HYDRA WP5 PARAMETERIZATION

2022/08/10

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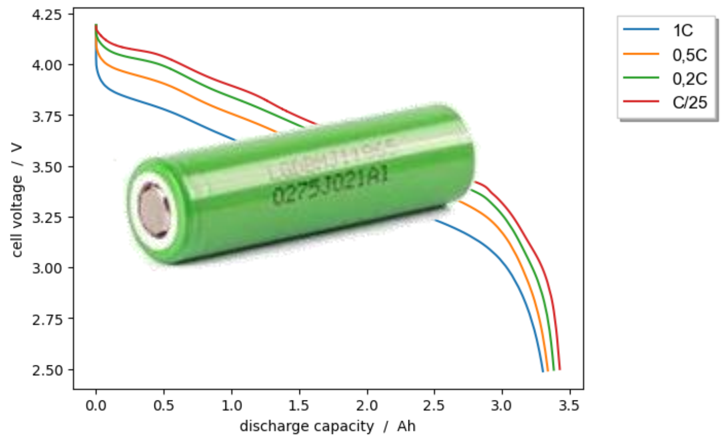
DLR

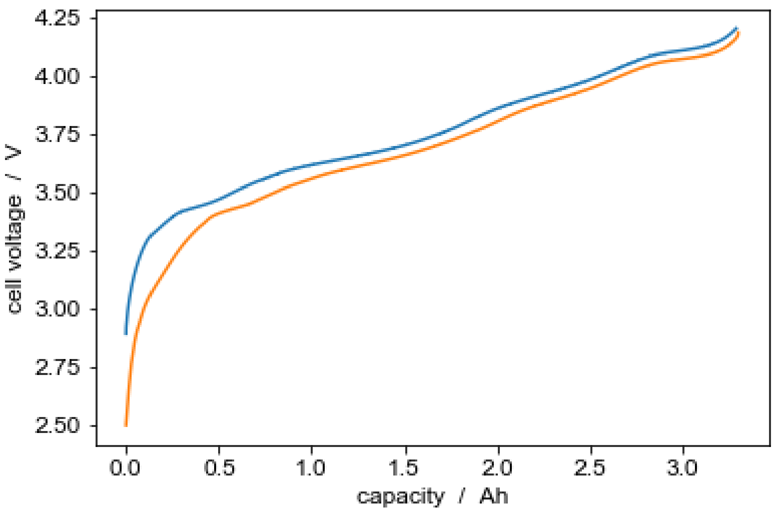
PARAMETRIZATION OF MJ1

LG MJ1

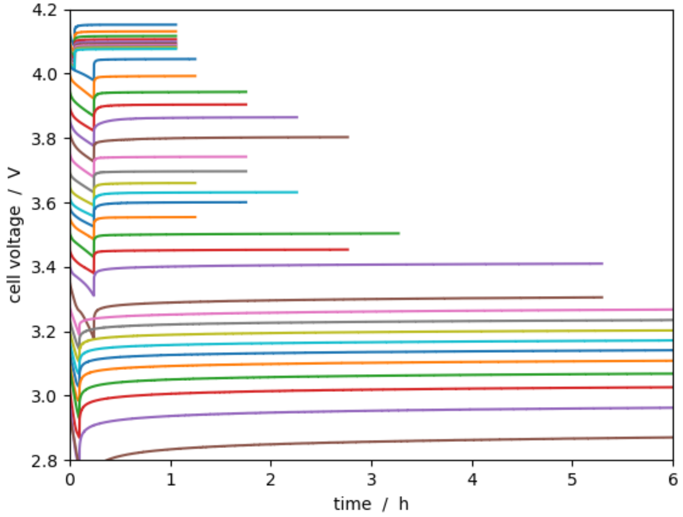
- <u>Nominal voltage</u>: 3.635 V / <u>nominal capacity</u>: 3500 mAh
- <u>Discharge energy</u>: 11.34 Wh @ 1C / 12.28 Wh @ 0.2C
- <u>Gravimetric energy density</u> (0.2C): 262 Wh/kg (weight: 46.93 g)
- <u>Volumetric energy density</u> (0.2C): 711 Wh/L (volume: 17.28 cm³)



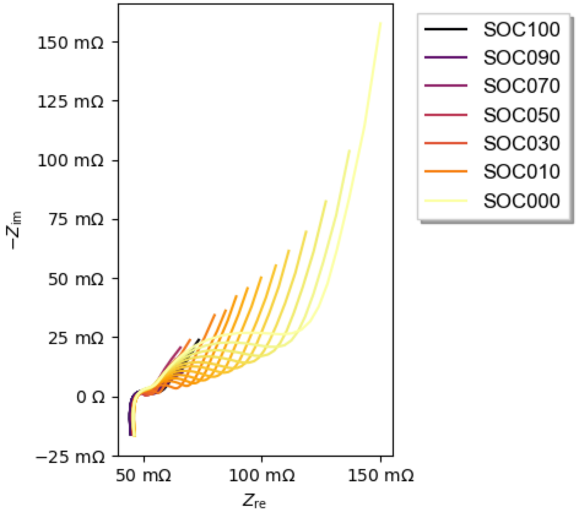
qOCV-curves



GITT (10, 25, 40 °C)

