**Optimisation of Industrially Relevant Electrode Formulations for LFP Cathodes in Lithium Ion Cells**

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**Table S1.** Design Of Experiment Mix Formulations.

| Mix No. | LFP/wt% | KS6L/wt% | SWCNT/wt% | PVDF/wt% |
|---------|---------|----------|-----------|----------|
| 01 | 96.98 | 1.91 | 0.19 | 0.92 |
| 02 | 96.40 | 2.80 | 0.05 | 0.75 |
| 03 | 95.32 | 2.80 | 0.12 | 1.76 |
| 04 | 94.67 | 2.80 | 0.20 | 2.33 |
| 05 | 96.49 | 1.49 | 0.05 | 1.97 |
| 06 | 97.12 | 0.70 | 0.20 | 1.98 |
| 07 | 96.20 | 1.83 | 0.12 | 1.85 |
| 08 | 94.17 | 2.78 | 0.05 | 3.00 |
| 09 | 98.47 | 0.70 | 0.08 | 0.75 |
| 10 | 96.00 | 2.80 | 0.20 | 1.00 |
| 11 | 94.91 | 1.92 | 0.16 | 3.01 |
| 12 | 96.20 | 0.70 | 0.10 | 3.00 |
| 13 | 96.84 | 1.75 | 0.12 | 1.29 |
| 14 | 97.00 | 0.70 | 0.05 | 2.25 |
| 15 | 96.27 | 1.75 | 0.14 | 1.84 |
| 16 | 97.10 | 2.10 | 0.05 | 0.75 |
| 17 | 95.86 | 2.80 | 0.09 | 1.25 |

**Table S2.** Electrochemical Cell Test Sequence.

| Step | Name | Cycles | Charge | Discharge | Voltage Range |
|------|------|--------|--------|-----------|---------------|
| #01 | Formation | 1 | C / 20 | C / 20 | 2.80 – 3.95 |
| #02 | Conditioning | 5 | C / 5 | C / 5 | 2.80 – 3.95 |
| #03 | Battery Cap. Determination | 1 | C / 5 | C / 5 | 2.80 – 3.95 |
| #04 | Rate Test | 5 | C / 5 | C / 2 – 10 C | 2.80 – 3.95 |
| #05 | ASI Measurement | 2 | C / 5 | 1.8 C | 2.80 – 3.95 |
| #06 | Impedance | 1 | C / 5 | C / 5 | 2.80 – 3.95 |
| #07 | Battery Cap. Determination | 1 | C / 5 | C / 5 | 2.80 – 3.95 |
| #08 | Capacity Measure | 1 | C / 10 | C / 10 | 2.80 – 3.95 |
| #09 | Cycling | 50 | C / 2 | C / 2 | 2.80 – 3.95 |
| #10 | Capacity Measure | 1 | C / 10 | C / 10 | 2.80 – 3.95 |
| #11 | Impedance | 1 | C / 5 | C / 5 | 2.80 – 3.95 |

