

Benchmarking the reproducibility of all-solid-state battery cell performance

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Table of Contents

1. Supplementary Tables

Supplementary Table 1. All-solid-state battery (ASSB) cell assembly parameters.	2
Supplementary Tables 2 - 20. Cell data of groups A to U.	3
Supplementary Table 21. In:Li ratios of the In/(InLi) _x alloy negative electrodes.	6
Supplementary Table 22. Parameters obtained from the DRT analysis.	7
Supplementary Table 23. Statistical results from the violin plots.	8
Supplementary Table 24. Elemental composition of the cathode active material.	9
Supplementary Table 25. Temperature-dependent conductivity of Li ₆ PS ₅ Cl.	9
Supplementary Table 26. PEIS measurement parameters.	9

2. Supplementary Figures

Supplementary Figure 1. Average initial discharge capacities per group.	10
Supplementary Figure 2. Specific discharge capacities of all working cells.	11
Supplementary Figure 3. Coulomb efficiencies of all working cells.	12
Supplementary Figure 4. Polarization voltages of all working cells.	13
Supplementary Figure 5. Accumulated irreversible capacities of all working cells.	14
Supplementary Figure 6. Violin plot of the capacity retentions.	15
Supplementary Figure 7. Definition of best performers and polarization evolution.	15
Supplementary Figure 8. Resistances obtained from DRT analysis.	16
Supplementary Figure 9. Correlation of resistance and cycling.	16
Supplementary Figure 10. DRT analysis of the PEIS spectrum of group K.	17
Supplementary Figures 11-27. Charge-/discharge curves of the groups A to U.	18
Supplementary Figure 28. Coin cell cycling results.	24
Supplementary Figures 29-43. Impedance spectra and DRT analysis of group D to U.	25
Supplementary Figure 44. X-ray diffraction (XRD) patterns.	40
Supplementary Figure 45. Scanning electron microscope (SEM) images of Li ₆ PS ₅ Cl.	40
Supplementary Figure 46. Scanning electron microscope (SEM) images of LiNi _{0.6} Mn _{0.2} Co _{0.2} O ₂ .	40
Supplementary Figure 47. Arrhenius plot for the pristine Li ₆ PS ₅ Cl.	41

1. Supplementary Tables

Supplementary Table 1. All-solid-state battery (ASSB) cell assembly parameters. Overview of the different ASSB cell dimensions, preparation parameters and cycling parameters used by the different groups.

		A	B	D	E	F	G	H	J*	K	L	M	N†	O	P	Q	R	S	T	U
Electrode diameter / mm		10	10	10	10	9.5	5	10	10	10	10	7	10	8	10	7	10	10	13	13
Separator	Pressure / MPa	100	hand-pressed	300	127	350	50	70	460	62	370	255	395	590	720	380	98	100	25	72
	Compression time / min	0.02	-	-	1	5	1	1	3.5	1	1	10	3	5	3	1	1	1	0.5	1
Cathode composite (CC)	Pressure / MPa	370	380	350	304	350	50	300	400	434	370	255	395	590	720	510	395	380	-	370
	Compression time / min	3	3	2	5	5	1	3	3.5	3	1	10	3	5	3	1	1	3	-	3
Complete battery cell	Pressure / MPa	100	-	10	50	25	400	hand-pressed	100	-	150	50	55	195	36	400	68	-	360	40
	Compression time / min	0.02	-	0	1	5	5	-	2	-	0.5	0.05	-	5	2	-	-	-	1	3
Cycling pressure / MPa		70	50	-	50	10	1	60	60	81	75	50	55	40	50	400	68	50	25	70
Pressure control		N	N	N	N	N	N	N	Y	N	N	N	N	Y	Y	N	Y	N	N	N
Cycling temperature / °C		25	25	25	25	25	25	25	25	25	23-26	25	25	25	25	23-25	24-26	25	25	30
Time elapsed between cell assembly and OCV measurement / h		12	5	0.5	1.67/0.83	12	0.5	0.17	3	0.25	0.2	2	3	2	24	0.5	4.25/6.5/0.3	0.5	0.5	6

* Cycling was performed between 3.0 V and 4.3 V vs. In/(InLi)_x, corresponding to a potential window of 3.62 V and 4.92 V vs. Li⁺/Li.

† Cycling was performed with 0.072C instead of 0.1C.

Supplementary Tables 2 –20 show the thicknesses l , weights m , loadings, OCVs and currents I used by every group. Abbreviations are used for the solid electrolyte separator (SE), positive composite electrode (CC) and positive electroactive material (CAM). Provided data is shown in black, calculated or assumed values are shown in blue.

Supplementary Table 2. Cell data of group A.

Cell ID	$l_{\text{In/InLi}} / \mu\text{m}$	$l_{\text{SE}} / \mu\text{m}$	$l_{\text{CC}} / \mu\text{m}$	$m_{\text{Li}} / \text{mg}$	$m_{\text{In}} / \text{mg}$	$m_{\text{SE}} / \text{mg}$	$m_{\text{CC}} / \text{mg}$	$m_{\text{CAM}} / \text{mg}$	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	I (0.1C) / mA
A1	130	500	60	1.8	36	58.8	11.2	7.8	10.0	1.803	0.157
A2	130	500	60	1.8	36	58.8	11.2	7.8	10.0	1.845	0.157

Supplementary Table 3. Cell data of group B.

Cell ID	$l_{\text{In/InLi}} / \mu\text{m}$	$l_{\text{SE}} + l_{\text{CC}} / \mu\text{m}$	$m_{\text{Li}} / \text{mg}$	$m_{\text{In}} / \text{mg}$	$m_{\text{SE}} / \text{mg}$	$m_{\text{CC}} / \text{mg}$	$m_{\text{CAM}} / \text{mg}$	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	I (0.1C) / mA
B1	150	500	1.5	47	60.09	12	8.40	10.7	2.6488	0.168
B2	150	460	1.5	47	60.01	11.93	8.35	10.6	2.6704	0.167
B3	150	480	1.5	47	60.20	11.98	8.39	10.7	2.6484	0.168

Supplementary Table 4. Cell data of group D.

Cell ID	$l_{\text{In/InLi}} / \mu\text{m}$	$l_{\text{SE}} + l_{\text{CC}} / \mu\text{m}$	$m_{\text{Li}} / \text{mg}$	$m_{\text{In}} / \text{mg}$	$m_{\text{SE}} / \text{mg}$	$m_{\text{CC}} / \text{mg}$	$m_{\text{CAM}} / \text{mg}$	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	I (0.1C) / mA
D1	100	500	0.67	48.2	60	11.2	7.8	10.0	2.570	0.156
D2	100	500	0.67	48.2	60	11.2	7.8	10.0	2.580	0.156
D3	100	500	0.67	48.2	60	11.2	7.8	10.0	2.710	0.156

Supplementary Table 5. Cell data of group E.

Cell ID	$l_{\text{In/InLi}} / \mu\text{m}$	$l_{\text{SE}} / \mu\text{m}$	$l_{\text{CC}} / \mu\text{m}$	$m_{\text{Li}} / \text{mg}$	$m_{\text{In}} / \text{mg}$	$m_{\text{SE}} / \text{mg}$	$m_{\text{CC}} / \text{mg}$	$m_{\text{CAM}} / \text{mg}$	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	I (0.1C) / mA
E1	200	500	50	1.1	62.9	58.7	11.5	8.1	10.3	2.354	0.161
E2	200	500	50	1.1	62.9	58.5	11.8	8.3	10.5	2.290	0.165

Supplementary Table 6. Cell data of group F.

Cell ID	$l_{\text{In/InLi}} / \mu\text{m}$	$l_{\text{SE}} / \mu\text{m}$	$l_{\text{CC}} / \mu\text{m}$	$m_{\text{Li}} / \text{mg}$	$m_{\text{In}} / \text{mg}$	$m_{\text{SE}} / \text{mg}$	$m_{\text{CC}} / \text{mg}$	$m_{\text{CAM}} / \text{mg}$	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	I (0.1C) / mA
F1	195	450	70	1	88	54	10	7.0	9.9	2.122	0.15
F2	195	450	70	1.2	92.16	53.51	10.7	7.5	10.5	1.822	0.15

Supplementary Table 7. Cell data of group G.

Cell ID	$l_{\text{In/InLi}} / \mu\text{m}$	$l_{\text{SE}} / \mu\text{m}$	$l_{\text{CC}} / \mu\text{m}$	$m_{\text{Li}} / \text{mg}$	$m_{\text{In}} / \text{mg}$	$m_{\text{SE}} / \text{mg}$	$m_{\text{CC}} / \text{mg}$	$m_{\text{CAM}} / \text{mg}$	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	I (0.1C) / mA
G1	-	-	-	0.61	13.73	16.03	2.94	2.1	10.5	2.384	0.0412
G2	-	-	-	0.55	14.18	14.08	3.1	2.2	11.1	2.473	0.0434
G3	-	-	-	0.51	13.67	14.76	2.7	1.9	9.6	2.209	0.0378

Supplementary Table 8. Cell data of group H.

Cell ID	$I_{In}/I_{InLi} / \mu\text{m}$	$I_{SE} / \mu\text{m}$	$I_{CC} / \mu\text{m}$	m_{Li} / mg	m_{In} / mg	m_{SE} / mg	m_{CC} / mg	m_{CAM} / mg	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	$I / (0.1C) / \text{mA}$
H1	200	452	58	0.51	55.78	60	12	8.4	10.7	2.620	0.168
H2	200	451	77	0.51	55.78	60	12	8.4	10.7	2.640	0.168

Supplementary Table 9. Cell data of group J.

Cell ID	$I_{In}/I_{InLi} / \mu\text{m}$	$I_{SE} / \mu\text{m}$	$I_{CC} / \mu\text{m}$	m_{Li} / mg	m_{In} / mg	m_{SE} / mg	m_{CC} / mg	m_{CAM} / mg	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	$I / (0.1C) / \text{mA}$
J1	140	527.9	159.1	1.06	52.14	60	11.79	8.2	10.5	1.720	0.165

Supplementary Table 10. Cell data of group K.

Cell ID	$I_{In}/I_{InLi} / \mu\text{m}$	$I_{SE} / \mu\text{m}$	$I_{CC} / \mu\text{m}$	m_{Li} / mg	m_{In} / mg	m_{SE} / mg	m_{CC} / mg	m_{CAM} / mg	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	$I / (0.1C) / \text{mA}$
K1	100	430	85	0.6	48.9	60.1	12.1	8.5	10.8	2.500	0.1694
K2	100	430	85	0.5	49.8	60.2	12.2	8.5	10.9	2.500	0.1708
K3	100	430	85	0.5	50.8	60.6	12.2	8.5	10.9	2.500	0.1708

Supplementary Table 11. Cell data of group L. For the weight of the In and Li, average values taken from the cells of the other groups standardized to the diameter were used for the calculation of energy and power densities.

Cell ID	$I_{In}/I_{InLi} / \mu\text{m}$	$I_{SE} / \mu\text{m}$	$I_{CC} / \mu\text{m}$	m_{Li} / mg	m_{In} / mg	m_{SE} / mg	m_{CC} / mg	m_{CAM} / mg	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	$I / (0.1C) / \text{mA}$
L1	150	511	180	1.6	48.9	58.9	11.8	8.3	10.0	2.790	0.1569

Supplementary Table 12. Cell data of group M.

Cell ID	$I_{In}/I_{InLi} / \mu\text{m}$	$I_{SE} / \mu\text{m}$	$I_{CC} / \mu\text{m}$	m_{Li} / mg	m_{In} / mg	m_{SE} / mg	m_{CC} / mg	m_{CAM} / mg	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	$I / (0.1C) / \text{mA}$
M1	90	500	50	0.5	20.5	28.8	5.6	3.9	14.5 (10.19)	2.550	0.079
M2	90	510	50	0.5	20.5	28.8	5.6	3.9	14.5 (10.19)	2.540	0.078
M3	100	520	40	0.5	21	29	5.5	3.9	14.3 (10.0)	2.550	0.077

Supplementary Table 13. Cell data of group N. The applied current I is lower than it should be from calculation (0.164 mA at 0.1C).

Cell ID	$I_{In}/I_{InLi} / \mu\text{m}$	$I_{SE} / \mu\text{m}$	$I_{CC} / \mu\text{m}$	m_{Li} / mg	m_{In} / mg	m_{SE} / mg	m_{CC} / mg	m_{CAM} / mg	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	$I / (0.1C) / \text{mA}$
N1	-	420	42.6	4.8	61.2	57.3	11.7	8.2	10.0	2.610	0.118

Supplementary Table 14. Cell data of group O.

Cell ID	$I_{In/InLi} / \mu\text{m}$	$I_{SE} / \mu\text{m}$	$I_{CC} / \mu\text{m}$	m_{Li} / mg	m_{In} / mg	m_{SE} / mg	m_{CC} / mg	m_{CAM} / mg	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	$I / (0.1\text{C}) / \text{mA}$
O1	120	499	112	1.299	39.61	36.12	7.46	5.2	10.4	2.626	0.0522
O2	120	478	132	1.304	40.04	35.79	7.99	5.6	11.1	2.593	0.0559
O3	120	482	109	1.290	39.33	35.99	7.50	5.3	10.5	2.531	0.0525

Supplementary Table 15. Cell data of group P.

Cell ID	$I_{In/InLi} / \mu\text{m}$	$I_{SE} / \mu\text{m}$	$I_{CC} / \mu\text{m}$	m_{Li} / mg	m_{In} / mg	m_{SE} / mg	m_{CC} / mg	m_{CAM} / mg	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	$I / (0.1\text{C}) / \text{mA}$
P1	100	520	160	3.7	40.1	57.5	11.7	8.2	10.4	2.309	0.1637

Supplementary Table 16. Cell data of group Q.

Cell ID	$I_{In/InLi} / \mu\text{m}$	$I_{SE} / \mu\text{m}$	$I_{CC} / \mu\text{m}$	m_{Li} / mg	m_{In} / mg	m_{SE} / mg	m_{CC} / mg	m_{CAM} / mg	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	$I / (0.1\text{C}) / \text{mA}$
Q1	100	543	23	0.7	28.7	29.6	5.8	4.1	10.6	2.720	0.0813
Q2	100	517	14	0.7	30.8	29.2	5.6	3.9	10.2	2.620	0.0826
Q3	100	513	62	0.7	27.9	29.0	5.9	4.1	10.2	2.720	0.0784

Supplementary Table 17. Cell data of group R.

Cell ID	$I_{In/InLi} / \mu\text{m}$	$I_{SE} / \mu\text{m}$	$I_{CC} / \mu\text{m}$	m_{Li} / mg	m_{In} / mg	m_{SE} / mg	m_{CC} / mg	m_{CAM} / mg	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	$I / (0.1\text{C}) / \text{mA}$
R1	-	-	-	2.1	48.0	43	12.1	8.5	10.8	2.580	0.169
R2	-	-	-	2.2	48.4	59	12.2	8.5	10.9	2.510	0.1712
R3	-	-	-	2.2	50.1	59	12.4	8.7	11.1	2.450	0.174

Supplementary Table 18. Cell data of group S.

Cell ID	$I_{In/InLi} / \mu\text{m}$	$I_{SE} / \mu\text{m}$	$I_{CC} / \mu\text{m}$	m_{Li} / mg	m_{In} / mg	m_{SE} / mg	m_{CC} / mg	m_{CAM} / mg	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	$I / (0.1\text{C}) / \text{mA}$
S1	140	200	36	1.71	46.7	60.03	11.58	8.1	10.3	2.504	0.168
S2	140	200	36	1.71	46.7	59.95	12.03	8.4	10.7	2.493	0.168

Supplementary Table 19. Cell data of group T.

Cell ID	$I_{In/InLi} / \mu\text{m}$	$I_{SE} / \mu\text{m}$	$I_{CC} / \mu\text{m}$	m_{Li} / mg	m_{In} / mg	m_{SE} / mg	m_{CC} / mg	m_{CAM} / mg	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	$I / (0.1\text{C}) / \text{mA}$
T1	140	-	-	1.90	100.44	99.81	13.63	9.5	10.04 (7.2)	2.500	0.2
T2	140	-	-	2.06	98.10	100.17	13.60	9.5	10.02 (7.2)	2.520	0.2

Supplementary Table 20. Cell data of group U. For the weight of the In and Li, average values taken from the cells of the other groups standardized to the diameter were used for the calculation of energy and power densities.

Cell ID	$I_{In/InLi} / \mu\text{m}$	$I_{SE} / \mu\text{m}$	$I_{CC} / \mu\text{m}$	m_{Li} / mg	m_{In} / mg	m_{SE} / mg	m_{CC} / mg	m_{CAM} / mg	CAM loading / $\text{mg} \cdot \text{cm}^{-2}$	Initial OCV / V vs. Li^+/Li	$I / (0.1\text{C}) / \text{mA}$
U1	-	-	-	2.1	63.5	100	19	13.3	10.0	2.689	0.266

Supplementary Table 21: In:Li ratios of the In/(InLi)_x alloy negative electrodes. The ratios are calculated from the respective weights of In and Li. Groups L and U are missing, as no weights were reported.