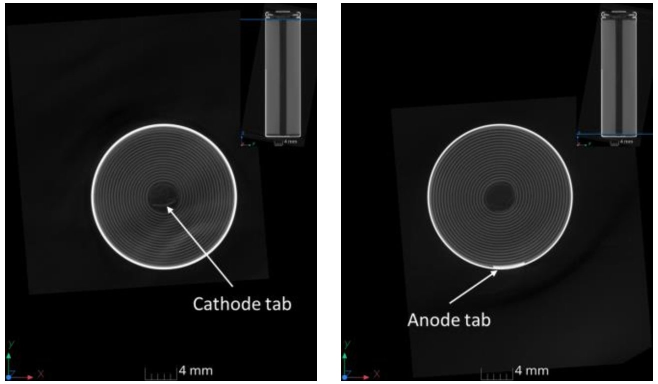


EIS (10, 25, 40 °C)

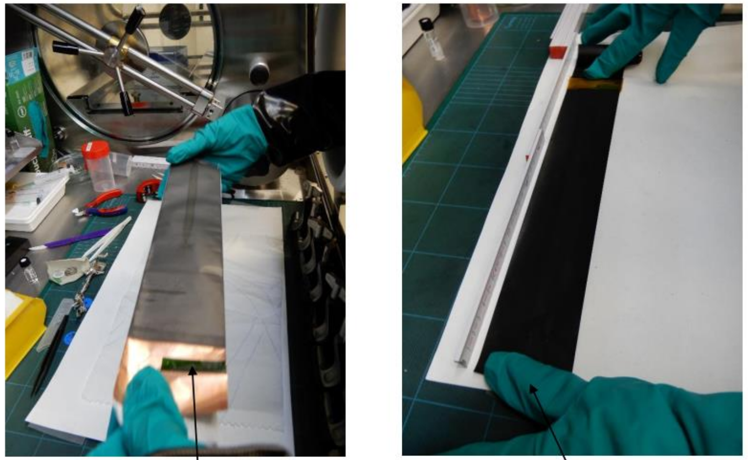


Geometry and dimensions

Computer tomography scans:



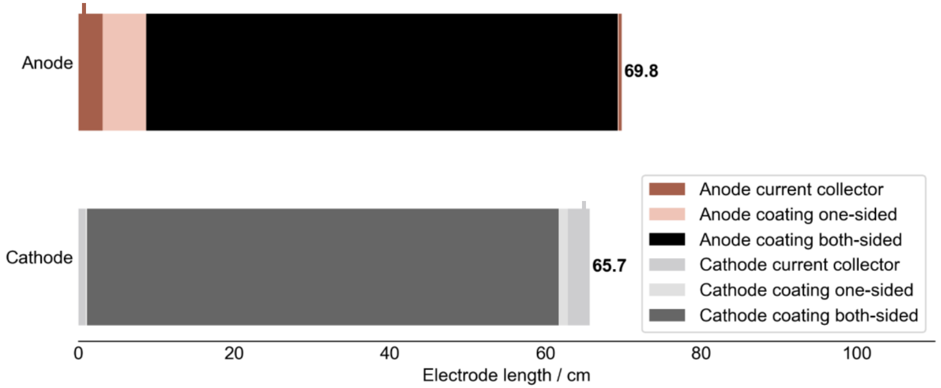
Anode | Cathode



Outer part: first current collector, then coating on one side, then coating on both sides (anode)

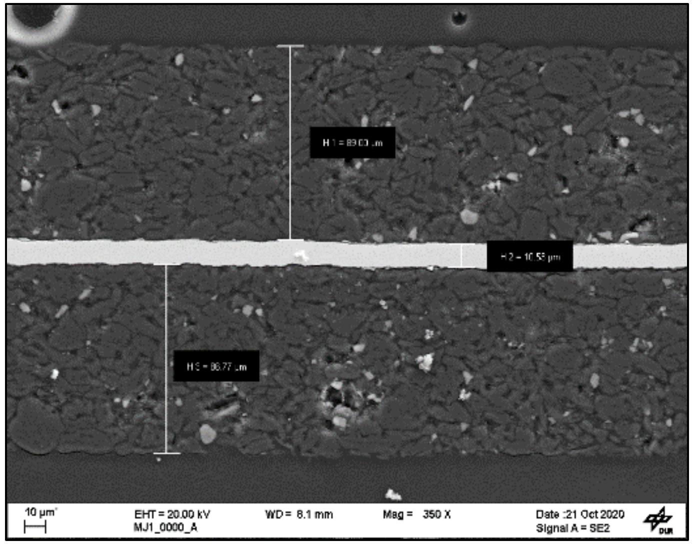
Outer part

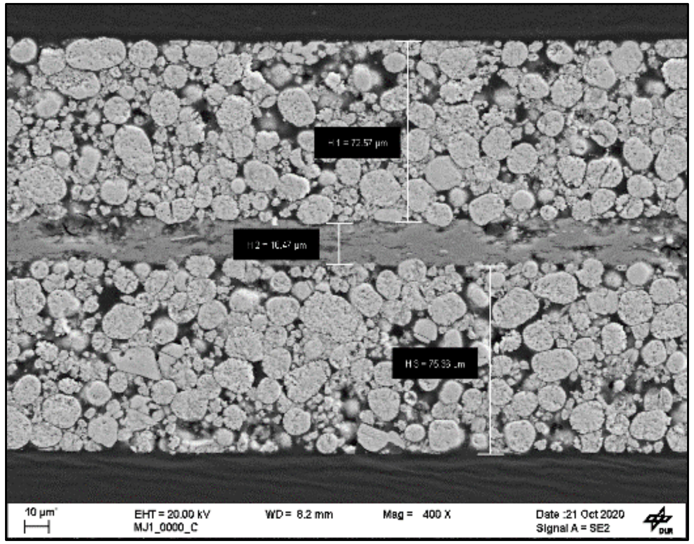
- Exact geometry is required to reproduce electrode arrangement and overlap in 3D-model
- Overlap of windings is not resolved at the moment