**Table S3.** Electrode Mixing, Coating And Adhesion.

| Mix No. | Blade Gap /m | Speed /m min$^{-1}$ | Viscosity @ 10 s$^{-1}$ /Pa s | Adhesion / kPa | |
|---|---|---|---|---|---|
| | | | | Uncalendered | Calendered |
| 01 | 270 | 0.24 | 1.83 | 71.5 | 101.3 |
| 02 | 280 | 0.24 | 0.48 | 29.6 | 62.2 |
| 03 | 300 | 0.30 | 2.10 | 263.1 | 321.2 |
| 04 | 280 | 0.18 | 4.49 | 282.9 | 227.3 |
| 05 | 310 | 0.36 | 0.99 | 339.7 | 370.0 |
| 06 | 310 | 0.39 | 2.95 | 279.5 | 445.6 |
| 07 | 300 | 0.30 | 1.78 | 270.1 | 450.5 |
| 08 | 310 | 0.30 | 1.98 | 434.2 | 477.8 |
| 09 | 260 | 0.30 | 0.61 | 32.3 | 96.9 |
| 10 | 270 | 0.30 | 2.38 | 28.0 | 163.3 |
| 11 | 400 | 0.12 | 2.51 | 277.4 | 478.2 |
| 12 | 340 | 0.30 | 2.71 | 540.2 | 561.7 |
| 13 | 340 | 0.43 | 1.26 | 56.4 | 85.6 |
| 14 | 340 | 0.36 | 1.13 | 325.7 | 432.5 |
| 15 | 300 | 0.24 | 1.92 | 154.5 | 414.6 |
| 16 | 280 | 0.24 | 0.25 | 56.3 | 73.9 |
| 17 | 280 | 0.27 | 0.62 | 181.0 | 150.8 |

**Table S4.** Coating Resistivity Measurements.

| Mix No. | Vol. Resistivity/$\Omega$ cm | | Interface Res./$\Omega$ cm$^2$ | | Total Through Plane/$\Omega$ cm$^2$ | |
|---|---|---|---|---|---|---|
| | Uncalendered | Calendered | Uncalendered | Calendered | Uncalendered | Calendered |
| 01 | 3.3 | 1.9 | 1.0 | 0.6 | 1.0 | 0.7 |
| 02 | 9.3 | 4.5 | 9.6 | 0.9 | 9.7 | 0.9 |
| 03 | 3.5 | 1.9 | 0.9 | 0.3 | 1.0 | 0.4 |
| 04 | 1.6 | 1.0 | 0.5 | 0.2 | 0.5 | 0.2 |
| 05 | 14.6 | 12.8 | 6.4 | 2.0 | 6.6 | 2.1 |
| 06 | 3.6 | 2.5 | 0.6 | 0.7 | 0.6 | 0.7 |
| 07 | 6.9 | 3.3 | 1.9 | 0.5 | 2.0 | 0.5 |
| 08 | 8.4 | 5.3 | 5.9 | 0.8 | 6.0 | 0.8 |
| 09 | 17.7 | 11.5 | 8.7 | 2.8 | 9.0 | 2.9 |
| 10 | 4.3 | 2.3 | 0.4 | 0.5 | 0.5 | 0.5 |
| 11 | 1.4 | 0.9 | 0.4 | 0.2 | 0.4 | 0.2 |
| 12 | 8.6 | 7.4 | 3.4 | 1.0 | 3.5 | 1.1 |
| 13 | 6.5 | 4.0 | 2.0 | 0.4 | 2.2 | 0.5 |
| 14 | 24.2 | 17.8 | 13.8 | 2.9 | 14.2 | 3.0 |
| 15 | 6.6 | 3.0 | 1.8 | 0.3 | 3.3 | 0.3 |
| 16 | 21.9 | 15.4 | 11.9 | 2.6 | 12.3 | 2.7 |
| 17 | 10.9 | 9.0 | 4.9 | 2.5 | 5.1 | 2.6 |