Group	Cell ID	In:Li ratio / wt%:wt%	In:Li ratio / at%:at%
A	A1	20.00	1.21
	A2	20.00	1.21
B	B1	31.33	1.89
	B2	31.33	1.89
	B3	31.33	1.89
D	D1	71.94	4.35
	D2	71.94	4.35
	D3	71.94	4.35
E	E1	57.18	3.46
	E2	57.18	3.46
F	F1	88.00	5.32
	F2	76.80	4.64
G	G1	22.51	1.36
	G2	25.78	1.56
	G3	26.80	1.62
H	H1	109.37	6.61
	H2	109.37	6.61
J	J1	49.33	2.98
K	K1	81.50	4.93
	K2	99.60	6.02
	K3	101.60	6.14
L	L1	-	-
M	M1	41.00	2.48
	M2	41.00	2.48
	M3	42.00	2.54
N	N1	12.75	0.77
O	O1	30.49	1.84
	O2	30.71	1.86
	O3	30.49	1.84
P	P1	10.84	0.66
Q	Q1	41.00	2.48
	Q2	44.00	2.66
	Q3	39.86	2.41
R	R1	22.86	1.38
	R2	22.00	1.33
	R3	22.77	1.38
S	S1	27.31	1.65
	S2	27.31	1.65
T	T1	52.86	3.20
	T2	47.62	2.88
U	U1	-	-

Supplementary Table 22. Parameters obtained from the DRT analysis. The parameters are area-normalized.

Cell	1 st charge (0.05 C)				2 nd charge (0.05 C)				51 st charge (0.1 C)				λ
	$R_{\text{tot}} / \Omega \cdot \text{cm}^2$	$R_0 / \Omega \cdot \text{cm}^2$	C_0 / F	L_0 / H	$R_{\text{tot}} / \Omega \cdot \text{cm}^2$	$R_0 / \Omega \cdot \text{cm}^2$	C_0 / F	L_0 / H	$R_{\text{tot}} / \Omega \cdot \text{cm}^2$	$R_0 / \Omega \cdot \text{cm}^2$	C_0 / F	L_0 / H	
D1	263.8	50.9	7.56E-01	1.12E-07	293.6	54.7	6.58E-01	0	265.3	61	9.22E-01	0	0.2
D2	240	54.4	0	0	295.5	54.5	0	0	290.8	58.6	2.73E-01	0	0.2
E1	533.7	29.8	1.12E-01	0	651.7	31.1	7.87E-02	0	942.5	35	7.21E-02	0	0.1
E2	330.7	23.4	1.84E-01	0	416.5	23.9	1.97E-01	0	638.1	31	1.38E-01	0	0.1
G1	140.6	24.3	1.80E-01	0	149	25	1.97E-01	0	569.4	29.1	8.31E-02	0	0.1
G2	124.9	24.6	1.61E-01	0	154.1	24.7	1.73E-01	0	274.9	33.9	1.45E-01	0	0.1
G3	186.6	24.4	2.22E-01	0	133.1	25.4	1.63E-01	0	3887.7	31.6	1.10E-03	0	0.1
H1	129.8	48.5	7.59E-01	0	130.5	49.1	1.1	0	114.4	59.2	1.6	0	0.3
H2	149.7	49.4	7.98E-01	0	156.8	50.6	1	0	149.7	62.9	1.2	0	0.3
K1	114.1	27.8	7.43E-01	0	140.7	27.9	9.33E-01	0	181.6	34.3	3.36E-01	0	0.1
K2	114	27.8	7.69E-01	0	137	29.3	0	0	182.6	34.4	4.55E-01	0	0.1
K3	164.5	31.6	6.34E-01	0	193.5	31.6	6.78E-01	0	207.4	39.4	2.85E-01	0	0.1
L1	83.1	33.5	1	9.75E-07	82.7	33.4	1.5	9.57E-07	78.9	35.5	8.62E-01	2.50E-06	0.2
M1	186.6	39.8	5.87E-01	0	224.7	45.2	5.05E-01	0	210.4	59.5	7.52E-01	0	0.2
M2	260.8	43.4	3.85E-01	0	295.7	44.6	4.00E-01	0	500.3	51.7	3.62E-01	0	0.2
M3	226.9	41.9	3.42E-01	0	253.9	41.1	3.95E-01	0	216	59.2	3.78E-01	0	0.2
N1	139.5	39.5	9.67E-01	0	165.4	46.6	5.15E-01	0	227.6	55	8.27E-01	0	0.2
O1	175.5	22.5	4.49E-02	0	202.5	22.4	4.97E-01	0	277.5	22.8	4.24E-01	0	0.1
O2	157.9	21.2	7.17E-01	0	168.9	21.3	4.63E-01	0	215.7	20.6	4.42E-01	0	0.1
O3	169.5	17.9	4.28E-01	0	172	17.1	3.65E-01	0	233.5	16.3	2.66E-01	0	0.1
P1	536	49.9	4.26E-02	7.70E-06	485.5	51.2	2.99E-02	7.50E-06	1649.3	51.4	6.50E-03	7.70E-06	0.1
Q1	189	19.4	2	0	222.7	19.4	1.4	0	278.1	18.7	3.52E-01	0	0.1
Q2	193.3	21.7	9.81E-01	0	234.4	23.2	1.7	0	282.7	22.9	2.46E-01	0	0.1
R1	198.5	27.4	4.15E-01	4.46E-07	227.4	29.5	468.3	3.46E-07	240.1	36	2.33E-01	0	0.1
R2	213.4	37	3.35E-01	6.61E-08	243.9	37.9	227.2	0	233.7	45.3	2.69E-01	0	0.1
R3	217.9	37.5	2.49E-01	0	272.5	37.2	178.4	0	262.3	43.6	3.75E-01	0	0.1
S1	169.1	32.6	5.43E-01	0	174.4	32.5	6.80E-01	0	176.5	37.5	5.57E-01	0	0.1
S2	151	31.4	7.02E-01	0	159.6	31.5	6.22E-01	0	169.6	35.3	7.51E-01	0	0.1
T1	675.7	42.2	1.36E-01	0	789.5	40.2	1.30E-01	0	1294.3	36.6	1.63E-01	0	0.3
T2	565.7	32.3	1.69E-01	0	675.5	32.7	1.37E-01	0	-	-	-	-	0.3
U1 [†]	149.3	39	2.5	0	235.7	46.2	2.9	0	286.4	45.5	1.9	0	0.3

[†] Impedance measurement down to 5 mHz, which influences the values of C_0 and R_{tot}

Supplementary Table 23. Statistical results from the violin plots. The corresponding violin plots are depicted in Figure 2 in the main manuscript.

	OCV / V	Polarization voltage / V		Capacity retention / %	Specific charge capacity (0.05C) / mAh·g ⁻¹	Specific discharge capacity / mAh·g ⁻¹			Coulomb efficiency / %		
		Cycle 1	Cycle 50			Pre-treatment (0.05C)	Cycle 1 (0.1C)	Cycle 50 (0.1C)	Pre-treatment (0.05C)	Cycle 1 (0.1C)	Cycle 50 (0.1C)
Box limit (low)	2.47	0.15	0.20	76.1	127.0	95.3	75.8	44.2	75.4	90.0	99.3
Box limit (high)	2.63	0.24	0.30	97.6	188.5	144.5	132.1	125.3	78.1	95.0	100.1
Whisker limit (low)	2.29	0.13	0.13	49.5	71.2	46.6	20.7	12.7	73.8	86.1	99.0
Whisker limit (high)	2.79	0.33	0.43	107.0	194.6	152.9	143.1	145.9	80.6	100.0	100.7
Median	2.54	0.20	0.23	91.6	179.3	137.2	121.9	105.6	77.0	93.4	99.8
Average	2.49	0.25	0.29	81.1	160.6	122.6	103.7	86.0	75.7	90.8	98.6

Supplementary Table 24. Elemental composition of the cathode active material. The composition was determined by 12 EDX point measurements.

Element	Fraction / atomic%
Ni	60.3 ± 0.5
Mn	20.0 ± 0.4
Co	19.7 ± 0.3

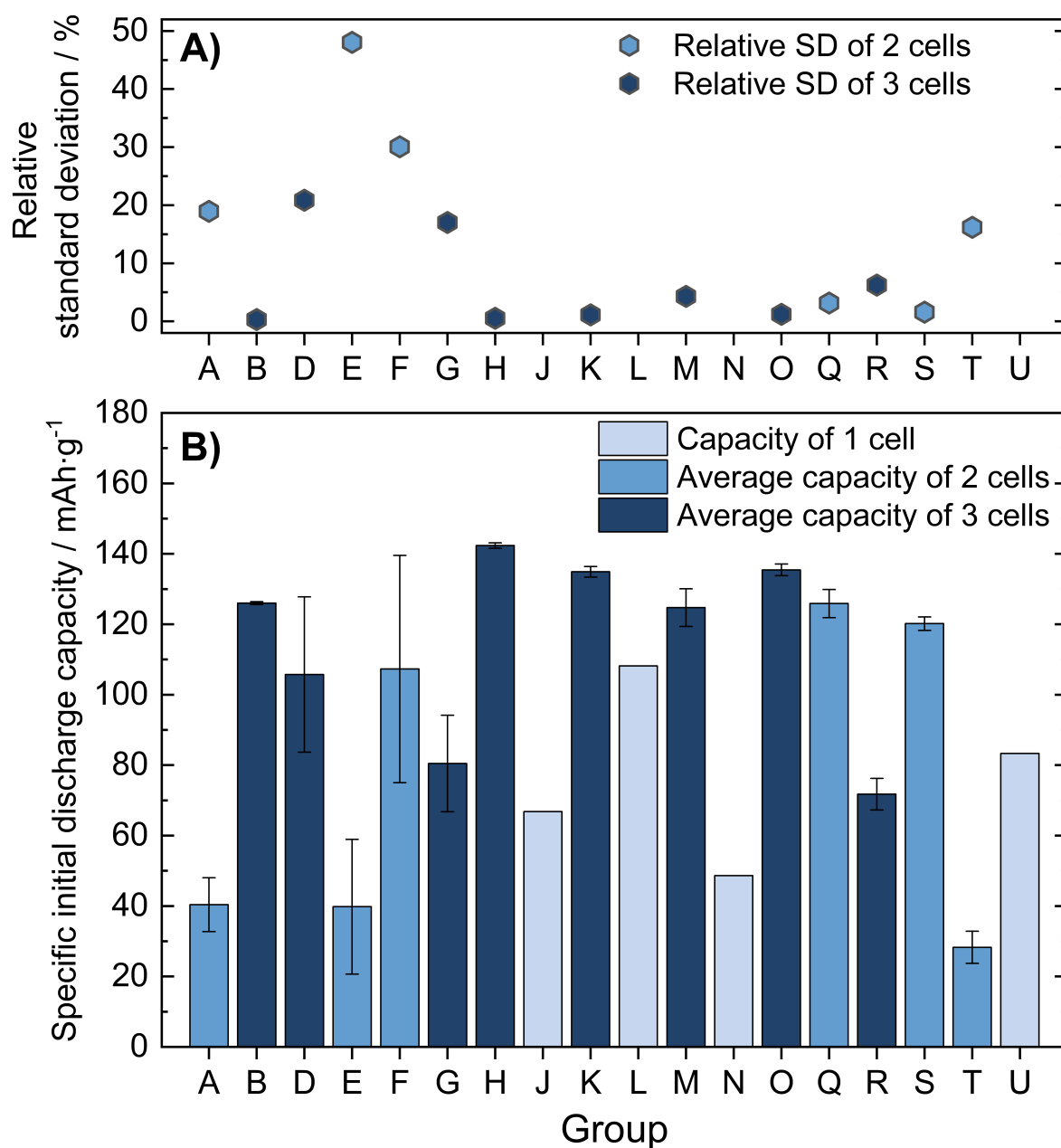
Supplementary Table 25. Temperature-dependent conductivity of Li₆PS₅Cl. The conductivity was determined by two independent PEIS measurements. The ionic conductivity σ_{ion} of the Li₆PS₅Cl was calculated by $\sigma_{\text{ion}} = \frac{l}{R \cdot A}$ with l being the thickness of the pellet, A the area of the electrodes and R the resistance, determined from the fitting of the PEIS data with a R-CPE equivalent circuit.

Temperature / °C	$\sigma_{\text{ion}} / \text{mS} \cdot \text{cm}^{-1}$	
	Measurement 1	Measurement 2
10	0.83	0.89
13	0.89	1.01
16	1.06	1.19
20	1.28	1.46
25	1.63	1.85
30	2.14	2.45
40	3.22	3.72
50	4.95	5.81
60	7.35	8.72

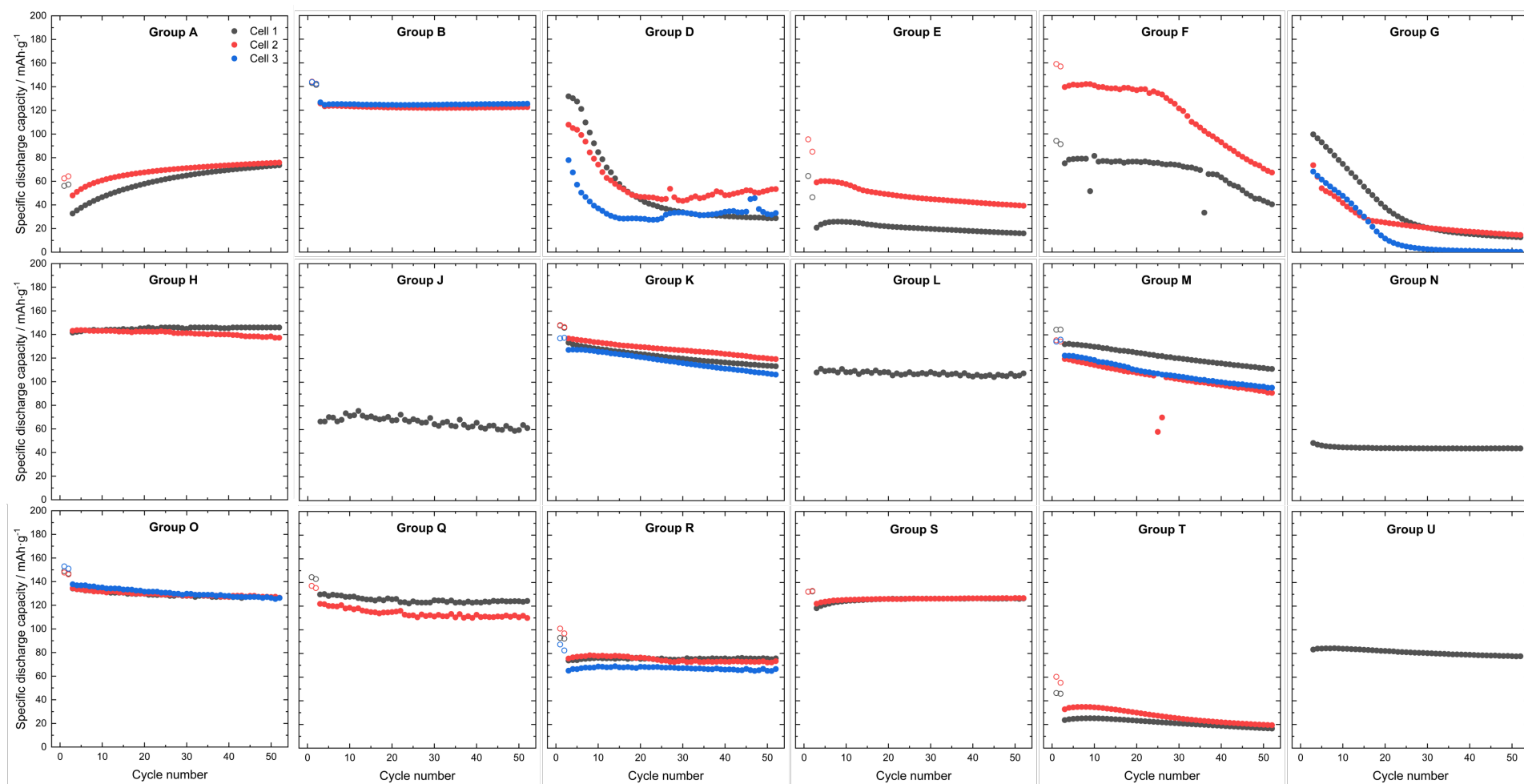
Supplementary Table 26. PEIS measurement parameters. The activation energy was determined from the slope of the temperature-dependent PEIS measurement in Supporting Figure 47 **Error! Reference source not found..**

	Measurement 1	Measurement 2
Weight / mg	249.5	250.7
Thickness / mm	2.02	2.19
E_a / eV	0.3884	0.3996