Geometry and dimensions

Anode | Cathode





<u>Thickness</u> (SEM of cross-section):

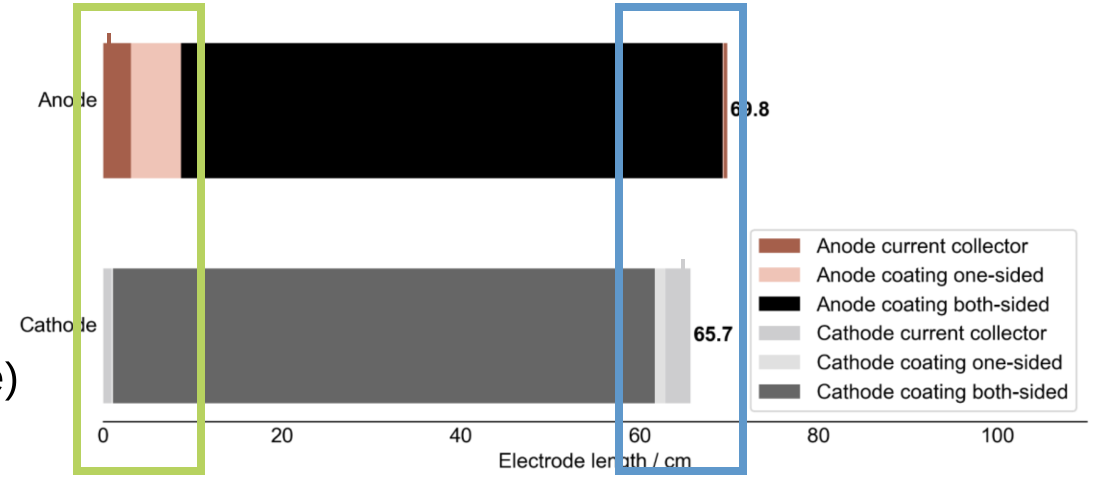
- Anode: 87 µm
- Cathode: 73 µm
- CC Anode: 10 µm
- CC Cathode: 16 µm
- Separator: 12 µm

<u>Area</u> (measured manually):

- Anode inside: 66.6 x 6 cm^2
  outside: 61 x 6 cm^2

- Cathode inside: 61.6 x 5.9 cm^2
  outside: 62.9 x 5.9 cm^2

- Length of anode foil inside: 3.1 cm / 0.4 cm
  Length of anode foil outside: 8.7 cm / 0.5 cm
- Length of cathode foil inside: 1.1 cm / 4 cm
  Length of cathodefoil outside: 0.9 cm / 2.8 cm
- Dimensions of tab: 6 x 0.4 cm^2 (anode), 5.9 x 0.3 cm^2 (cathode)



Electrode composition

Positive electrode:

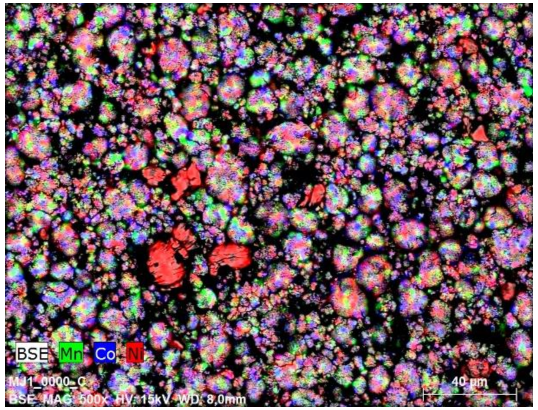
- Composition: $Ni_{0.83}$ $Mn_{0.05}$ $Co_{0.12}$ (avg. from ICP-OES (3 times) and SEM-EDX, see table)
- Active material content: 0.92 (typical for high energy cell)

		Ni	Mn	Co
ICP-OES	**at.-%**	83.66	4.93	11.40
SEM-EDX	**at.-%**	82.50	5.40	12.10

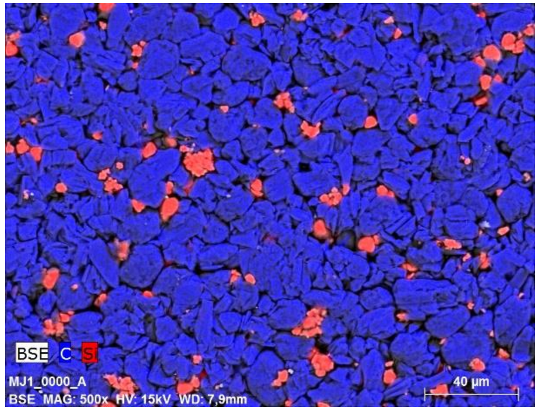
Negative electrode:

- Composition: Graphite/SiOx composite 96.5 : 3.5% (µCT)
- Active material content: 0.95 (typical for high energy cell)

