Supporting Information

Time-Resolved and Robust Lithium Plating Detection for Automotive Lithium-Ion Cells with the Potential for Vehicle Application

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Temperature dependence

The data shown in Figure 2 has been measured with an initial cell temperature of 25 °C. During fast charging, the cell temperature increases by up to $20\mathrm{K}$ (see Figure 4a in [1] for details). In addition, even though initial conditions were identical, the maximum temperature varies for different fast charging cycles for about $10\mathrm{K}$. Hence, the proposed method is valid in the temperature range relevant for automotive fast charging. The fact that the increase of $\Delta\mathrm{T}_{\mathrm{Re-Im}}$ is not simply an artefact of the increased cell temperature can also be highlighted by a comparison of 2C CCCV vs P03 CCCA charging: Only the former 2C CCCV charging shows an increase of $\Delta\mathrm{T}_{\mathrm{Re-Im}}$ (Figure 2b) while both charging protocols cause identical maximum temperatures of around $37°\mathrm{C}$ (Figure 4a in [1]).

The applicability of the proposed method for low temperature charging needs to be checked; this is beyond the scope of the present manuscript.

Correlation between impedance measurement and cell temperature

The data shown in Table S1 summarizes the information of the linear regression line to calculate the cell temperature based on the real or imaginary part of the impedance measurement for each cycle during the test procedure. The regression coefficients x_0 and α are determined for each cycle during a temperature change at open circuit voltage conditions. Therefore, the cell temperature and impedance are measured at the same time. The maximum value of the real and imaginary part ($\mathrm{Re}_{\mathrm{max}}$ and $\mathrm{Im}_{\mathrm{max}}$) depict a measurement point in a temperature range between $27.8°\mathrm{C}$ and $28.3°\mathrm{C}$. The

minimum value of the real and imaginary part ($\mathrm{Re}_{\mathrm{min}}$ and $\mathrm{Im}_{\mathrm{min}}$) depict a measurement point in a temperature range between 24.8 °C and 25.3 °C.

Table S1: Information of the linear regression line to calculate the cell temperature based on impedance data.

Cycle	Fast-charge profile	$\mathrm{Re}_{\mathrm{max}}$ [mΩ]	$\mathrm{Re}_{\mathrm{min}}$ [mΩ]	$x_{0,\mathrm{Re}}$	α_{Re}	$\mathrm{Im}_{\mathrm{max}}$ [mΩ]	$\mathrm{Im}_{\mathrm{min}}$ [mΩ]	$x_{0,\mathrm{Im}}$	α_{Im}
1	1C CCCV	0.843	0.826	172.209	-174.721	0.361	0.346	-36.267	177.586
2	1C CCCV	0.842	0.826	164.524	-165.594	0.362	0.347	-31.262	163.185
3	1C CCCV	0.842	0.826	170.697	-173.056	0.362	0.347	-35.160	173.892
4	1C CCCV	0.842	0.827	185.228	-190.468	0.36	0.348	-37.577	180.489
5	P01	0.842	0.825	160.836	-161.210	0.364	0.35	-34.808	172.196
6	P01	0.841	0.825	158.296	-158.251	0.364	0.35	-32.783	166.144
7	P01	0.841	0.825	158.008	-157.944	0.365	0.351	-34.183	169.788
8	P01	0.841	0.822	180.630	-185.014	0.367	0.352	-45.123	200.502
9	P02	0.841	0.825	158.575	-158.637	0.366	0.352	-34.876	171.262
10	P02	0.84	0.824	159.699	-160.041	0.366	0.353	-35.688	173.194
11	P02	0.841	0.825	158.942	-159.136	0.367	0.353	-37.327	177.555
12	P02	0.84	0.822	181.447	-186.100	0.368	0.354	-50.296	213.764
13	P03	0.84	0.825	159.653	-160.053	0.368	0.355	-38.526	180.409
14	P03	0.84	0.824	159.082	-159.406	0.368	0.355	-39.603	183.085
15	P03	0.84	0.824	155.324	-154.909	0.369	0.355	-38.461	179.691
16	P03	0.84	0.822	187.101	-192.994	0.37	0.356	-55.078	225.921
17	1.5C CCCV	0.839	0.824	171.398	-174.422	0.369	0.356	-41.851	188.453
18	1.5C CCCV	0.838	0.823	172.057	-175.391	0.37	0.357	-41.160	186.281
19	1.5C CCCV	0.837	0.822	174.487	-178.546	0.37	0.358	-43.544	192.512
20	1.5C CCCV	0.836	0.817	206.449	-217.171	0.373	0.358	-56.814	229.082
21	P04	0.836	0.821	179.292	-184.627	0.372	0.359	-47.334	202.548
22	P04	0.834	0.82	181.078	-187.145	0.371	0.359	-47.418	202.540
23	P04	0.831	0.818	197.456	-207.412	0.372	0.36	-49.653	208.538
24	P04	0.829	0.812	232.199	-249.918	0.373	0.36	-69.026	261.816
25	P05	0.829	0.817	218.450	-233.456	0.372	0.362	-60.013	235.549
26	P05	0.829	0.817	208.089	-220.944	0.373	0.362	-55.864	223.997
27	P05	0.829	0.817	217.967	-233.001	0.373	0.362	-59.270	232.978
28	P05	0.828	0.818	225.467	-242.149	0.372	0.363	-70.706	264.171
29	P01	0.828	0.817	215.389	-229.945	0.374	0.364	-60.503	235.742
30	P01	0.828	0.817	214.493	-228.913	0.375	0.364	-60.241	234.883
31	P01	0.829	0.816	211.197	-224.862	0.374	0.364	-62.048	239.603
32	P01	0.829	0.809	208.167	-221.196	0.379	0.365	-72.423	267.614
33	2C CCCV	0.828	0.816	222.595	-238.927	0.376	0.366	-66.624	251.370
34	2C CCCV	0.828	0.816	217.493	-232.692	0.376	0.366	-65.703	248.873
35	2C CCCV	0.827	0.815	224.537	-241.484	0.377	0.365	-66.906	252.109
36	2C CCCV	0.825	0.81	237.247	-257.415	0.378	0.366	-82.772	295.076
37	2.5C CCCV	0.825	0.811	235.254	-254.793	0.378	0.367	-71.809	264.794
38	2.5C CCCV	0.825	0.811	238.739	-259.125	0.377	0.366	-75.398	274.922
39	2.5C CCCV	0.824	0.81	240.620	-261.410	0.378	0.367	-96.297	331.566
40	2.5C CCCV	0.825	0.807	244.619	-266.358	0.379	0.367	-106.176	358.042