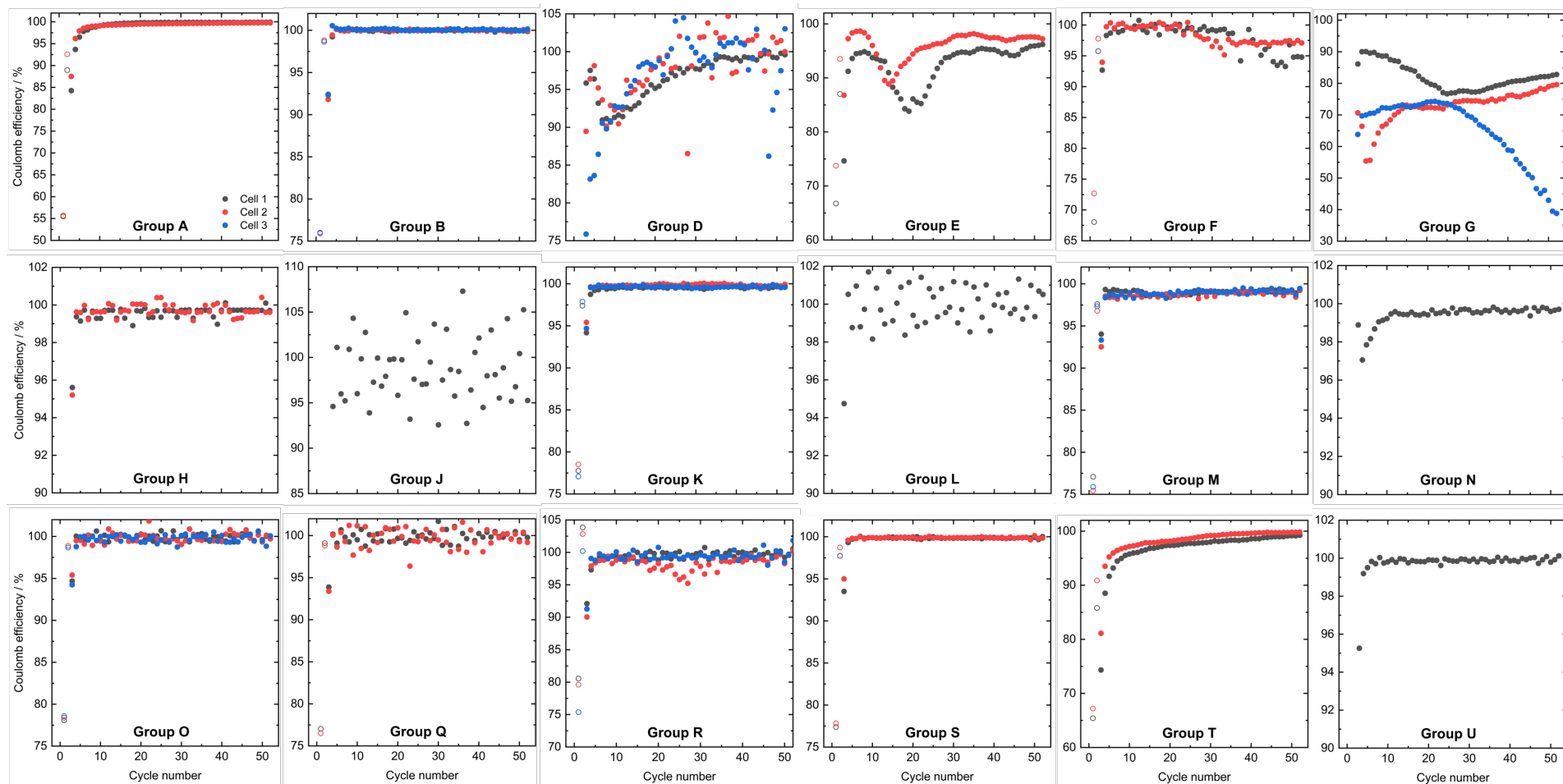
2. Supplementary Figures



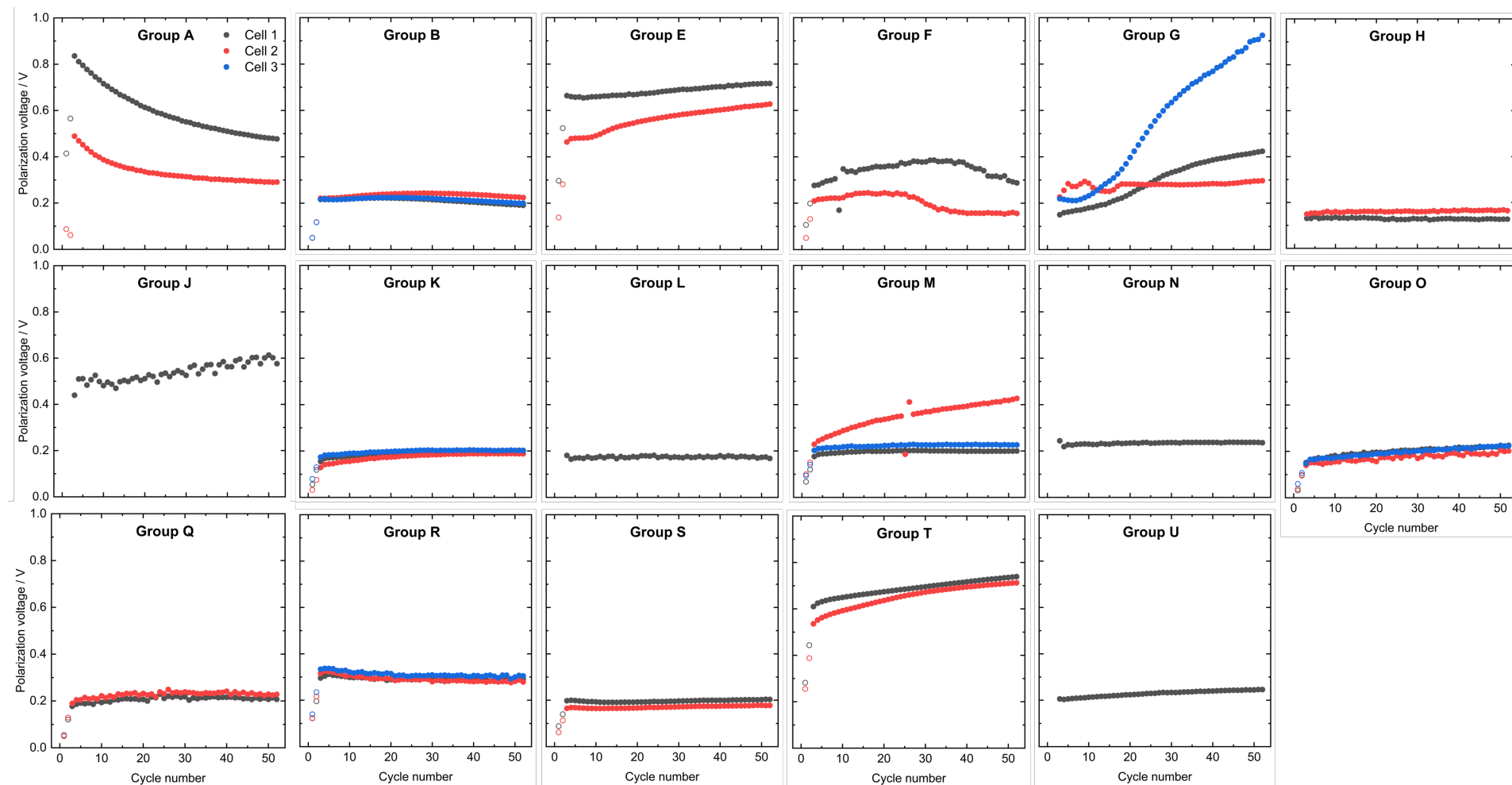
Supplementary Figure 1. Average initial discharge capacity per group. A) shows the relative standard deviation, the color marks if two or three cells were considered. B) shows the (average) capacity, the number of cells averaged per group is marked by the color. For the groups with two or three cells working, the standard deviation is shown as error bars.



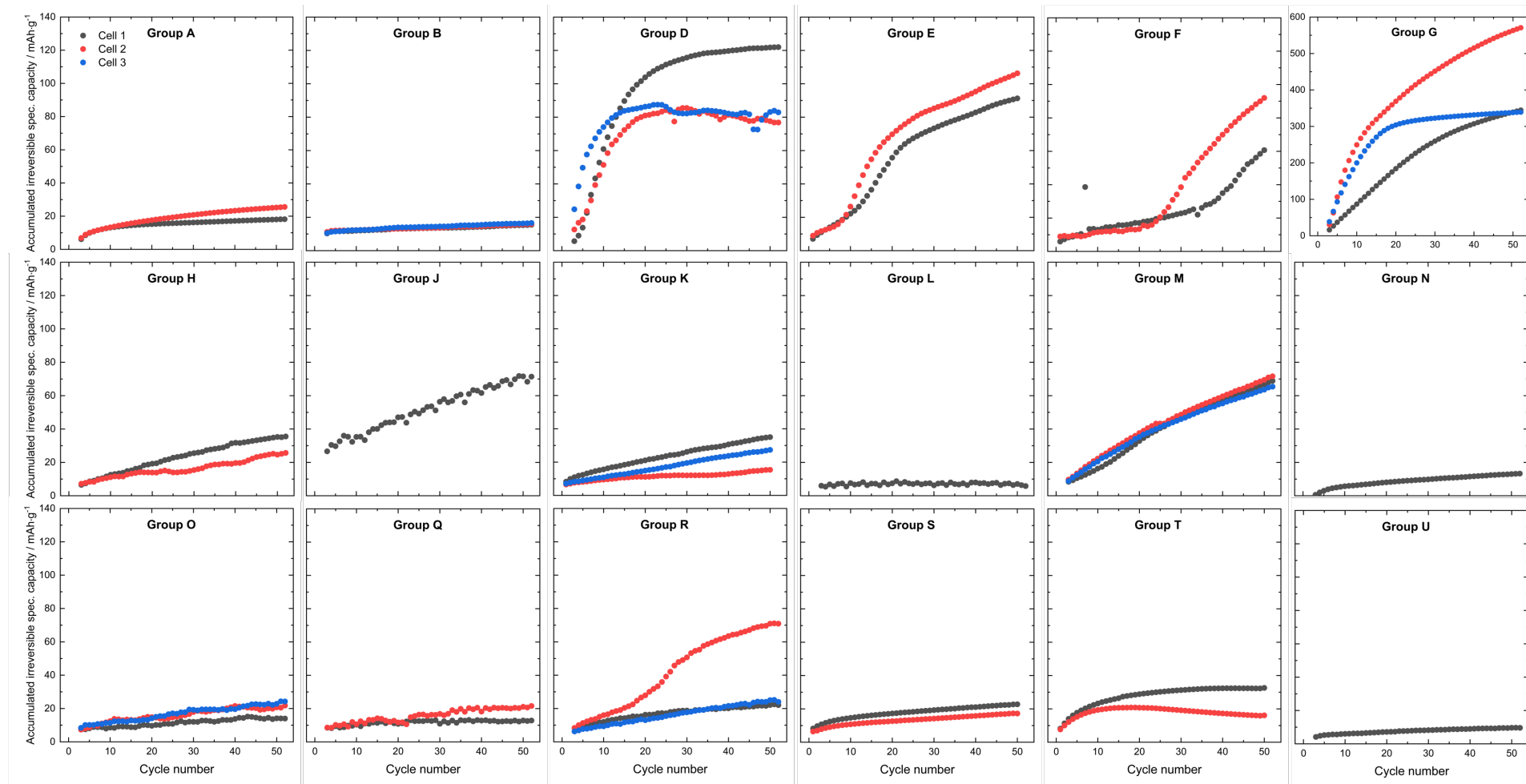
Supplementary Figure 2. Specific discharge capacities of all working cells. Open circles mark the 0.05C pretreatment data, if it was provided by the respective group.



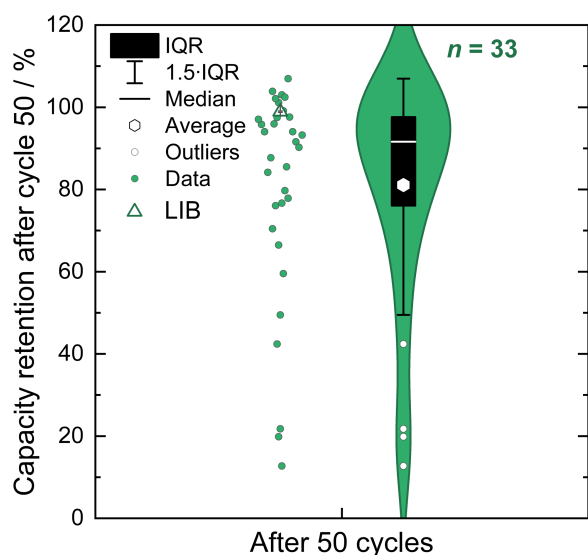
Supplementary Figure 3. Coulomb efficiencies of all working cells. Open circles mark the 0.05C pretreatment data, if it was provided by the respective group.



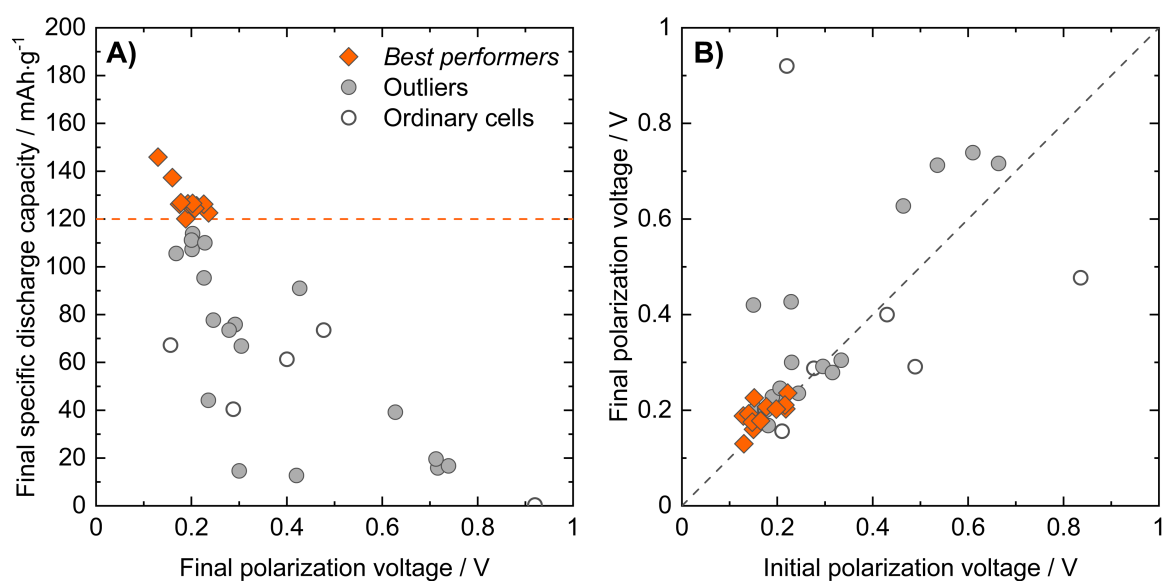
Supplementary Figure 4. Polarization voltages of all working cells. Open circles mark the 0.05C pretreatment data, if it was provided by the respective group. Group D is not shown, because the voltage data was not reported.



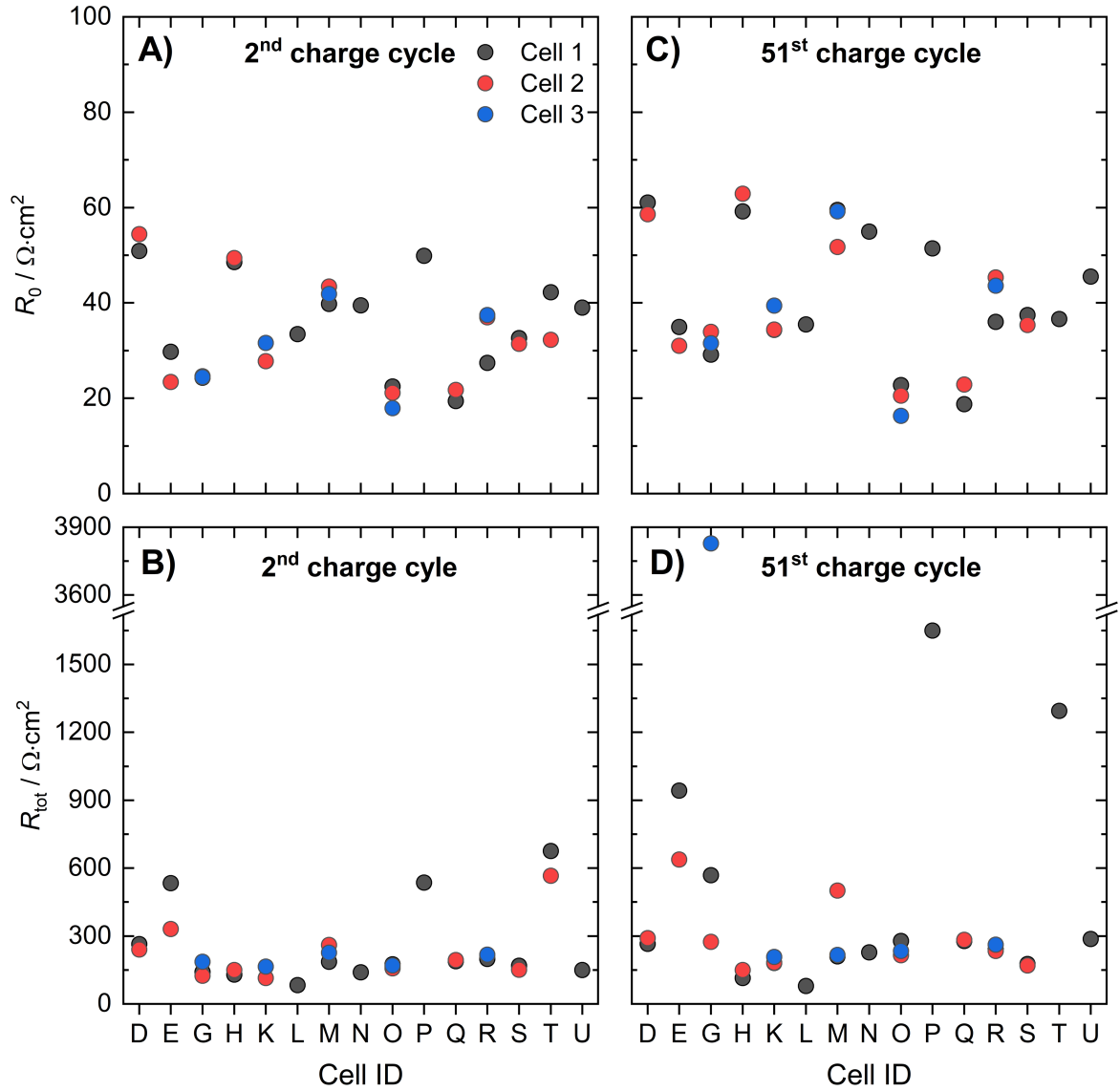
Supplementary Figure 5. Accumulated irreversible capacities of all working cells. Only the 0.1C cycling is considered.