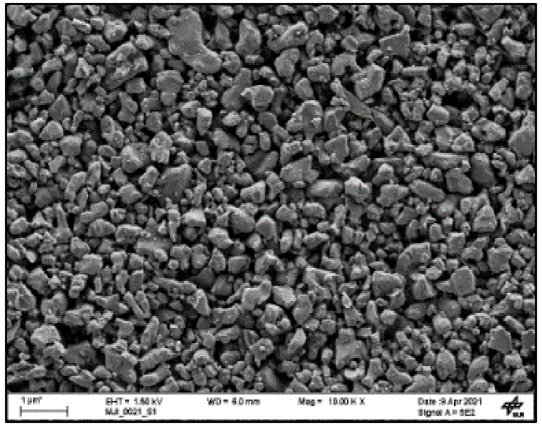
Positive electrode:
High-Ni NMC



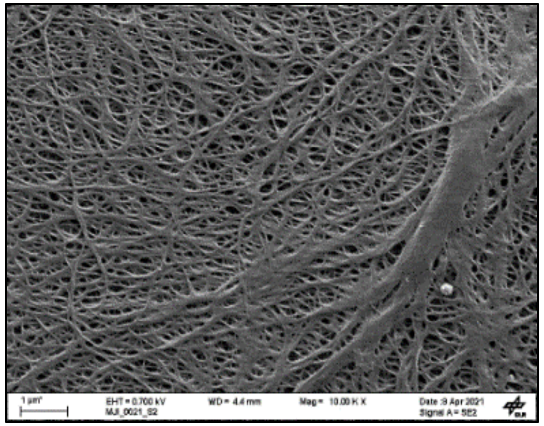
Negative electrode:
Graphite-SiOx



Separator: ceramic coating



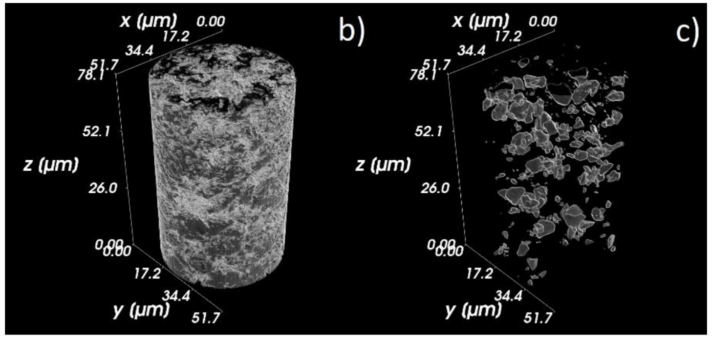
Separator: polymer matrix

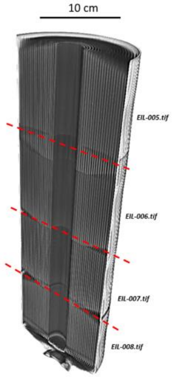


Structural properties – Negative electrode

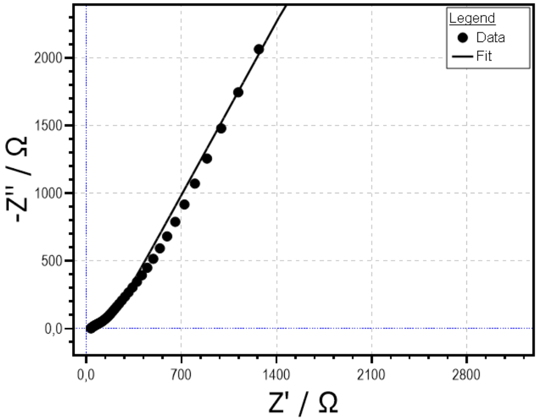
- Porosity:
  - **MIP:** **26.1 % mercury intrusion porosimetry**
  - µCT: 29.3 % 3D reconstruction from lit.
  - calc: 27.6 % via weight, thickness and density

$$\varepsilon = 1 - \frac{M_{coat}}{L\,\rho}$$



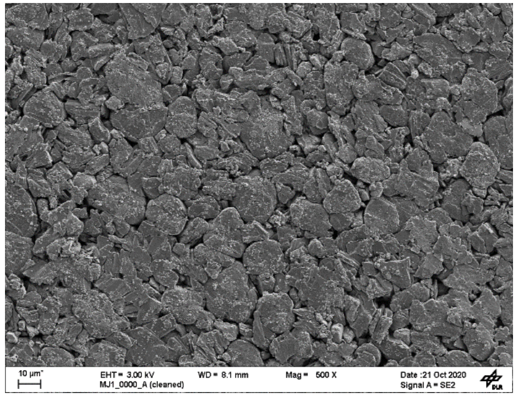


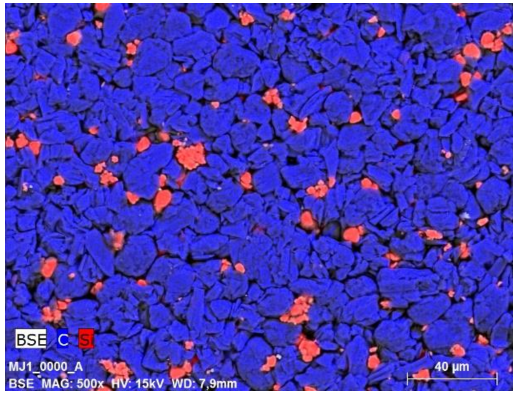
- Tortuosity: 4.6 ± 0.9 derived from EIS
- Mac Mullin number: 17.5 ± 3.5
- Bruggeman coefficient α: 1.1
- Mean particle diameter: 9.57 µm graphite
  2.25 µm SiOx
- BET surface area: $1.23\ m^2/g$
- Mass loading: $0.019\ g/cm^2$