**Table S5.** Electrode Performance.

| Mix No. | Coat Weight /gsm | Porosity /% | FCL /% | ASI (Min.) /S cm$^{-2}$ | $D_{51}/D_{00}$ /% | $D_{50}/D_{01}$ /% |
|---|---|---|---|---|---|---|
| 01 | 175.4 | 38.9 | 1.9 | 0.041 | 98.8 | 99.7 |
| 02 | 177.8 | 39.7 | 1.6 | 0.039 | 99.8 | 97.4 |
| 03 | 169.9 | 37.1 | 2.0 | 0.037 | 99.3 | 100.6 |
| 04 | 165.9 | 38.2 | 1.9 | 0.044 | 99.5 | 100.6 |
| 05 | 172.0 | 41.2 | 2.2 | 0.019 | 98.5 | 97.2 |
| 06 | 177.2 | 37.6 | 1.8 | 0.042 | 97.9 | 97.9 |
| 07 | 173.4 | 37.4 | 2.0 | 0.038 | 98.8 | 99.7 |
| 08 | 154.9 | 36.8 | 1.8 | 0.023 | 99.4 | 99.3 |
| 09 | 167.4 | 40.1 | 1.9 | 0.026 | 97.9 | 96.9 |
| 10 | 170.8 | 38.9 | 1.9 | 0.033 | 99.0 | 99.7 |
| 11 | 168.7 | 36.5 | 2.4 | 0.041 | 99.4 | 100.0 |
| 12 | 169.1 | 35.1 | 2.0 | 0.033 | 98.9 | 98.5 |
| 13 | 184.6 | 37.7 | 1.9 | 0.037 | 98.1 | 96.7 |
| 14 | 179.3 | 35.4 | 1.9 | 0.028 | 98.4 | 97.8 |
| 15 | 157.1 | 36.3 | 1.8 | 0.043 | 99.2 | 100.3 |
| 16 | 170.3 | 39.6 | 1.7 | 0.029 | 96.6 | 90.1 |
| 17 | 161.8 | 39.2 | 1.6 | 0.035 | 97.3 | 93.7 |

**Table S6.** Rate Performance Measurements.

| Mix No. | Discharge Capacity/mA hr g$^{-1}$ | | | | | | |
|---|---|---|---|---|---|---|---|
| | C/20 | C/5 | C/2 | C | 2C | 5C | 10 C |
| 01 | 159.7 | 152.5 | 144.1 | 133.8 | 116.1 | 73.8 | 12.1 |
| 02 | 160.1 | 152.3 | 143.5 | 130.7 | 113.1 | 78.6 | 8.3 |
| 03 | 159.6 | 152.1 | 143.7 | 133.0 | 115.6 | 71.6 | 5.1 |
| 04 | 159.8 | 152.4 | 144.1 | 133.8 | 118.8 | 89.2 | 24.9 |
| 05 | 159.1 | 151.3 | 137.6 | 124.9 | 95.6 | 3.5 | 0.1 |
| 06 | 160.6 | 153.2 | 144.8 | 134.4 | 118.3 | 82.6 | 13.3 |
| 07 | 159.8 | 152.0 | 144.1 | 133.5 | 116.9 | 79.3 | 9.6 |
| 08 | 159.2 | 151.8 | 143.5 | 132.2 | 113.3 | 57.7 | 0.5 |
| 09 | 159.7 | 151.6 | 142.4 | 128.3 | 95.4 | 20.3 | 0.1 |
| 10 | 159.9 | 152.5 | 144.4 | 134.4 | 118.8 | 80.9 | 4.9 |
| 11 | 156.6 | 149.0 | 140.8 | 130.8 | 115.4 | 79.6 | 14.7 |
| 12 | 158.9 | 151.5 | 142.7 | 131.2 | 113.0 | 69.1 | 3.5 |
| 13 | 159.1 | 151.7 | 143.5 | 132.7 | 114.9 | 69.8 | 3.9 |
| 14 | 158.1 | 150.8 | 142.3 | 130.9 | 109.9 | 45.9 | 0.5 |
| 15 | 159.5 | 152.2 | 144.2 | 134.4 | 119.5 | 93.7 | 25.4 |
| 16 | 161.3 | 152.5 | 143.6 | 130.7 | 110.6 | 61.8 | 1.1 |
| 17 | 158.7 | 151.5 | 143.2 | 131.4 | 115.2 | 79.2 | 8.0 |