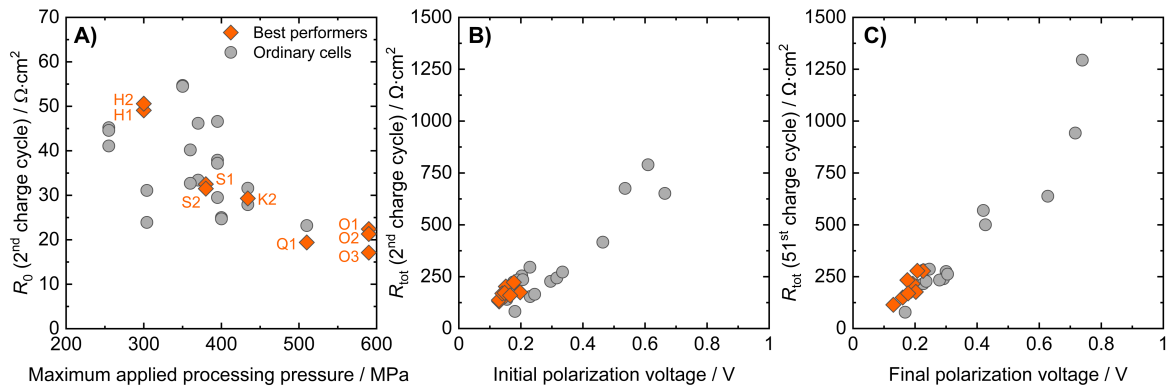
Supplementary Figure 6. Violin plot of the capacity retentions. The capacity retention is calculated from the quotient of the 50th and 1st 0.1C discharge capacity. The box shows the interquartile range (IQR), the whisker 1.5·IQR of the 33 ASSB cells considered. The violin plot was prepared with Kernel density estimation. The triangle shows the Li-ion battery (LIB) coin cell data of three coin cells we prepared as benchmark of the NMC 622 active material, the error bar shows the standard deviation of these three cells.



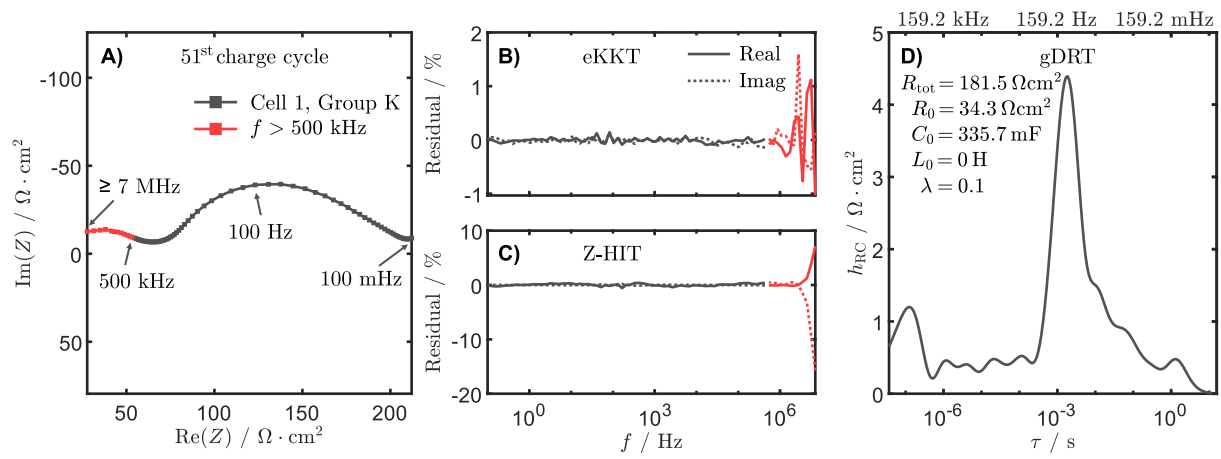
Supplementary Figure 7. Definition of best performers and polarization evolution. A) Final specific discharge capacity as a function of the final polarization voltage, the dotted line shows the defined limit at 120 mAh·g⁻¹ for the best performers. B) Evolution of the polarization voltage from the 1st to the 50th cycle.



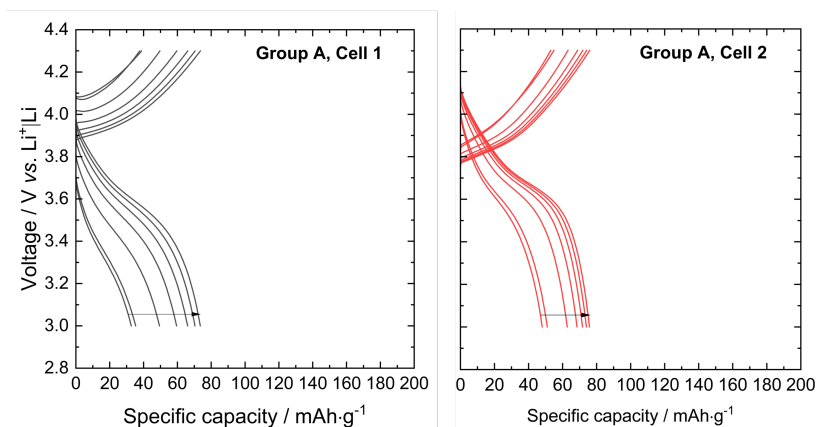
Supplementary Figure 8. Resistances obtained from DRT analysis. High-frequency intercept R_0 and total cell resistance R_{tot} after A), B) the second and C), D) the 51st charge cycle.



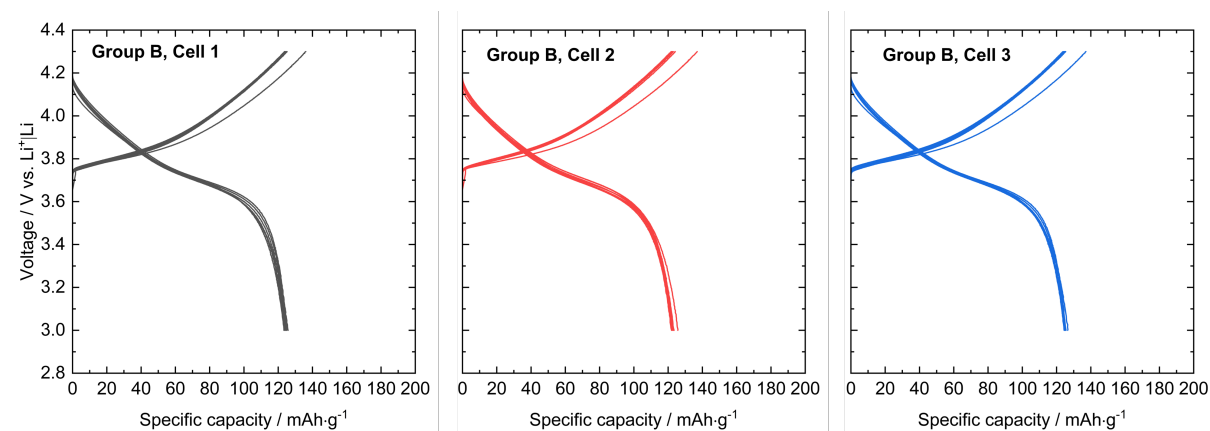
Supplementary Figure 9. Correlation of resistance and cycling. A) High-frequency intercept R_0 as a function of the maximum pressure used during the whole compression procedure, B) initial total cell resistance R_{tot} vs. the initial polarization voltage and C) final R_{tot} as a function of the final polarization voltage. The resistances are obtained from DRT analysis.



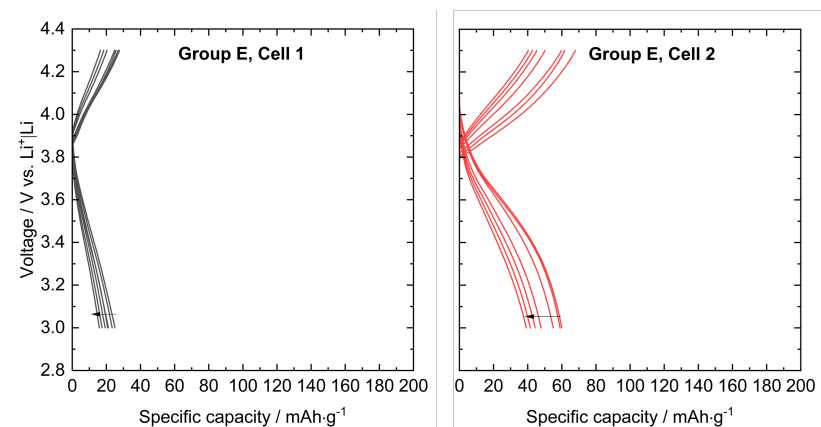
Supplementary Figure 10. DRT analysis of the PEIS spectrum of group K. A) Displays the impedance spectrum from the 51st charging cycle of cell 1 in group K, highlighting the high frequency components removed at $f \geq 500$ kHz in red. B) and C) present the residuals from the extended Kramers-Kronig Test (eKKT) and the Z-HIT impedance validation methods, respectively. D) Illustrates the generalized distribution function (gDRT) incorporating additional lumped serial elements, overall polarization, and the regularization parameter used.



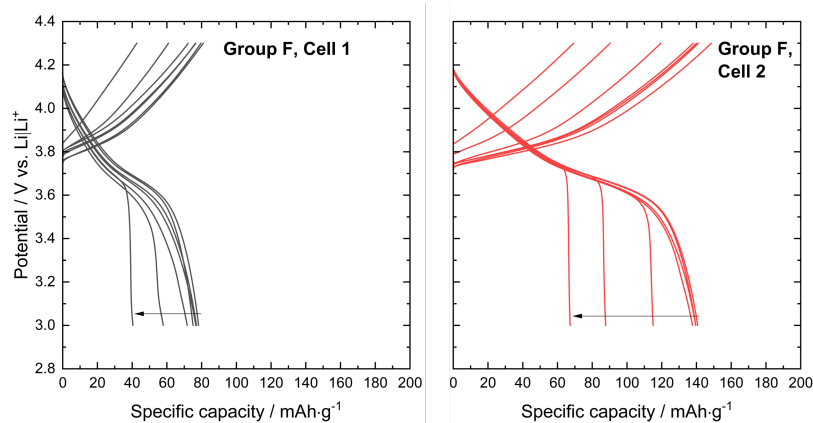
Supplementary Figure 11. Charge-/discharge curves for group A.



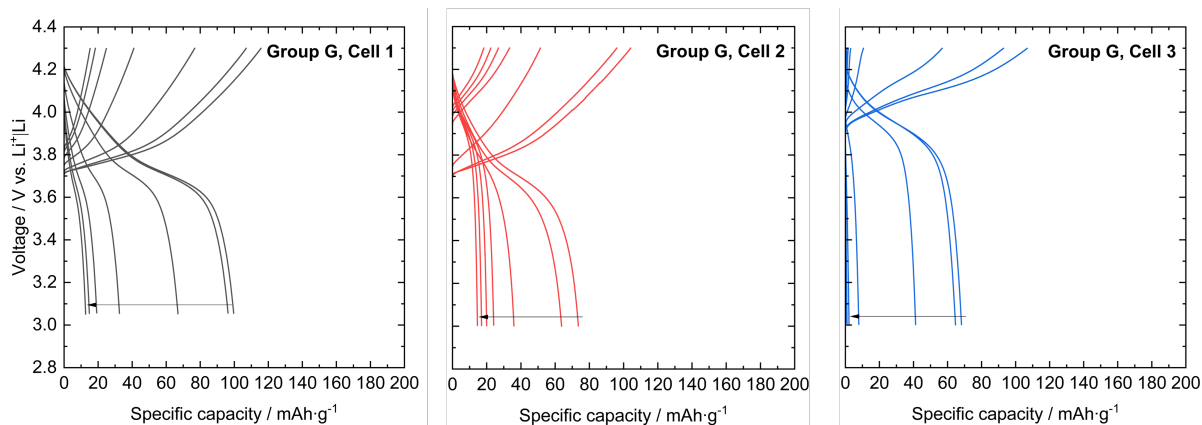
Supplementary Figure 12. Charge-/discharge curves for group B.



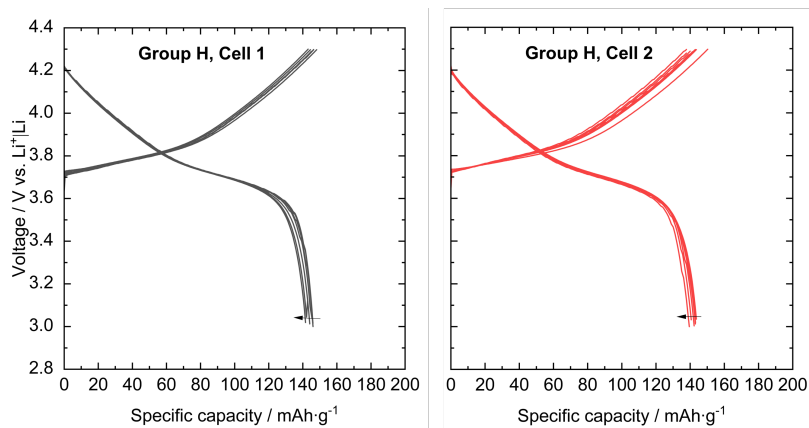
Supplementary Figure 13. Charge-/discharge curves for group E.



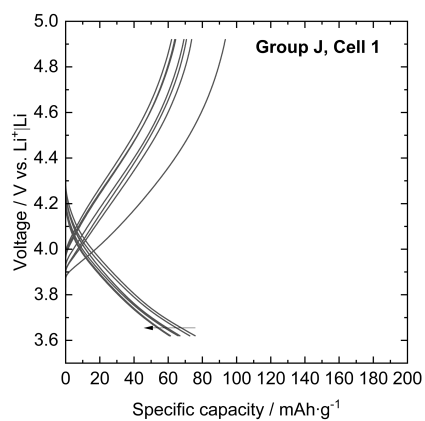
Supplementary Figure 14. Charge-/discharge curves for group F.



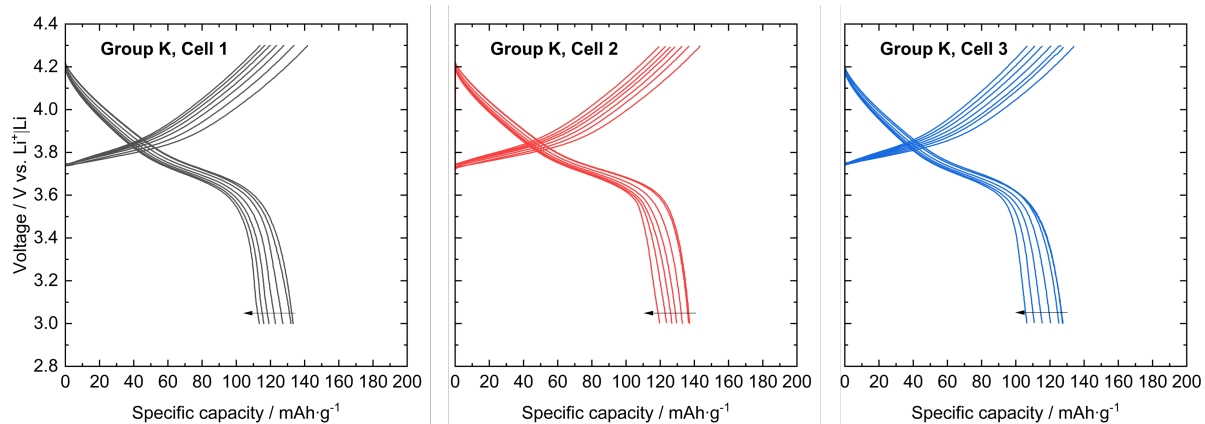
Supplementary Figure 15. Charge-/discharge curves for group G.



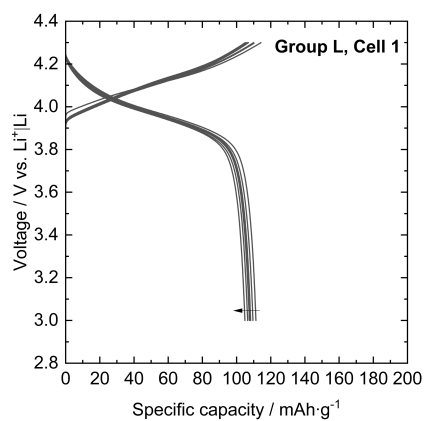
Supplementary Figure 16. Charge-/discharge curves for group H.



