Supplementary

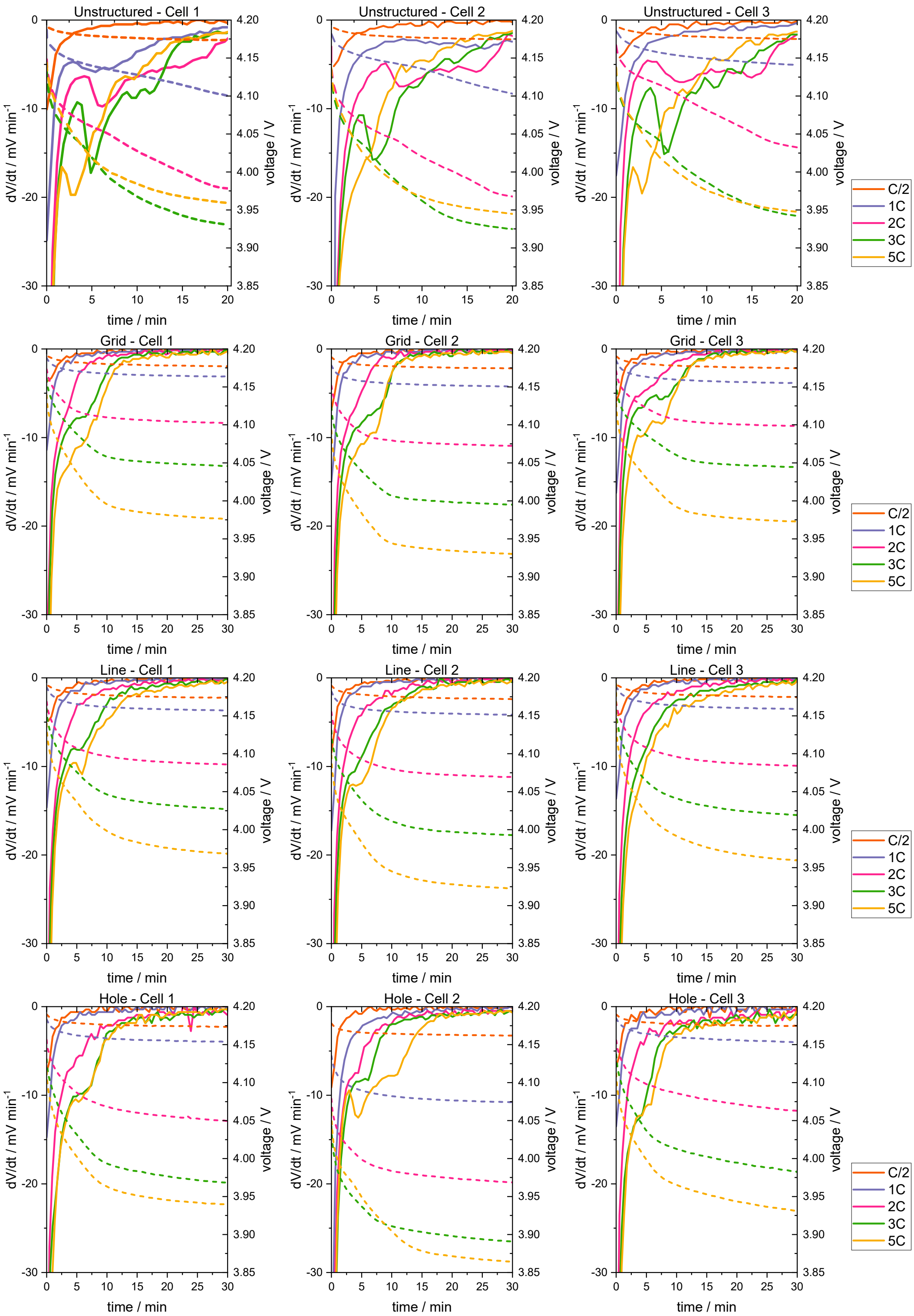

Figure S1. Differential voltage analysis of the voltage relaxation (solid line) and voltage as a function of time (dashed line) in the 4 h rest period after charging in the 5th cycle at each C-rate for all cells with unstructured and structured electrodes.