Figure S1 shows the regression lines based on the information of Table . The change of the regression line with respect to the fast-charging cycle results from the aging of the cell during cycling.

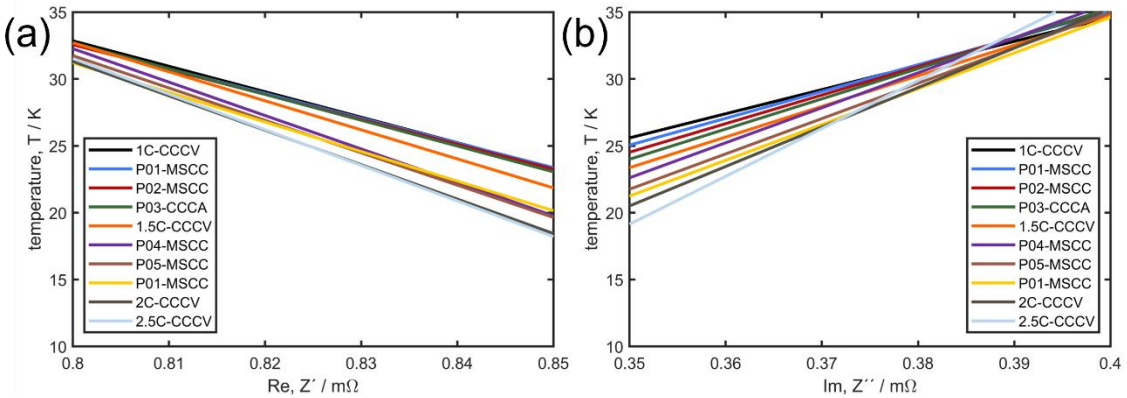

Figure S1: Regression lines to determine the cell temperature for each fast-charging profile based on impedance measurements: (a) real part in correlation to cell temperature and (b) imaginary part in correlation to cell temperature.

Critical fast charging with the occurrence of lithium plating

Figure S2 shows the current and voltage signal as well as the impedance and temperature behavior of a cell during a 2.5C-CCCV charging process as an example of critical fast charging with the occurrence of lithium plating. The difference of T_{Re} and T_{Im} can be attributed to the occurrence of lithium-plating.

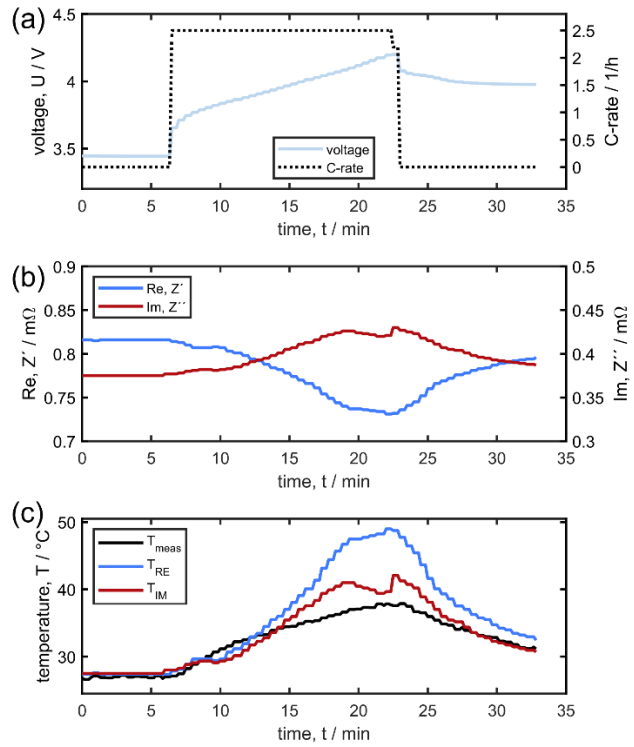

Figure S2: Critical fast charging with 2.5C at 25 °C initial temperature with the occurrence of lithium plating (see Figure 2b), a) cell potential and current, b) real and imaginary cell impedance measured at 3125 Hz, c) cell temperatures measured with an external temperature sensor and derived from the cell impedance.