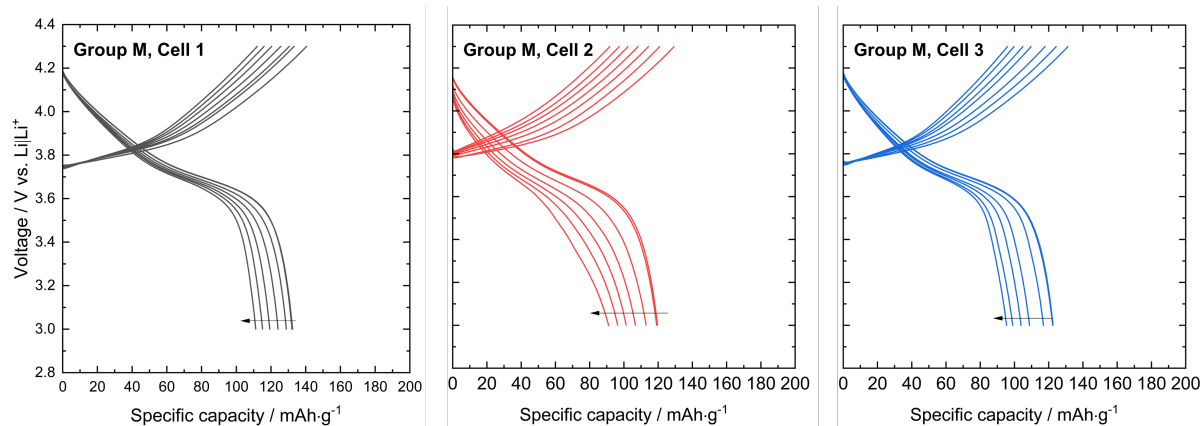
Supplementary Figure 17. Charge-/discharge curves for group J.



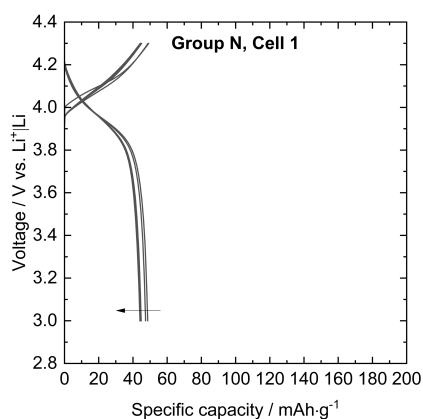
Supplementary Figure 18. Charge-/discharge curves for group K.



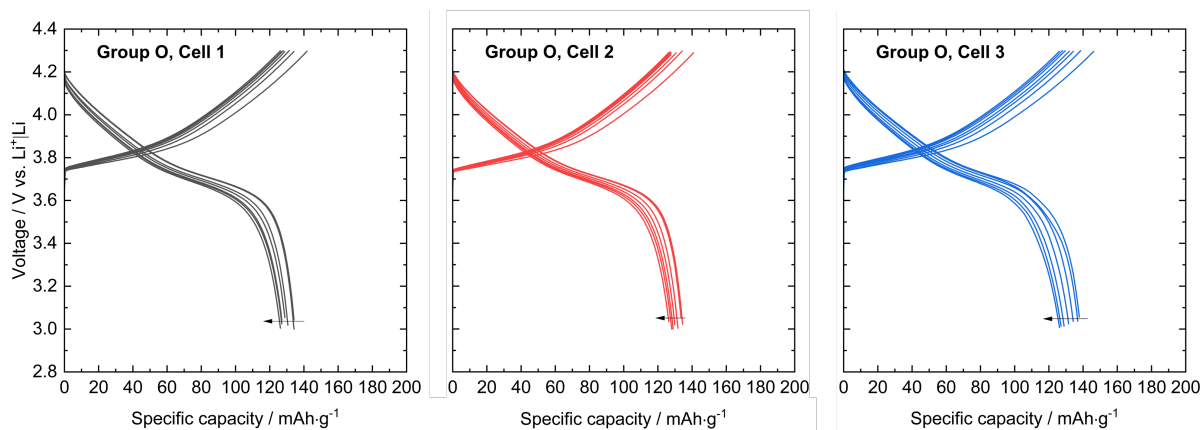
Supplementary Figure 19. Charge-/discharge curves for group L.



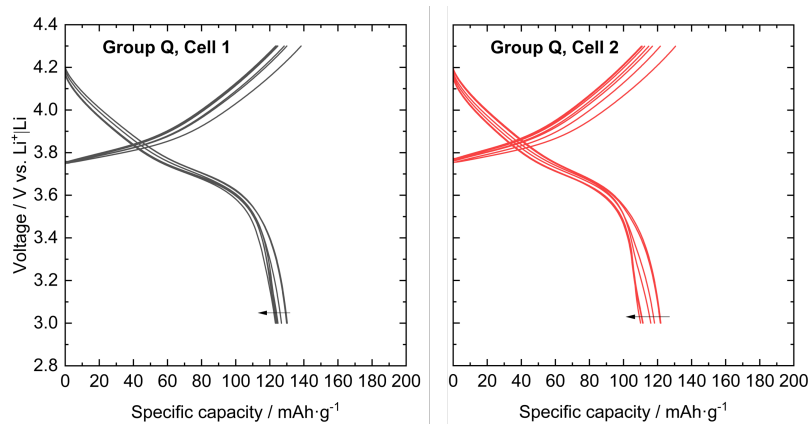
Supplementary Figure 20. Charge-/discharge curves for group M.



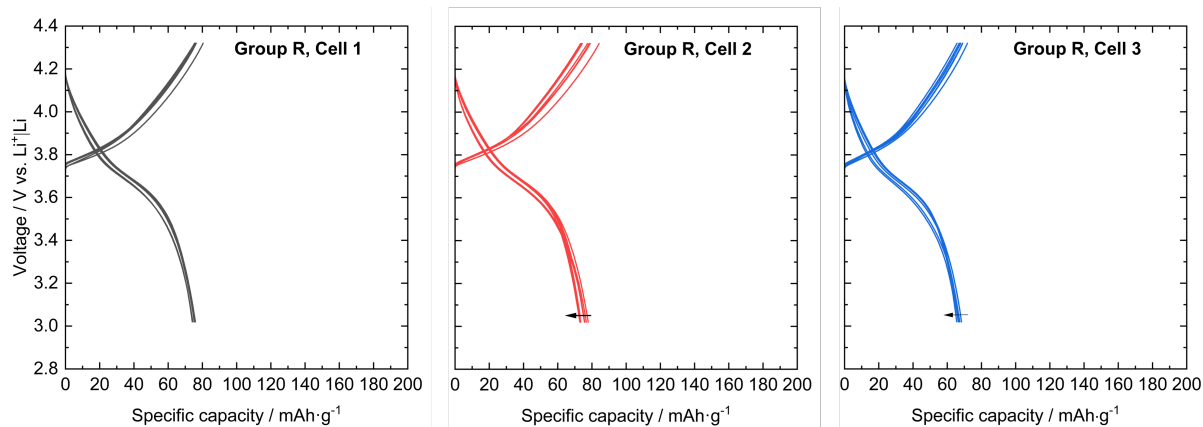
Supplementary Figure 21. Charge-/discharge curves for group N.



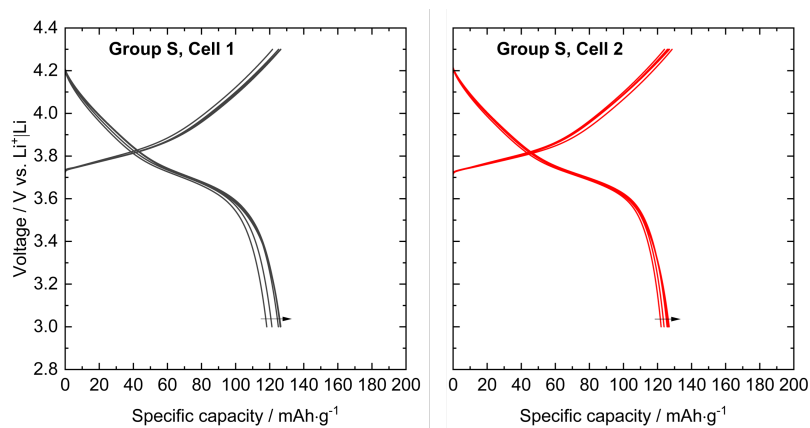
Supplementary Figure 22. Charge-/discharge curves for group O.



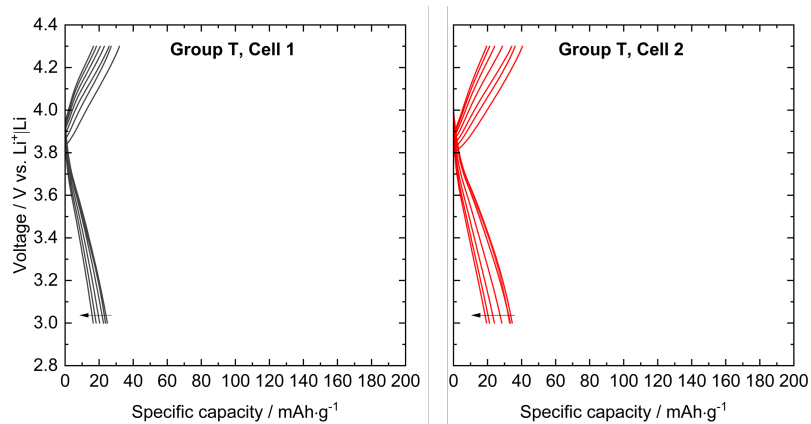
Supplementary Figure 23. Charge-/discharge curves for group Q.



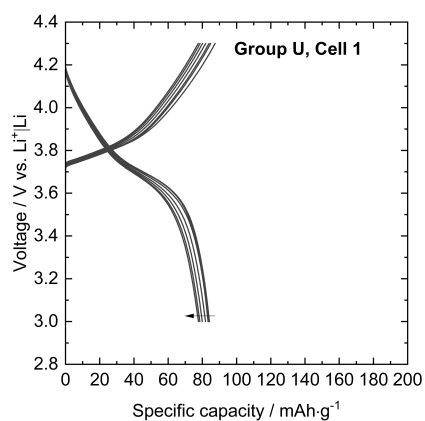
Supplementary Figure 24. Charge-/discharge curves for group R.



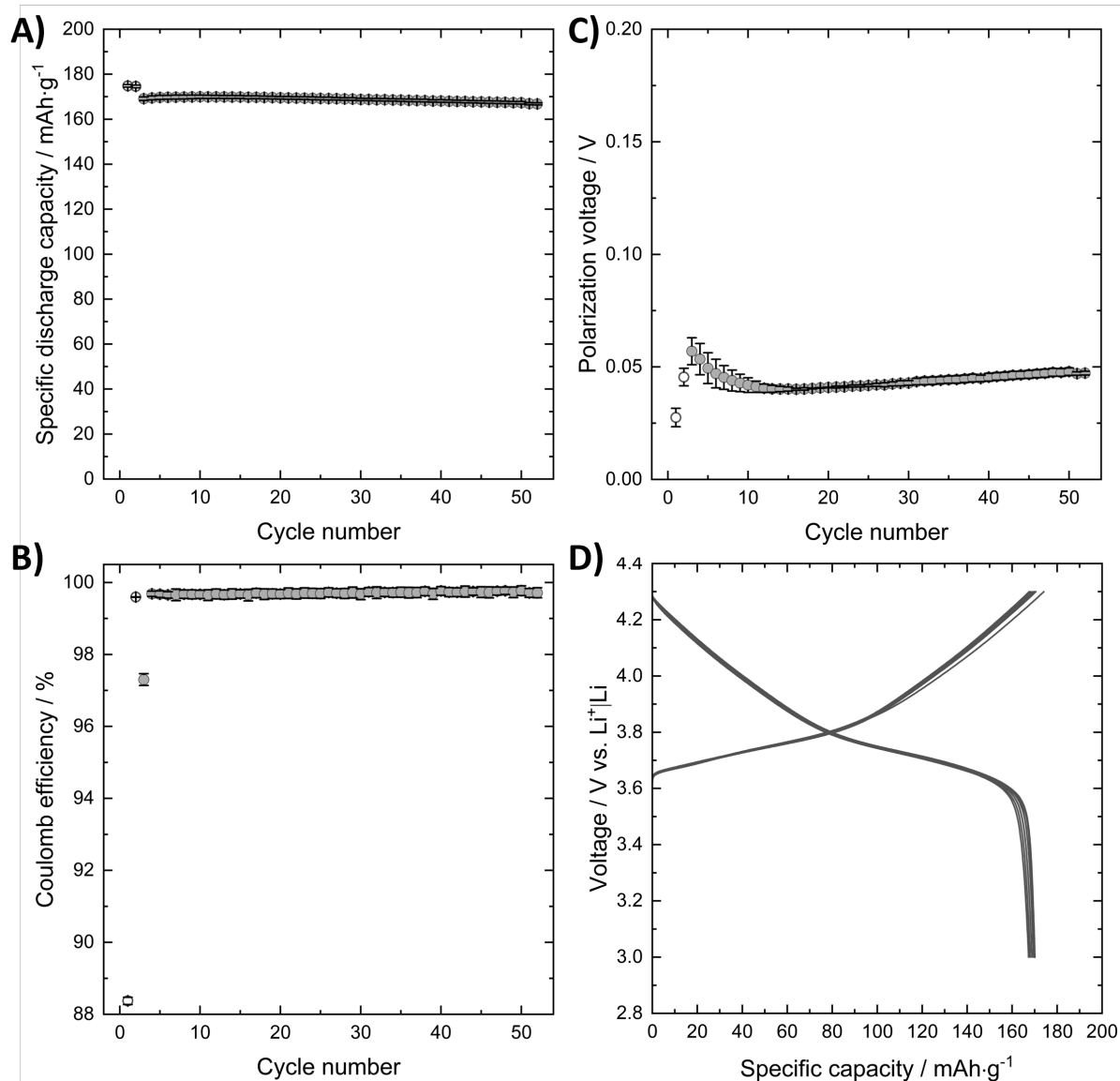
Supplementary Figure 25. Charge-/discharge curves for group S.



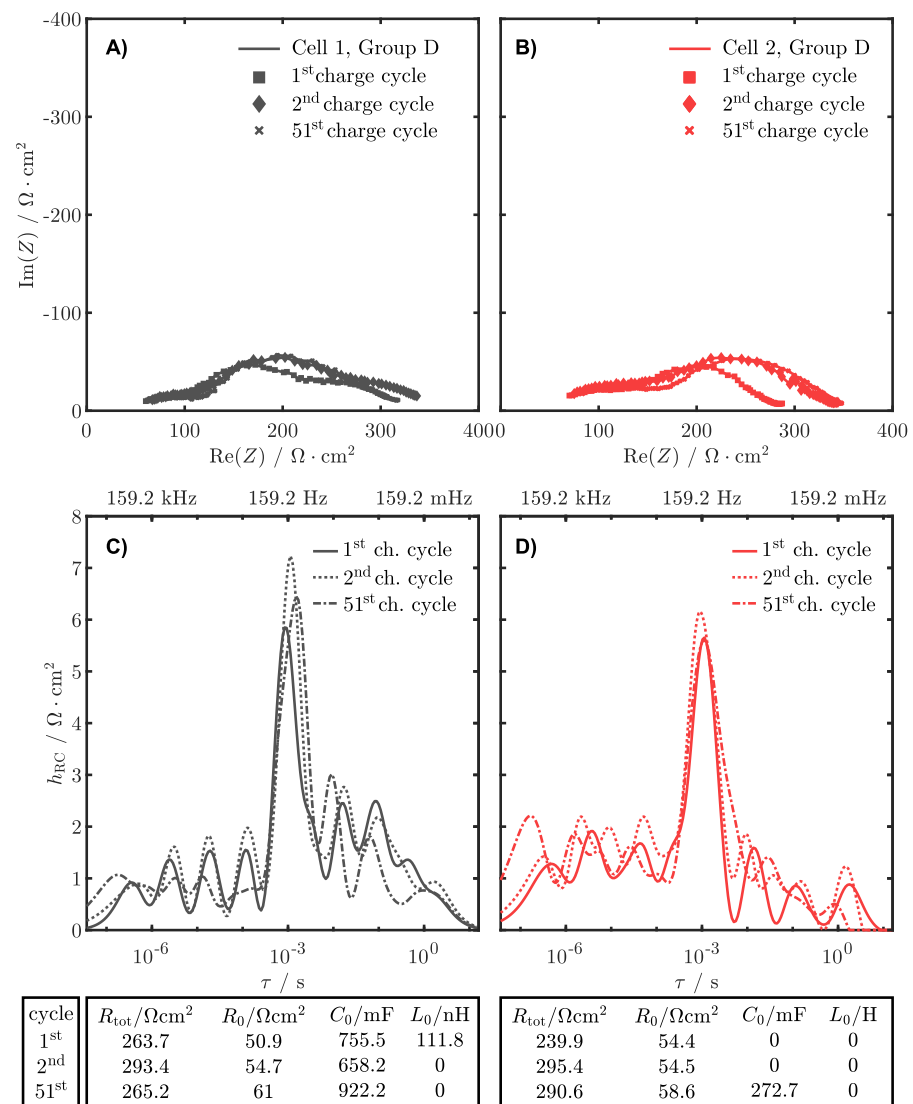
Supplementary Figure 26. Charge-/discharge curves for group T.