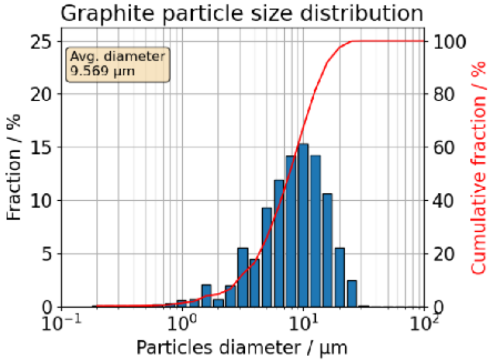


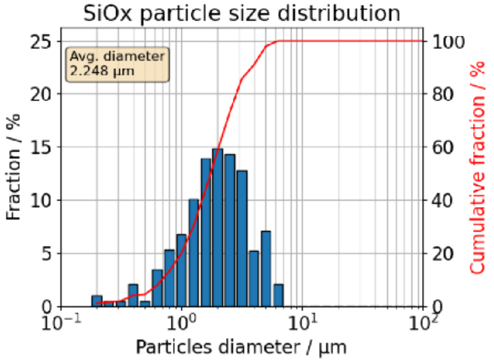
EIS under blocking conditions in 10 mM TBAClO4 in EC:EMC, Assumed porosity from MIP (26.1%)

$$\tau = \frac{\varepsilon \cdot R_{ion} \cdot A \cdot \sigma}{2d}$$









Structural properties – Positive electrode

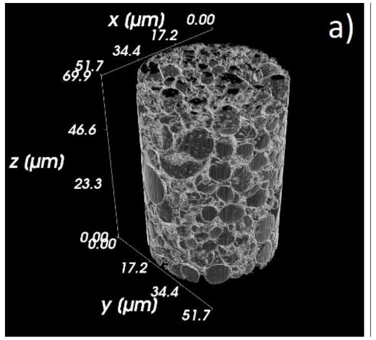
- Porosity:
  - **MIP:** **23.0 % mercury intrusion porosimetry**
  - µCT: 31.1 % 3D reconstruction from lit.
  - calc: 25.5 % via weight, thickness and density

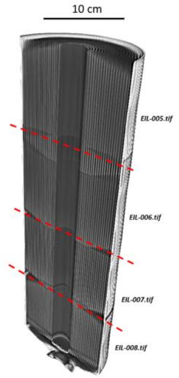
$$\varepsilon = 1 - \frac{M_{coat}}{L\,\rho}$$

- Tortuosity: 2.5 ± 0.1 derived from EIS
- Mac Mullin number: 11 ± 0.6
- Bruggeman coefficient α: 0.6
- Mean particle diameter: 4.75 µm
- BET surface area: 1.19m²/g
- Mass loading: 0.036 g/cm²

Separator:

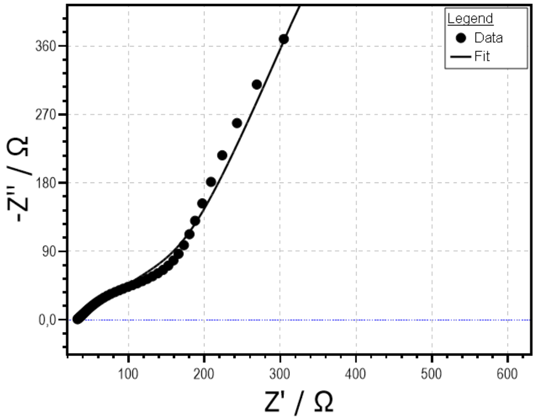
- Tortuosity: 4.64 ± 0.05 derived from EIS
- Mac Mullin number: 12.2 ± 0.1
- Bruggeman coefficient α: 1.59 ± 0.01





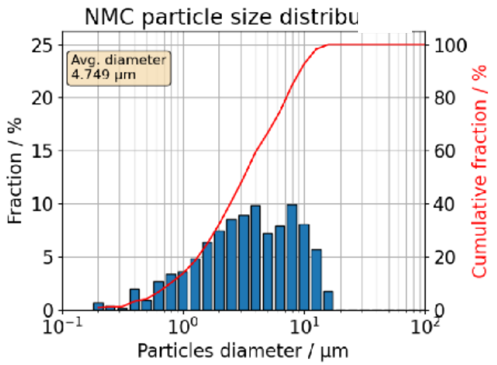
https://doi.org/10.1016/j.dib.2020.106033

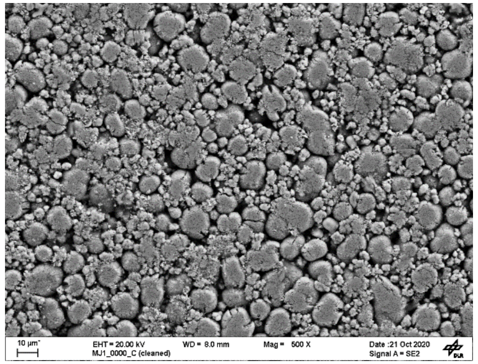
Reconstruction and evaluation performed by Roberto