Figure S2. Separator of the cell with unstructured electrodes with plated lithium sticking on it (cathode visible on the backside)

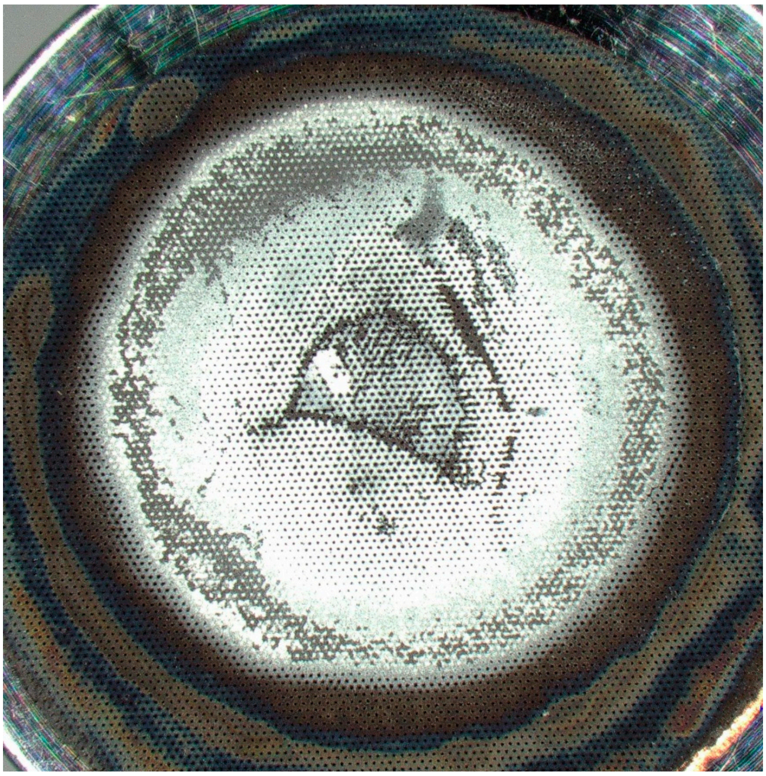

Figure S3. Post-mortem digital microscope image of the hole structured electrode with overgrown structures visible in the edge of the plated area.

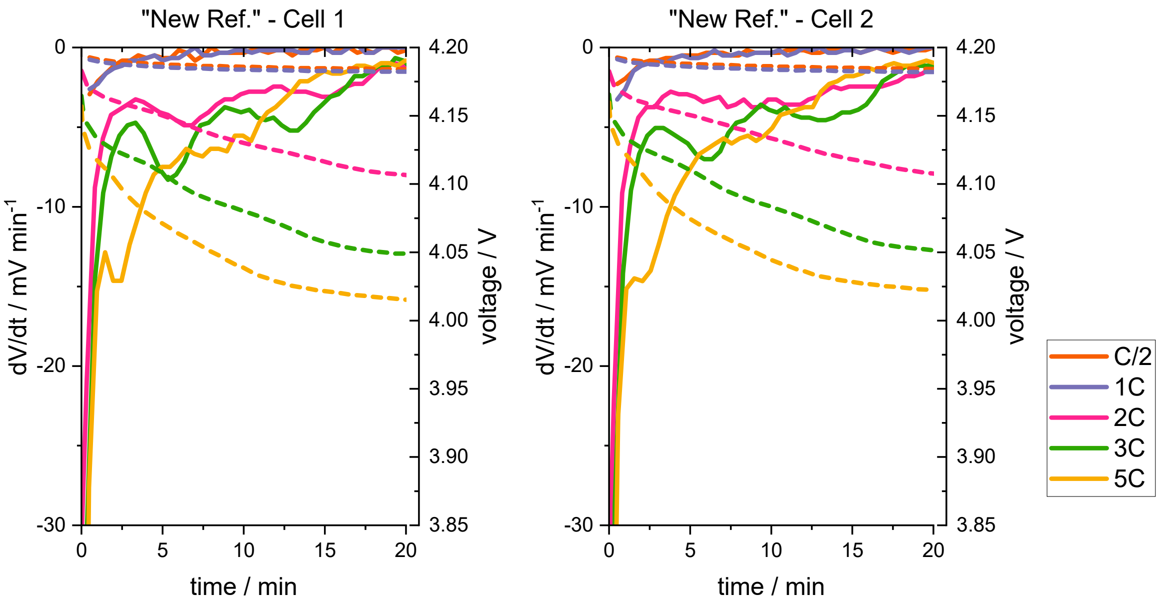

Figure S4. Differential voltage analysis of the voltage relaxation (solid line) and voltage as a function of time (dashed line) in the 4 h rest period after charging in the 5th cycle at each C-rate for the “New Ref.” cells.