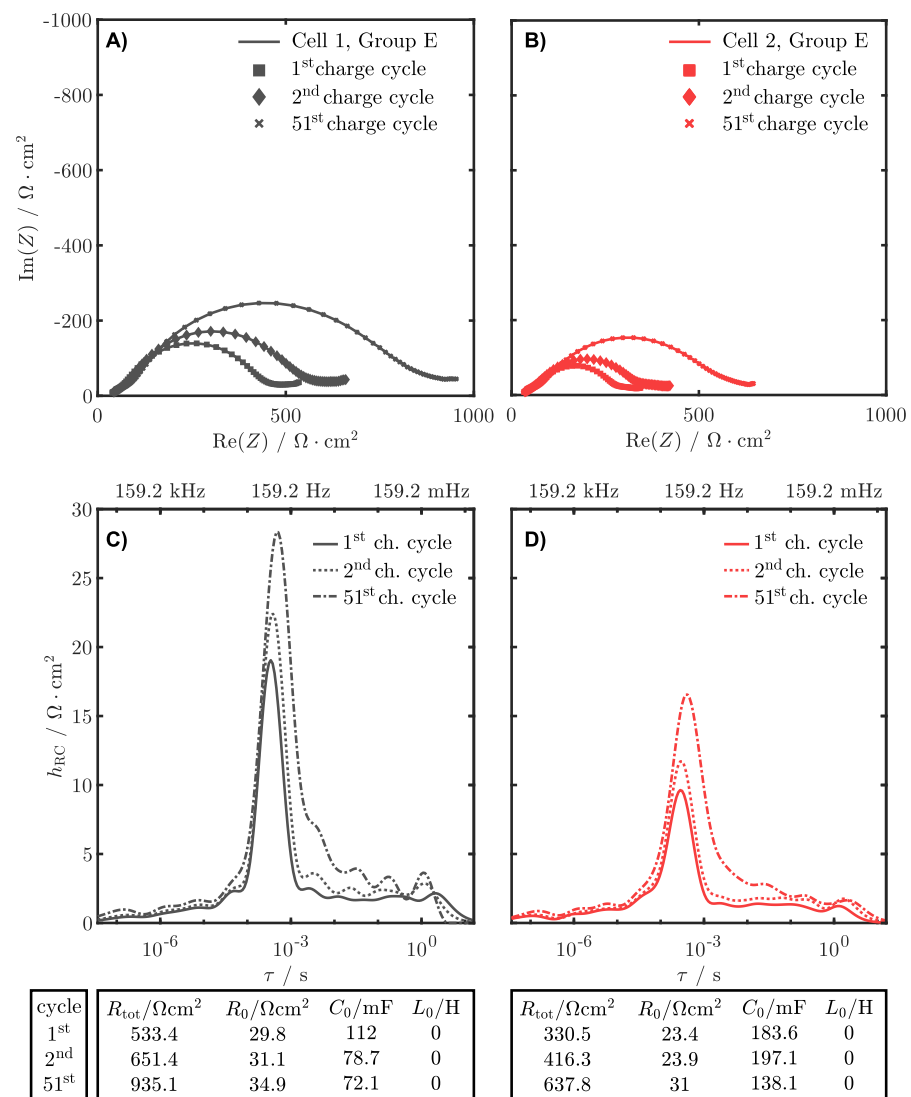
Supplementary Figure 27. Charge-/discharge curves for group U.



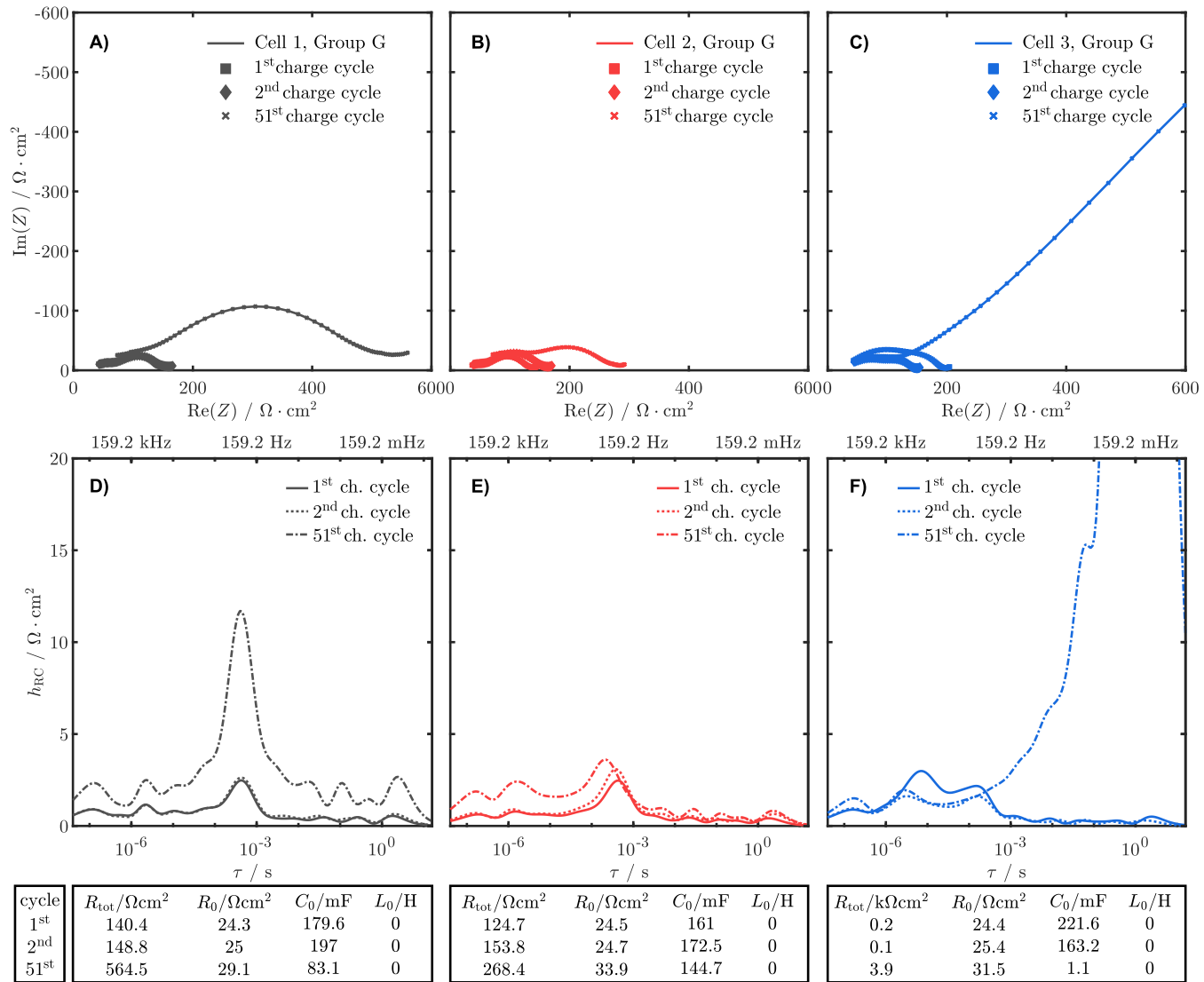
Supplementary Figure 28. Coin cell cycling results. A) Specific discharge capacity; B) Coulomb efficiency and C) polarization voltage of the reference coin cells. The average and standard deviation of three cells is shown, open circles mark the first two pretreatment cycles at 0.05C. D) Charge/ discharge curves of one representative coin cell, shown are the cycles 1, 2, 10, 20, 30, 40 and 50.



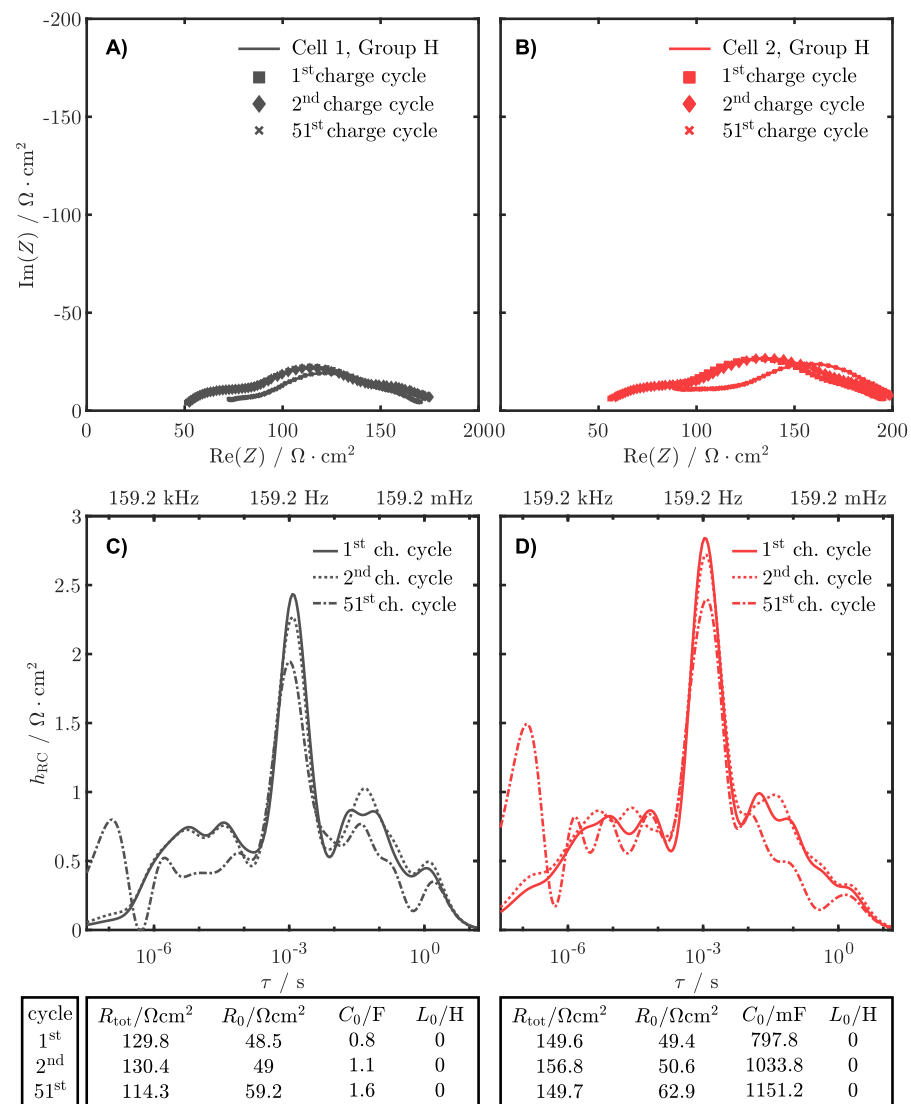
Supplementary Figure 29. Impedance spectra and DRT analysis of group D. A) and B) show the impedance spectra after specific charging steps and C), D) DRT analysis with the attached table of lumped serial elements for group D.



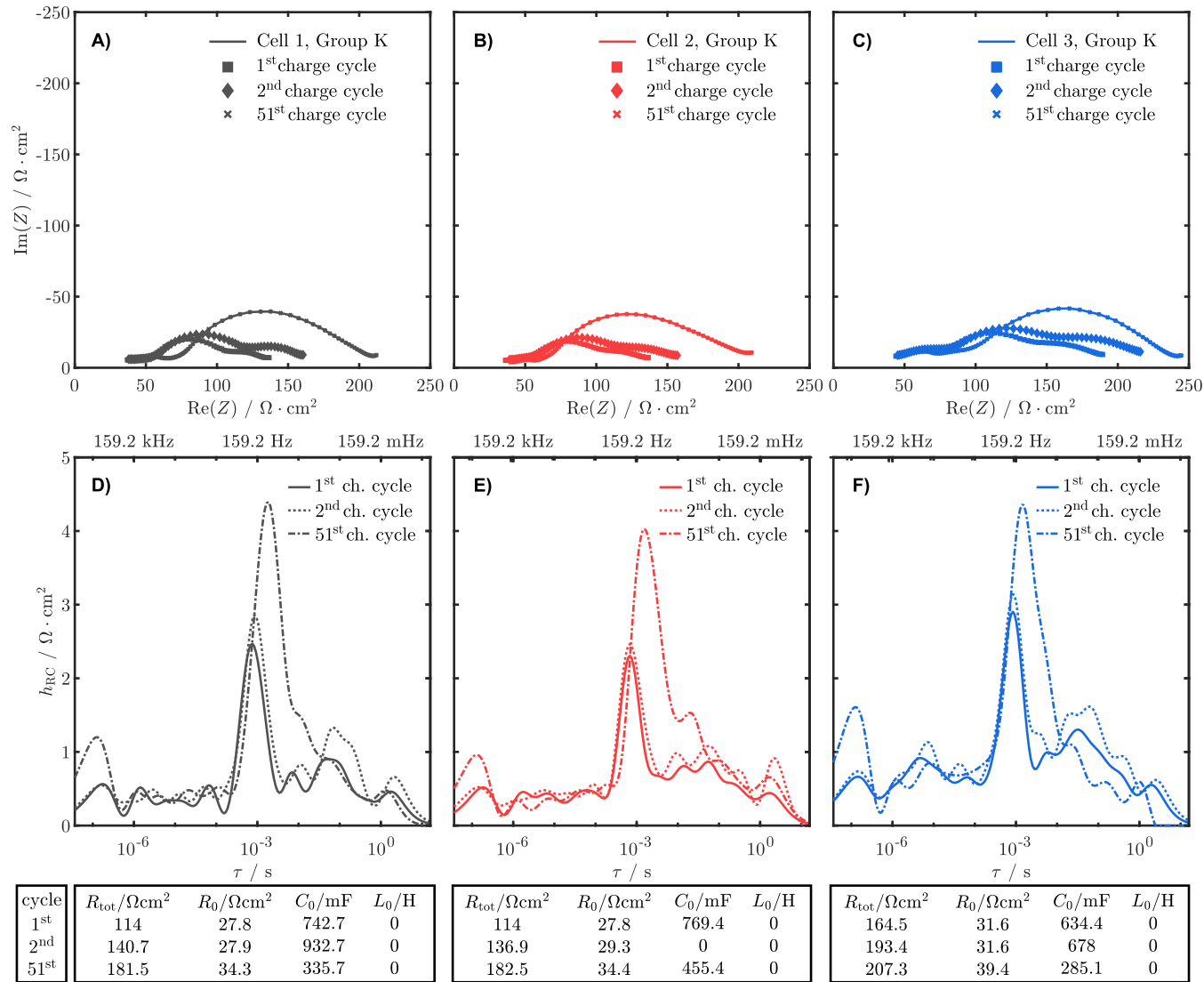
Supplementary Figure 30. Impedance spectra and DRT analysis of group E. A), B) Impedance spectra after specific charging steps and C), D) DRT analysis with the attached table of lumped serial elements for group E.



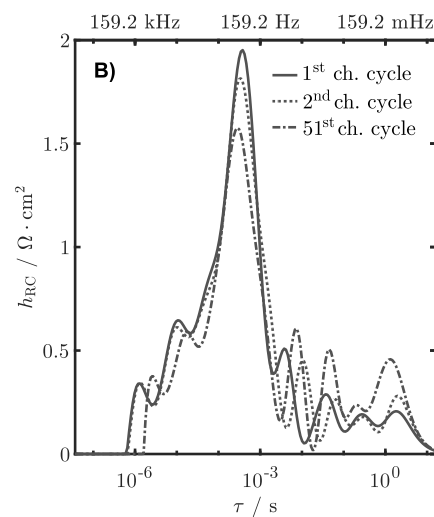
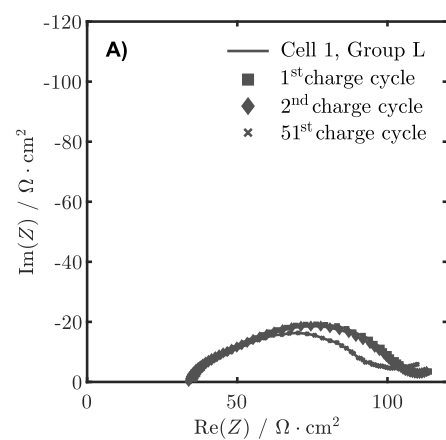
Supplementary Figure 31. Impedance spectra and DRT analysis of group G. A) to C) Impedance spectra after specific charging steps and D) to F) DRT analysis with the attached table of lumped serial elements for group G.



Supplementary Figure 32. Impedance spectra and DRT analysis of group H. A), B) Impedance spectra after specific charging steps and C), D) DRT analysis with the attached table of lumped serial elements for group H.