EIS under blocking conditions in 10 mM TBAClO4 in EC:EMC,
Assumed porosity: 23%

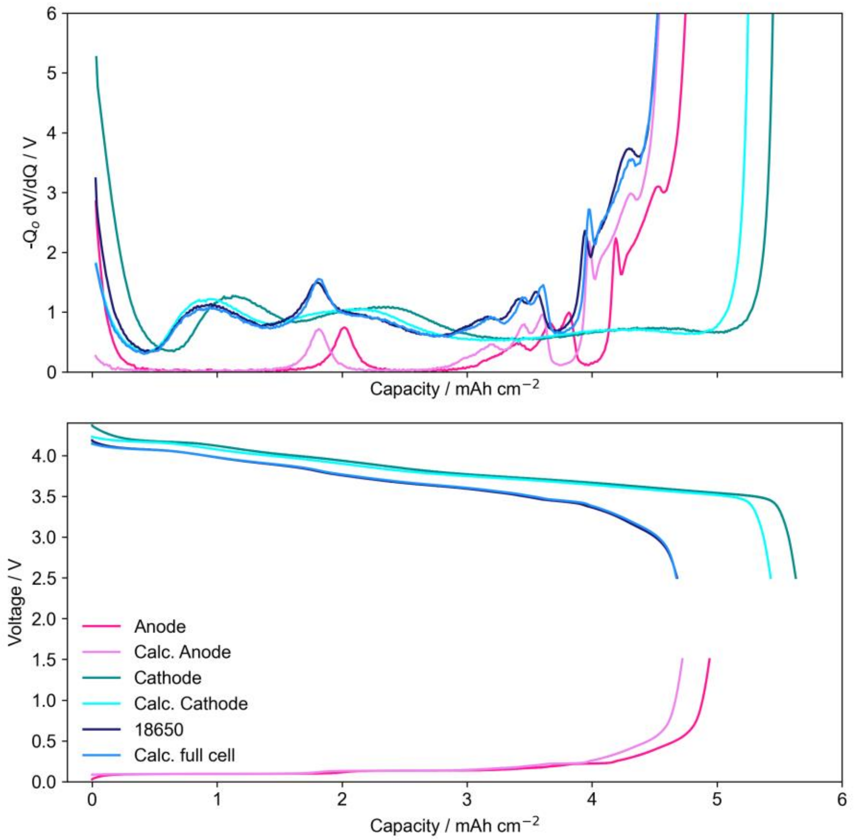
$$\tau = \frac{\varepsilon \cdot R_{ion} \cdot A \cdot \sigma}{2d}$$