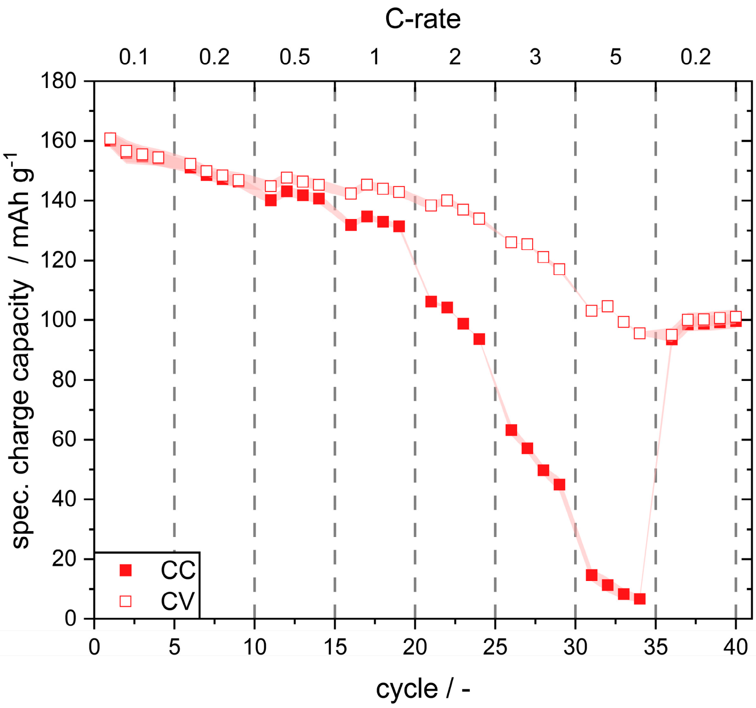

Figure S5. Specific charge capacity of the “New. Ref.” cells after the CC and CCCV phase (without the 5th cycle at each C-rate). The shading represents the standard deviation.

Table S1. Abbreviations and data for calculating the energy density and specific energy.

A_A [m^{-2}]	Area anode	0.049664 [52]
A_C [m^{-2}]	Area cathode	0.04845 [52]
A_S [m^{-2}]	Area separator	0.0517 [52]
B [-]	Balancing factor	1.14-1.17 (experiment)
d [m]	Coating thickness	-
d_A [m]	Anode coating thickness	calculated; Equation (S3)
d_{Al} [m]	Cathode current collector thickness	20e-6
d_C [m]	Cathode coating thickness	calculated; Equation (S5)
d_{Cu} [m]	Anode current collector thickness	9e-6
d_S [m]	Separator thickness	25e-6 (experiment)
d_{stack} [m]	Electrode stack thickness	calculated; Equation (S9)
E_m [Wh kg^{-1}]	Specific energy	calculated; Equation (S18)
E_V [Wh m^{-3}]	Energy density	calculated; Equation (S17)
F_E [-]	Electrolyte factor	2 [31]
m_A [kg]	Anode mass	calculated; Equation (S11)
m_C [kg]	Cathode mass	calculated; Equation (S12)
m_E [kg]	Electrolyte mass	calculated; Equation (S13)
$m_{electrode}$ [kg m^{-2}]	Area density electrode coating	-
m_S [kg]	Separator mass	calculated; Equation (S14)
m_{stack} [kg]	Stack mass	calculated; Equation (S15)
n_A [-]	Number of anodes	calculated; Equation (S7)
n_C [-]	Number of cathodes	calculated; Equation (S6)
n_S [-]	Number of separators	calculated; Equation (S8)
$Q_{A,A}$ [Ah m^{-2}]	Areal capacity anode	43.3-45.2 (experiment)
$Q_{A,C}$ [Ah m^{-2}]	Areal capacity cathode	calculated; Equation (S4)
Q_{cell} [Ah]	Cell capacity	>80
$Q_{spec,A}$ [Ah kg^{-1}]	Specific capacity anode active material	330 (experiment)
$Q_{spec,C}$ [Ah kg^{-1}]	Specific capacity cathode active material	172 (experiment)
S_A [%]	Mass loss anode	9.7-11 (experiment)
S_C [%]	Mass loss cathode	0
U_{cell} [V]	Nominal cell voltage	3.7 [1]
ε_A [-]	Anode porosity	0.4 (calculated; Equation (S2))
ε_C [-]	Cathode porosity	0.35 (calculated; Equation (S2))
ε_S [-]	Separator porosity	0.55 [42]
θ_A [-]	Anode active material mass fraction	0.93 (experiment)
θ_C [-]	Cathode active material mass fraction	0.92 (experiment)
θ_n [-]	Individual mass fractions of the electrode coating materials	-
ρ_{Al} [kg m^{-3}]	Aluminium density	2700
ρ_{Cu} [kg m^{-3}]	Copper density	8920
ρ_E [kg m^{-3}]	Electrolyte density	1260 [8]
ρ_n [kg m^{-3}]	Individual densities of the electrode coating materials	-
$\rho_{pmd,A}$ [kg m^{-3}]	Anode particle mass density	2090 (calculated; Equation (S1))
$\rho_{pmd,C}$ [kg m^{-3}]	Cathode particle mass density	4170 (calculated; Equation (S1))
ρ_S [kg m^{-3}]	Separator material density	900