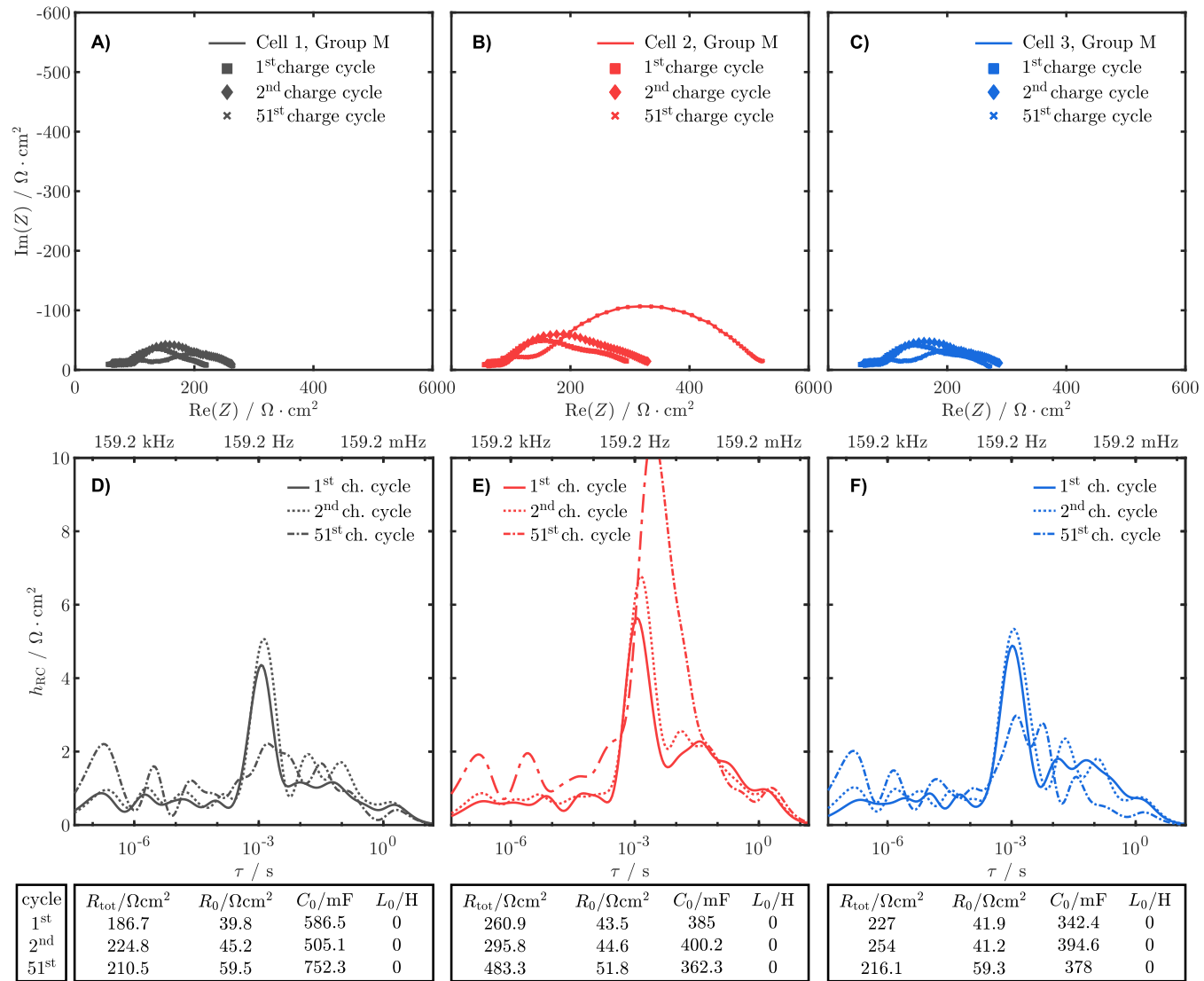


Supplementary Figure 33. Impedance spectra and DRT analysis of group K. A) to C) Impedance spectra after specific charging steps and D) to F) DRT analysis with the attached table of lumped serial elements for group K.

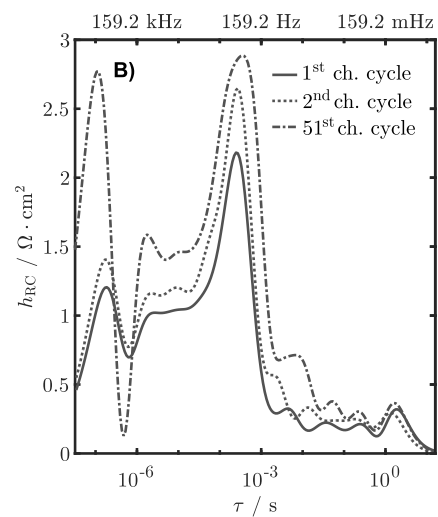
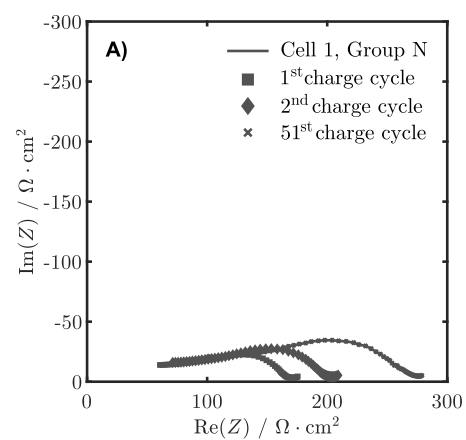


cycle	$R_{\text{tot}} / \Omega \text{cm}^2$	$R_0 / \Omega \text{cm}^2$	C_0 / F	$L_0 / \mu\text{H}$
1 st	83	33.4	1	1
2 nd	82.7	33.3	1.5	1
51 st	78.8	35.5	0.9	2.5

Supplementary Figure 34. Impedance spectra and DRT analysis of group L. A) Impedance spectra after specific charging steps and B) DRT analysis with the attached table of lumped serial elements for group L.

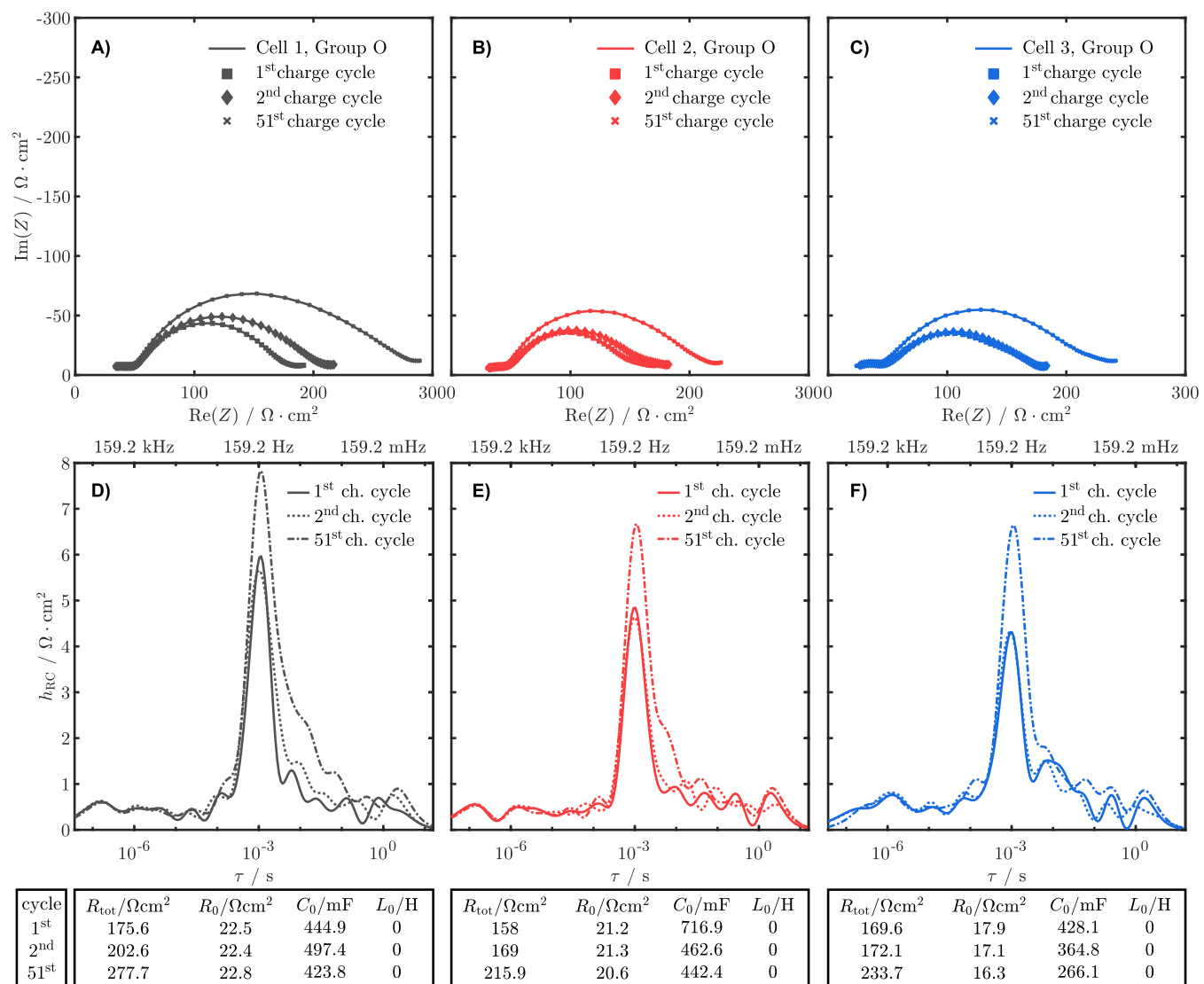


Supplementary Figure 35. Impedance spectra and DRT analysis of group M. A) to C) Impedance spectra after specific charging steps and D) to F) DRT analysis with the attached table of lumped serial elements for group M.

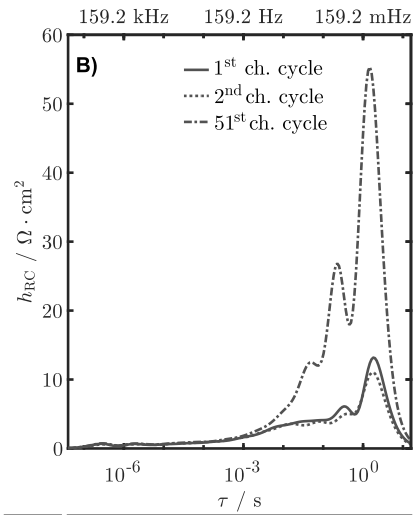
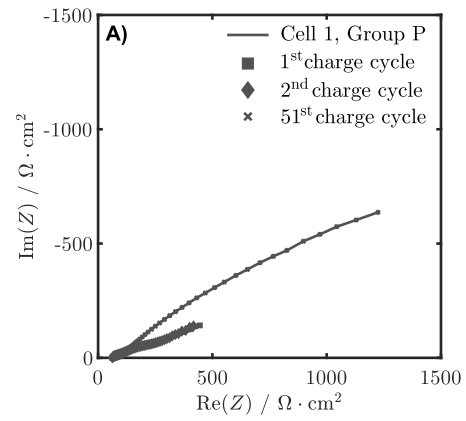


cycle	$R_{\text{tot}} / \Omega \text{cm}^2$	$R_0 / \Omega \text{cm}^2$	C_0 / mF	L_0 / H
1 st	139.4	39.5	967	0
2 nd	165.3	46.5	514.5	0
51 st	227.5	54.9	826.7	0

Supplementary Figure 36. Impedance spectra and DRT analysis of group N. A) Impedance spectra after specific charging steps and B) DRT analysis with the attached table of lumped serial elements for group N.

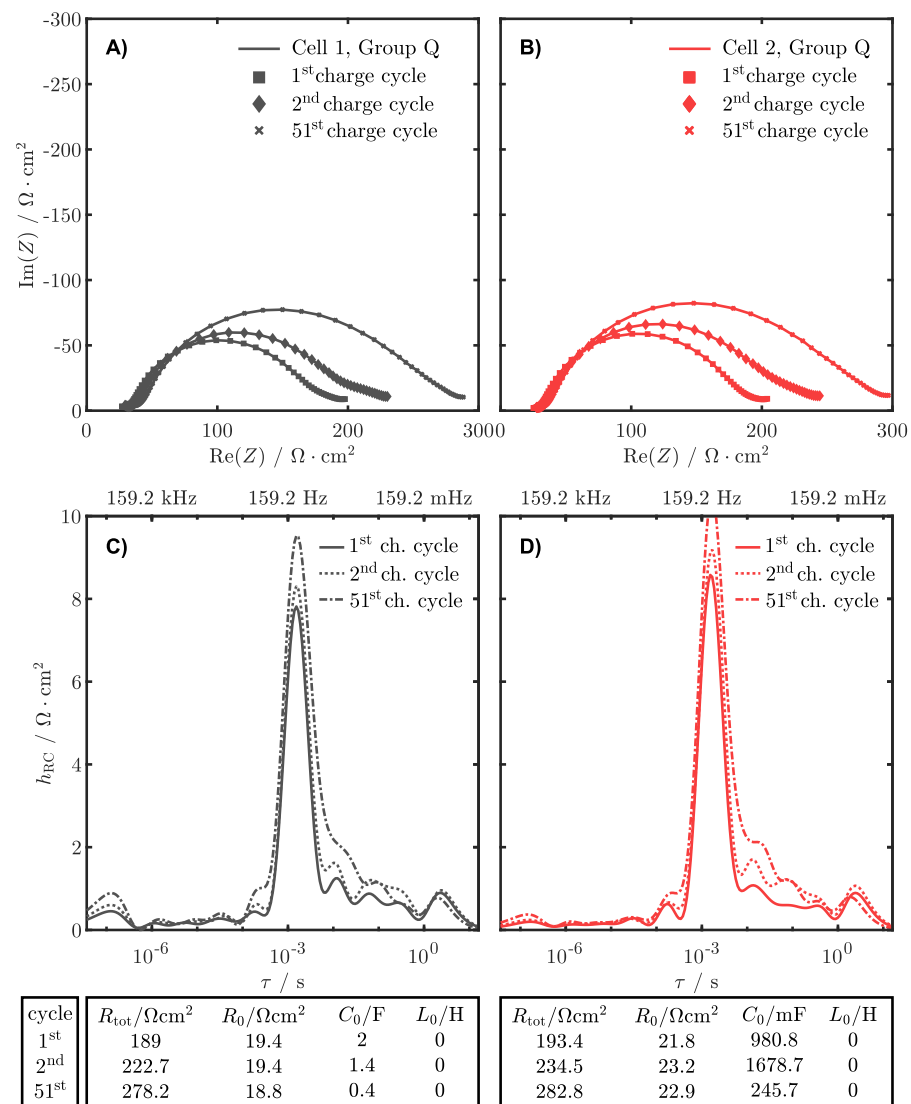


Supplementary Figure 37. Impedance spectra and DRT analysis of group O. A) to C) Impedance spectra after specific charging steps and D) to F) DRT analysis with the attached table of lumped serial elements for group O.

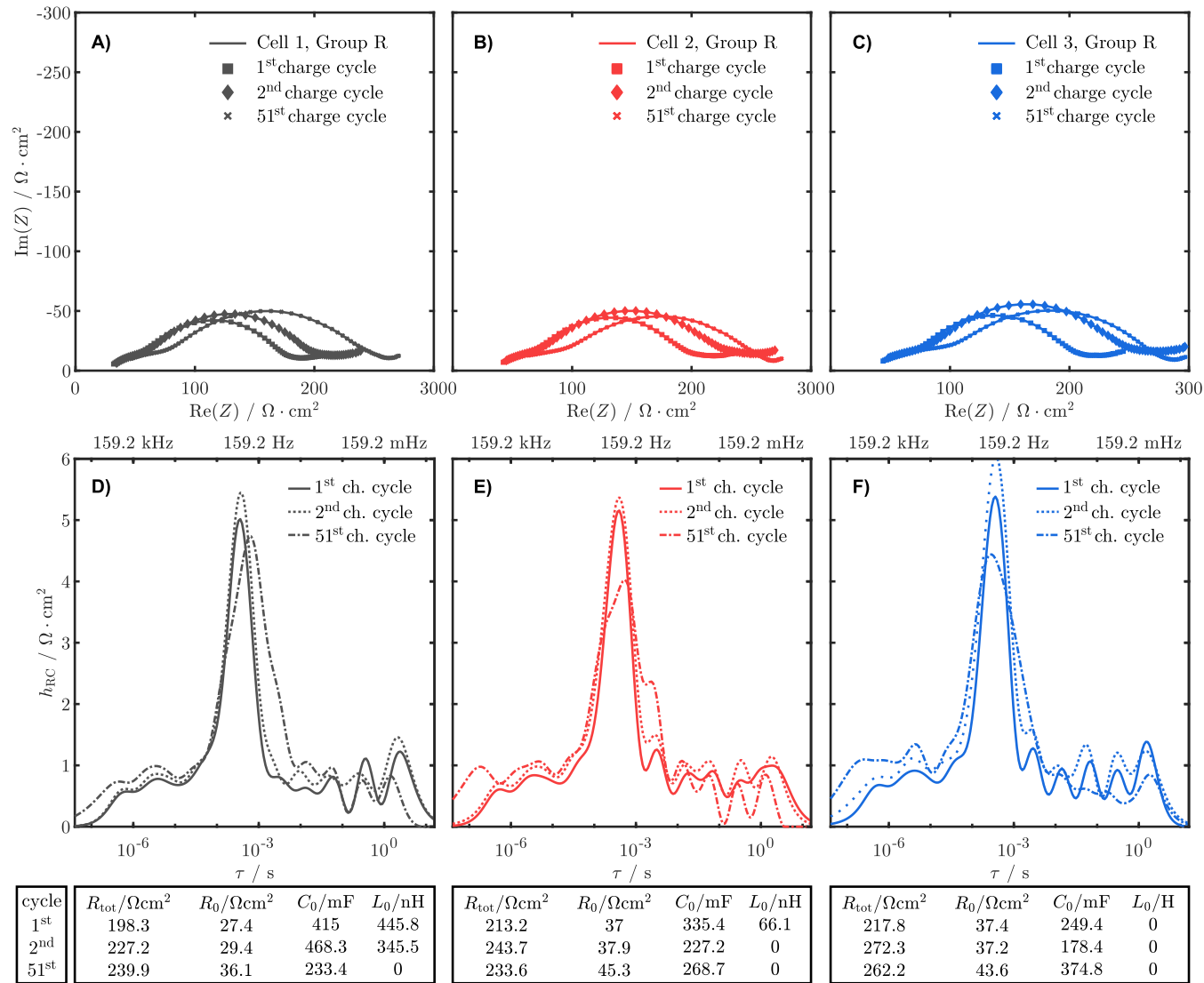


cycle	$R_{\text{tot}}/\Omega\text{cm}^2$	$R_0/\Omega\text{cm}^2$	C_0/mF	$L_0/\mu\text{H}$
1 st	535.6	49.9	42.6	7.7
2 nd	485.3	51.2	29.9	7.5
51 st	1640.8	51.4	6.5	7.7