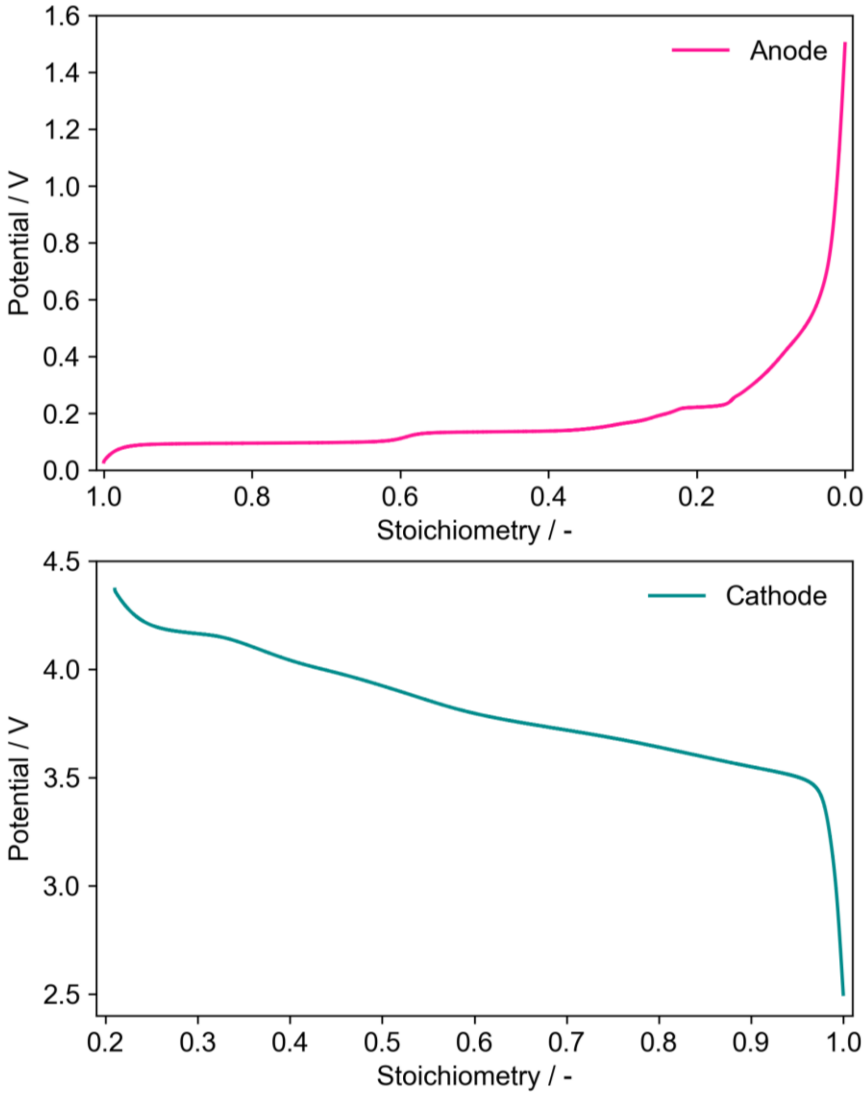




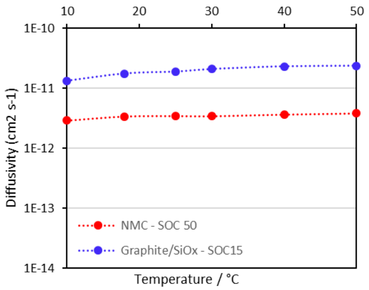
Thermodynamics and balancing

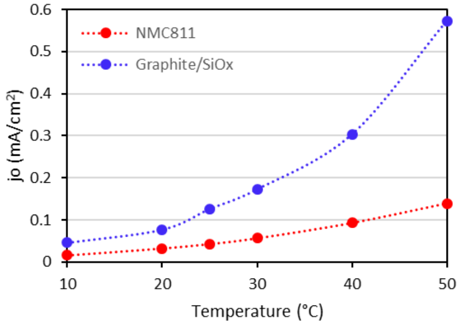
- Pseudo-OCV curves obtained @ C/50 charge/discharge
- Individual electrode OCV-curves shifted and scaled according to differential voltage (DV) curves
- Calculated full cell DV curve in accordance with DV curve from measured OCV of the 18650 cell





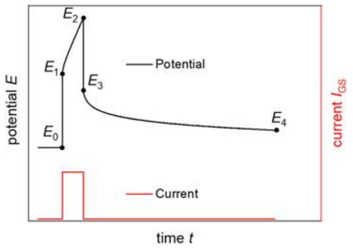
Kinetic parameters

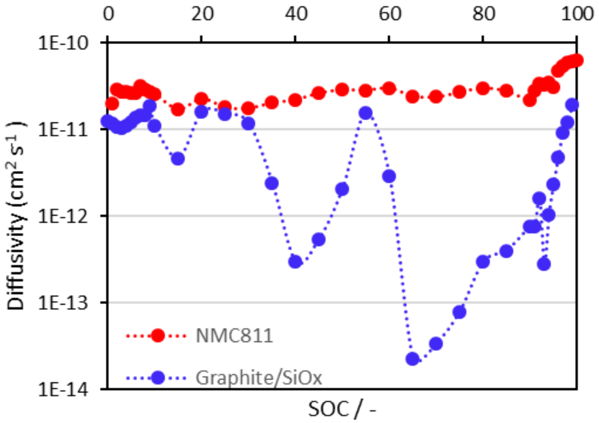


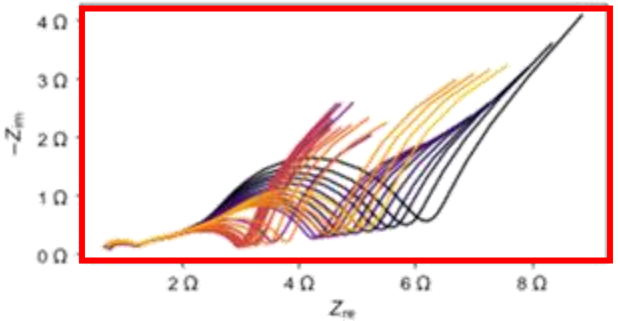


- **Solid-State Diffusion** determined by Galvanostatic Intermittent Titration Technique (GITT)

$$D = \frac{4}{9\pi}\left(\frac{E_4 - E_0}{E_3 - E_0}\right)^2 \cdot \frac{r_p^2}{t_p}$$

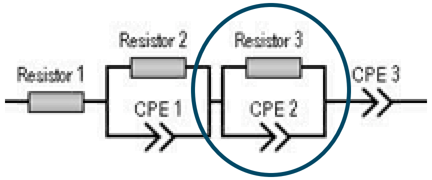




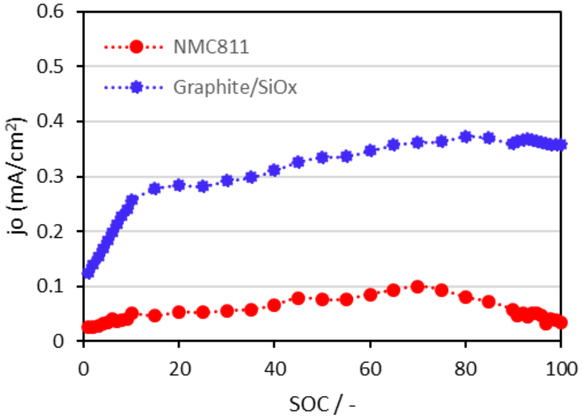


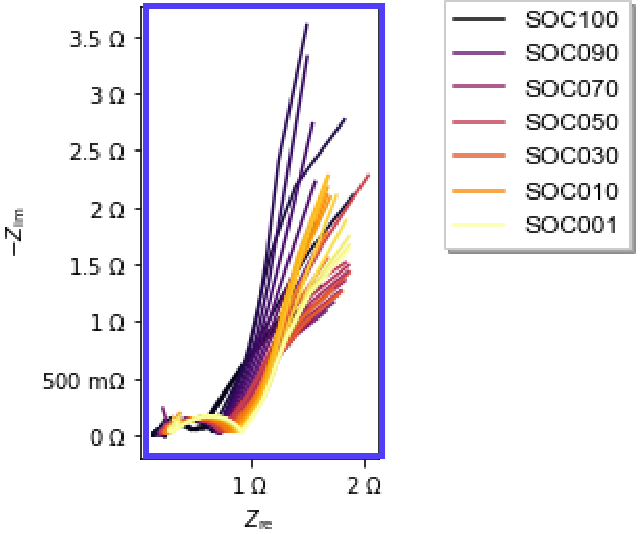
- **Exchange current density** determined via Impedance Spectroscopy in 3-electrode arrangement

$$R_{ct} = \frac{RT}{j_0 SF}$$



$$S = nSA_{particle} = \frac{\varepsilon_{AM} V_{electrode}}{V_{particle}} SA_{particle}$$





