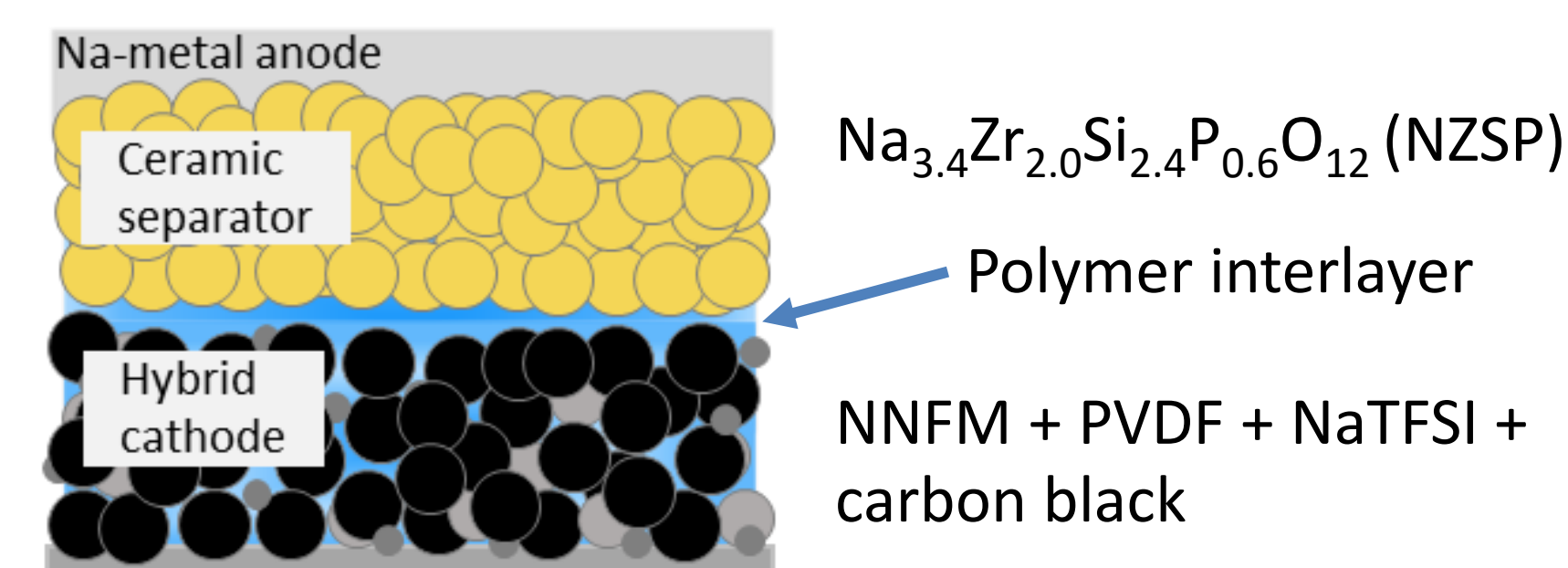
- Different cathode compositions
- For conventional cells: NNFM, carbon black, and PVDF (90:5:5 wt.%)
- For SSBs: NNFM, carbon black, PVDF, and NaTFSI (70:10:10:10 wt.%)