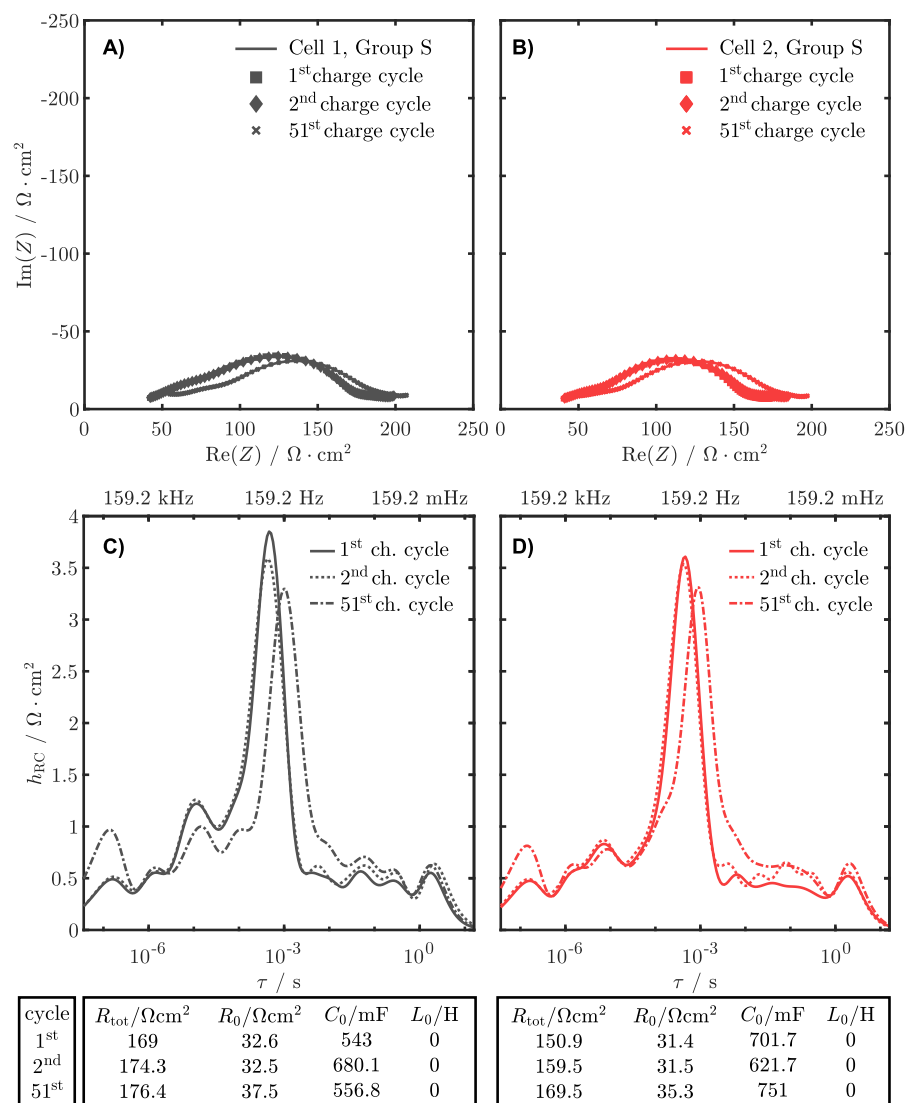
Supplementary Figure 38. Impedance spectra and DRT analysis of group P. A) Impedance spectra after specific charging steps and B) DRT analysis with the attached table of lumped serial elements for group P.



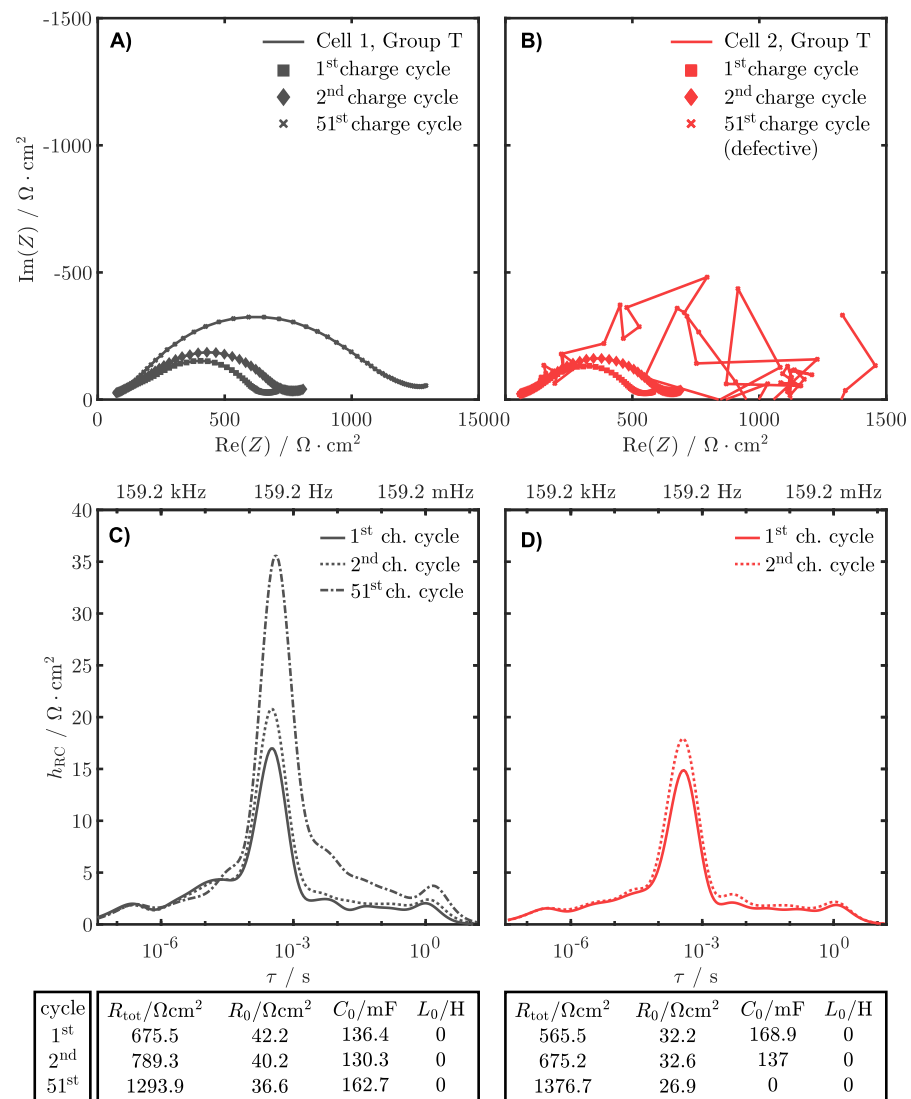
Supplementary Figure 39. Impedance spectra and DRT analysis of group Q. A), B) Impedance spectra after specific charging steps and C), D) DRT analysis with the attached table of lumped serial elements for group Q.



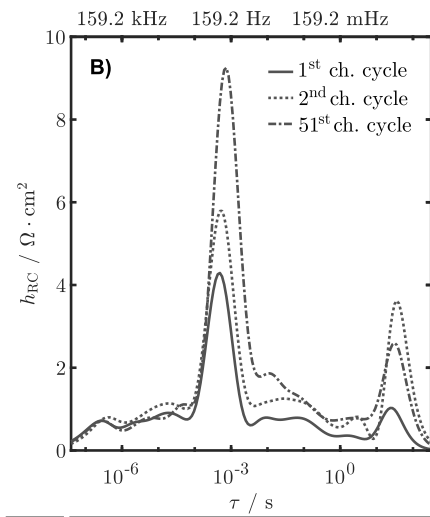
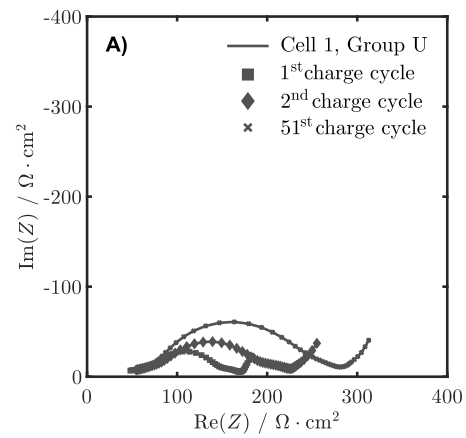
Supplementary Figure 40. Impedance spectra and DRT analysis of group R. A) to C) Impedance spectra after specific charging steps and D) to F) DRT analysis with the attached table of lumped serial elements for group R.



Supplementary Figure 41. Impedance spectra and DRT analysis of group S. A), B) Impedance spectra after specific charging steps and C), D) DRT analysis with the attached table of lumped serial elements for group S.

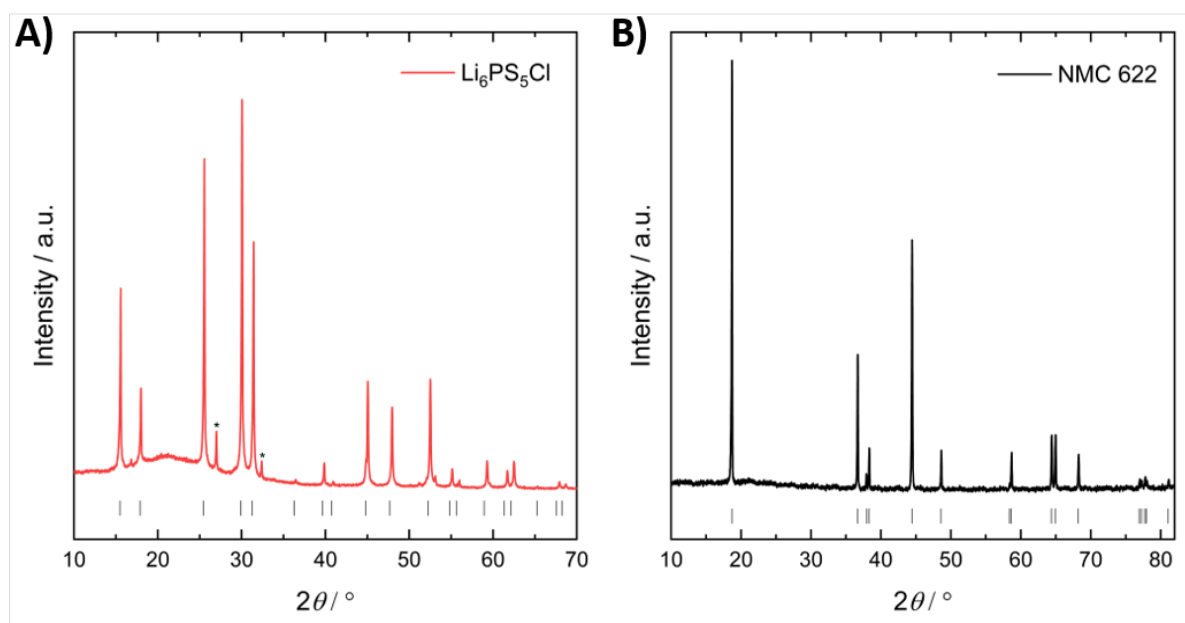


Supplementary Figure 42. Impedance spectra and DRT analysis of group T. A), B) Impedance spectra after specific charging steps and C), D) DRT analysis with the attached table of lumped serial elements for group T.

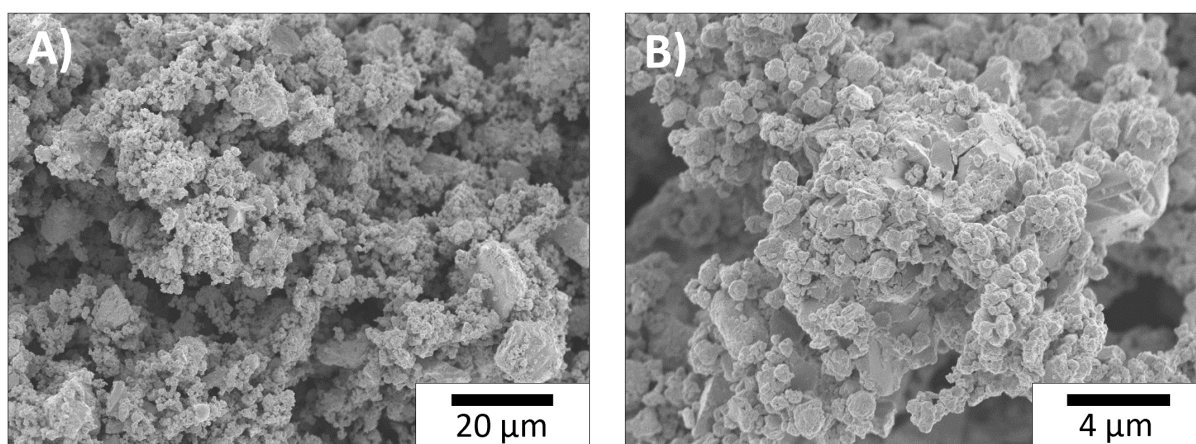


cycle	$R_{tot}/\Omega\text{cm}^2$	$R_0/\Omega\text{cm}^2$	C_0/F	L_0/H
1 st	149.3	39	2.5	0
2 nd	235.7	46.2	2.9	0
51 st	286.3	45.6	1.9	0

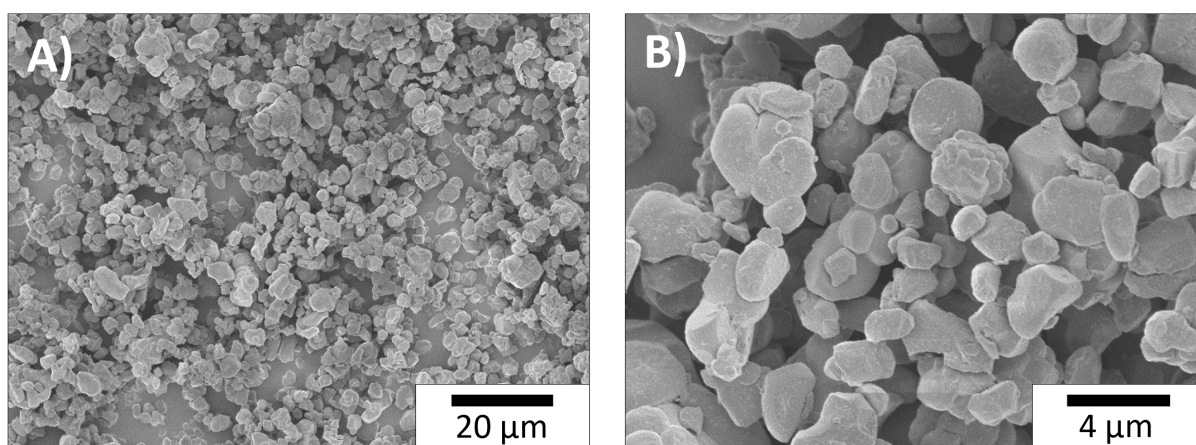
Supplementary Figure 43. Impedance spectra and DRT analysis of group U. A) Impedance spectra after specific charging steps and B) DRT analysis with the attached table of lumped serial elements for group U.



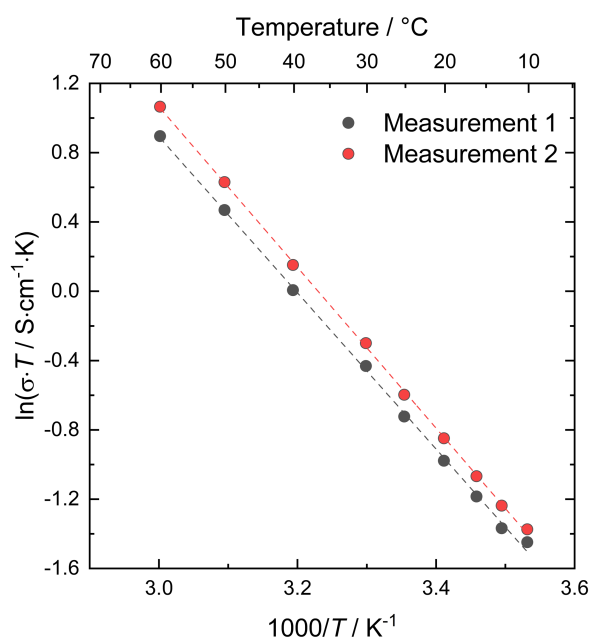
Supplementary Figure 44. X-ray diffraction (XRD) patterns. A) lithium argyrodite and B) NMC 622. The reflections marked with an asterisk (*) are originating from Li_2S .



Supplementary Figure 45. Scanning electron microscope (SEM) images of $\text{Li}_6\text{PS}_5\text{Cl}$. A) 1000 times and B) 5000 times magnification.



Supplementary Figure 46. Scanning electron microscope (SEM) images of $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$. A) 1000 times and B) 5000 times magnification.



Supplementary Figure 47. Arrhenius plot for the pristine Li₆PS₅Cl. The ionic conductivity σ_{ion} of the Li₆PS₅Cl was calculated by $\sigma_{\text{ion}} = \frac{l}{R \cdot A}$ with l being the thickness of the pellet, A the area of the electrodes and R the resistance, determined from the fitting of the PEIS data with a R-CPE equivalent circuit. Two measurements were performed.