- **Activation energy** (j0 / Di) via Arrhenius analysis :
  - cathode: 40.5 / 5.63 kJ/mol @SOC50
  - anode: 47.8 / 13.82 kJ/mol @SOC15

Electrolyte transport parameters

- **Electrolyte type** revealed by Sturm et al. via GC-MS, (verified by contacting main author): $LiPF_6$ in EC/EMC/DMC, 1M and 1:1:1 ratio assumed
- **Ionic conductivity** κ for this specific composition replicated from literature (Schmalstieg et al.)

$$\sqrt{\frac{\kappa}{C_{salt}}} = a_1 + a_2 T + a_3 c_{salt} T + a_4 \exp(a_5 c_{salt})$$

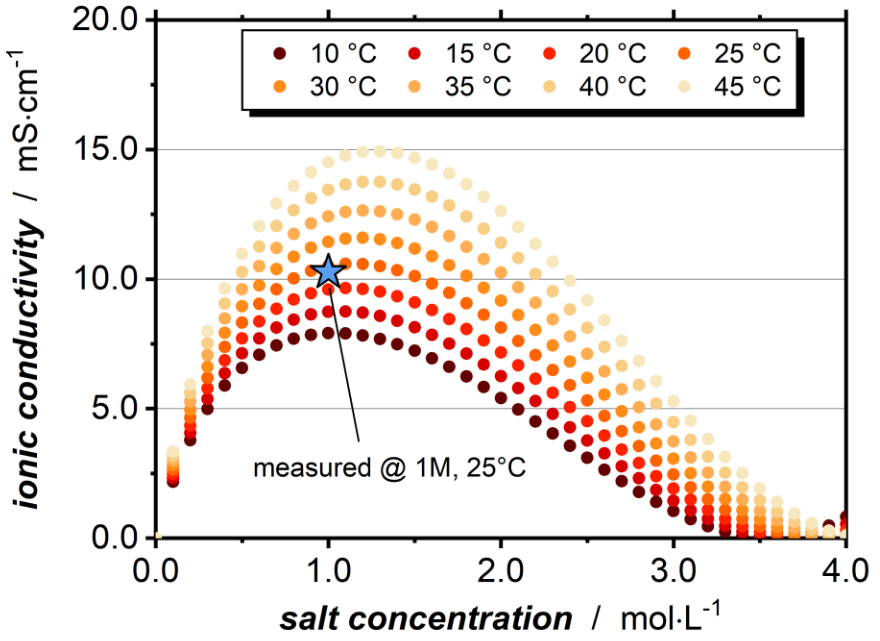
a1	a2	a3	a4	a5
-5.384	0.03213	-0.00368	1.320	-2.235

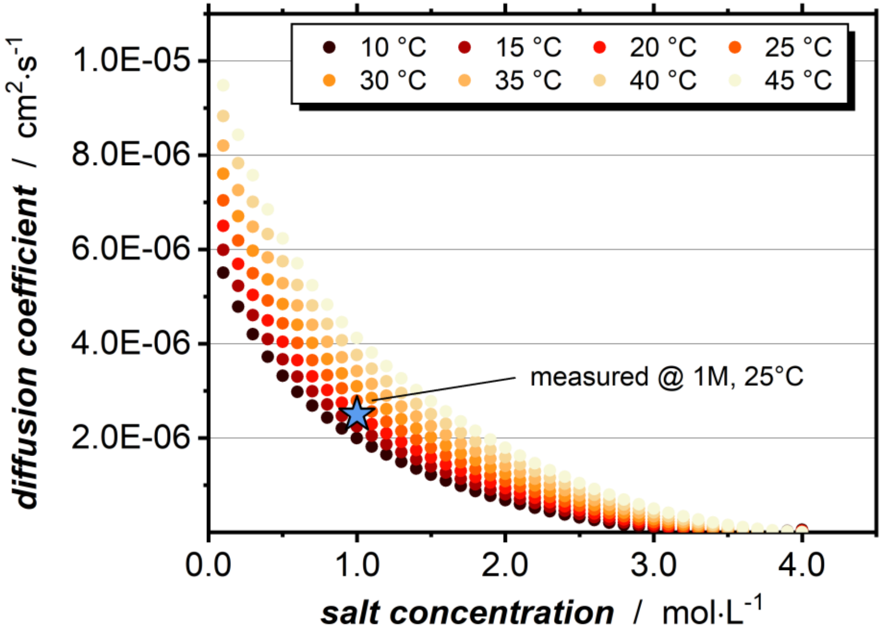
- **Diffusion coefficient** estimated via Einstein relationship

$$D_e = \frac{\kappa R T}{F^2 z^2 c_{salt}}$$

(only valid in dilute solutions, but results very consistent with literature)

- *Measured* values (D_e via current pulse, κ via EIS) fully consistent with other data (see graph)
- **Transference number** $t_+ = 0.226$ measured (galv. Polarization), assumed independent on c_{salt} and T in relevant range





J. Sturm *et al.* (2019) *J. Power Sources* **412** 204–223
J. Schmalstieg *et al* (2018) *J. Electrochem. Soc.* **165** A3799