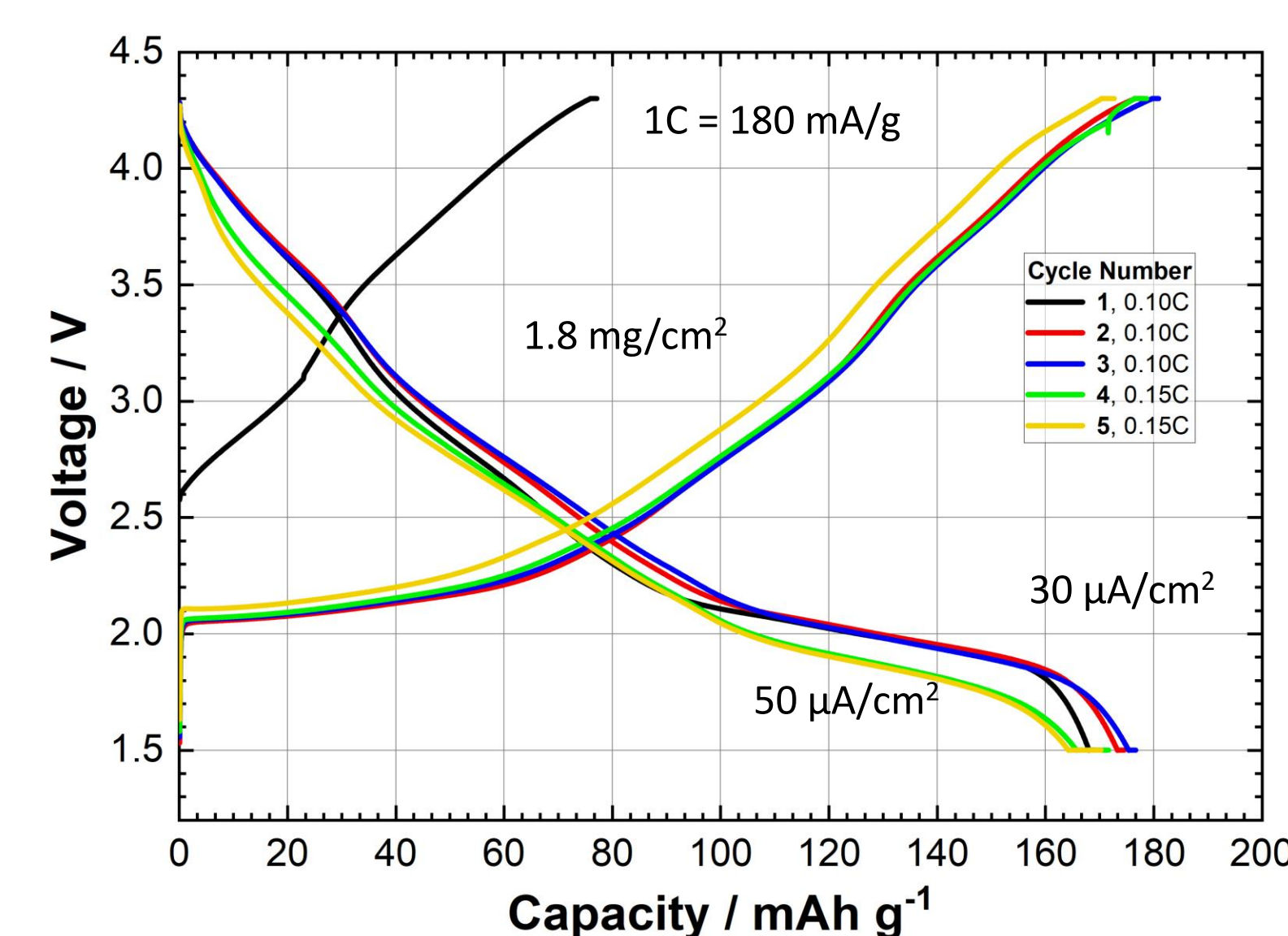
Cell type, p/ MPa	CR2032, ~0.2
Electrolyte	0.5 M NaPF ₆ , PC:FEC (98:2)
Φ_{CAM} / wt. %	90
q_A / mAh cm ⁻²	1.22
A / cm ²	1.77
$j_{\text{chg}}/j_{\text{dsch}}$ / mA cm ⁻²	0.03-1.63 (0.04C - 2.00C)
E / V vs. Na/Na ⁺	1.5 - 4.3
T / °C	25

- Li-doping improves the rate performance
- Li-doping most likely affects the transition metal interlayer ordering, which facilitates the Na^+ diffusion at high currents

Hybrid, polymer-ceramic all-solid-state battery



- The cathode was infiltrated with 20 μL polymer electrolyte (Bt-PCL + NaTFSI in THF) and attached to the NZSP separator (98% rel. density) using a polymer interlayer
- Vacuum drying to remove solvent residues (10^{-5} mbar) → fully dry all-SSBs



Cell type, p/ MPa	Swagelok, ~0.2
Electrolyte	NZSP; NaTFSI + Bt-PCL
Φ_{CAM} / wt. %	70
q_A / mAh cm ⁻²	0.32
A / cm ²	0.79
$j_{\text{chg}}/j_{\text{dsch}}$ / mA cm ⁻²	0.03, 0.05 (0.1C, 0.15C)
E / V vs. Na/Na ⁺	1.5 - 4.3
T / °C	60

- The first hybrid all-solid-state Na-batteries were successfully cycled
- Improving the cathode architecture and integrating the doped CAM toward high power and high capacity all-SSBs is ongoing

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