$$\rho_{pmd} = \frac{1}{\sum_n \frac{\theta_n}{\rho_n}} \tag{S1}$$

$$\varepsilon = 1 - \frac{\frac{m_{electrode}}{d}}{\rho_{pmd}} \quad \text{(S2)}$$

$$d_A = \frac{Q_{A,A}}{\rho_{pmd,A} \cdot \theta_{Ac,A} \cdot Q_{spec,A} \cdot (1-\epsilon_A) \cdot (1-S_A)} \quad \text{(S3)}$$

$$Q_{A,C} = \frac{Q_{A,A}}{B} \quad \text{(S4)}$$

$$d_C = \frac{Q_{A,C}}{\rho_{pmd,C} \cdot \theta_{AC,C} \cdot Q_{spec,C} \cdot (1-\epsilon_C) \cdot (1-S_C)} \quad \text{(S5)}$$

$$n_C = x \quad \text{(S6)}$$

$$n_A = n_C + 1 \quad \text{(S7)}$$

$$n_S = 2 \cdot n_A \quad \text{(S8)}$$

$$d_{stack} = n_C \cdot (d_{Al} + 2 \cdot d_C) + n_A \cdot (d_{Cu} + 2 \cdot d_A) + n_S \cdot d_S \quad \text{(S9)}$$

$$V_{stack} = A_A \cdot d_{stack} \quad \text{(S10)}$$

$$m_A = A_A \cdot n_A \cdot \left(d_{Cu} \cdot \rho_{Cu} + 2 \cdot d_A \cdot (1-\epsilon_A) \cdot \rho_{pmd,A} \cdot (1-S_A)\right) \quad \text{(S11)}$$

$$m_C = A_C \cdot n_C \cdot \left(d_{Al} \cdot \rho_{Al} + 2 \cdot d_C \cdot (1-\varepsilon_C) \cdot \rho_{pmd,C} \cdot (1-S_C)\right) \quad \text{(S12)}$$

$$m_E = \big(n_S \cdot \varepsilon_S \cdot A_S \cdot d_S + 2 \cdot n_A \cdot (d_A \cdot \varepsilon_A \cdot A_A \cdot (1-S_A) + d_A \cdot A_A \cdot S_A) + 2 \cdot n_C \cdot (d_C \cdot \varepsilon_C \cdot A_C \cdot (1-S_C) + d_C \cdot A_C \cdot S_C)\big) \cdot \rho_E \cdot F_E \quad \text{(S13)}$$

$$m_S = n_s \cdot A_s \cdot (1-\varepsilon_S) \cdot \rho_S \quad \text{(S14)}$$

$$m_{stack} = m_A + m_C + m_S + m_E \quad \text{(S15)}$$

$$Q_{cell} = 2 \cdot n_C \cdot A_C \cdot Q_{A,C} \quad \text{(S16)}$$

$$E_V = \frac{U_{cell} \cdot Q_{cell}}{V_{stack}} \quad \text{(S17)}$$

$$E_m = \frac{U_{cell} \cdot Q_{cell}}{m_{stack}} \quad \text{(S18)}$$

Generalized finite Warburg element Equation (S19) and the transformation into the transmission line model Equations (S20-S22) (imaginary unit i, angular frequency ω, ionic resistance R_{Ion}, constant-phase capacitance Q_s and the constant phase exponent α)

$$Z_{GFW} = R \cdot \frac{\coth((i \cdot T \cdot \omega)^P)}{(i \cdot T \cdot \omega)^P} \quad \text{(S19)}$$

$$R = R_{ion} \quad \text{(S20)}$$

$$T = (R_{Ion} \cdot Q_s)^{\frac{1}{\alpha}} \quad \text{(S21)}$$

$$P = \frac{\alpha}{2} \quad \text{(S22)}$$