| Viscosity vs. SWCNT content | Viscosity vs. PVDF content |
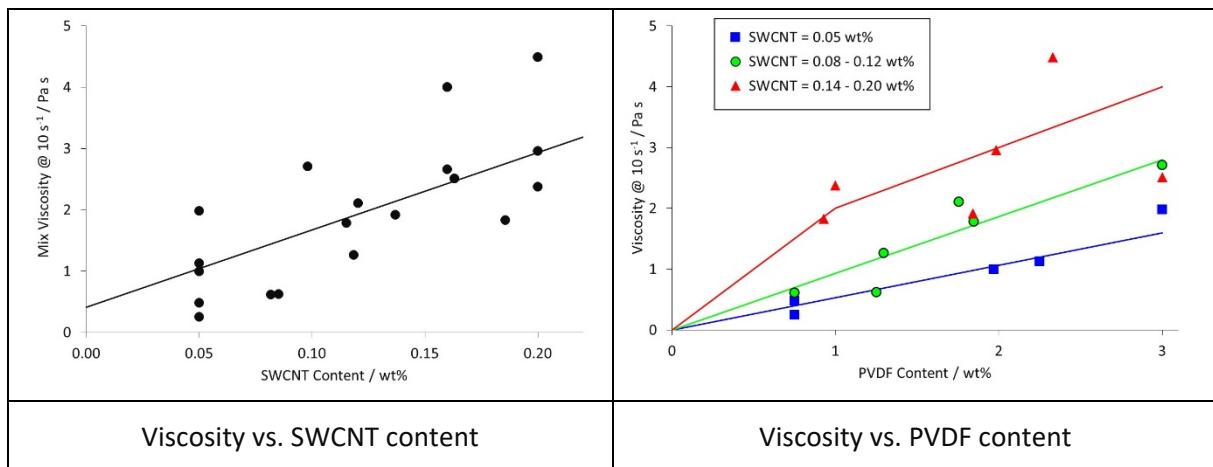
**Figure S1.** Variation Of Mix Viscosity With SWCNT And PVDF Content In Mix.



| Adhesion; Calendered vs. Uncalendered | Porosity vs. PVDF Content |
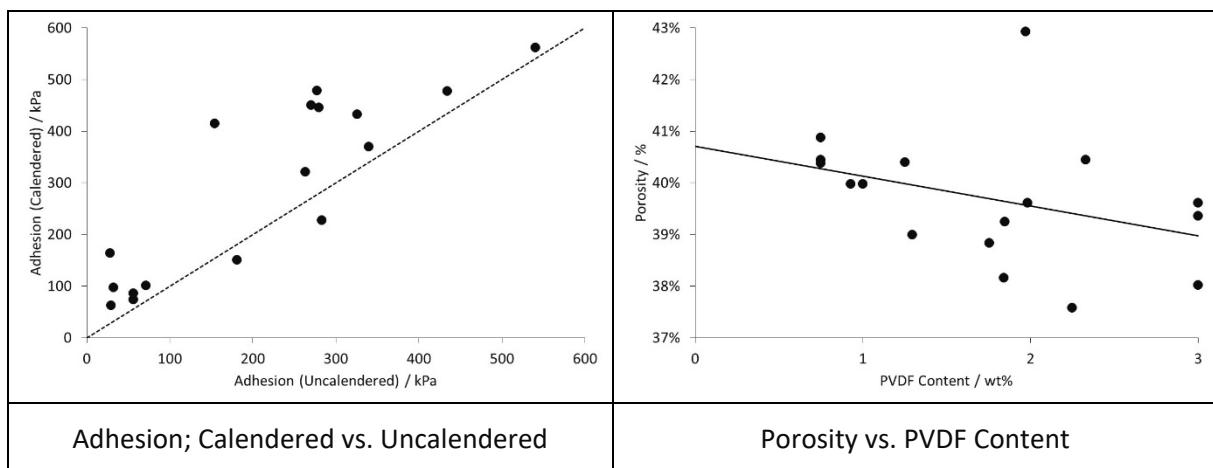
**Figure S2.** Adhesion And Porosity Plots.



| Volumetric Resistivities | Interface Resistances |
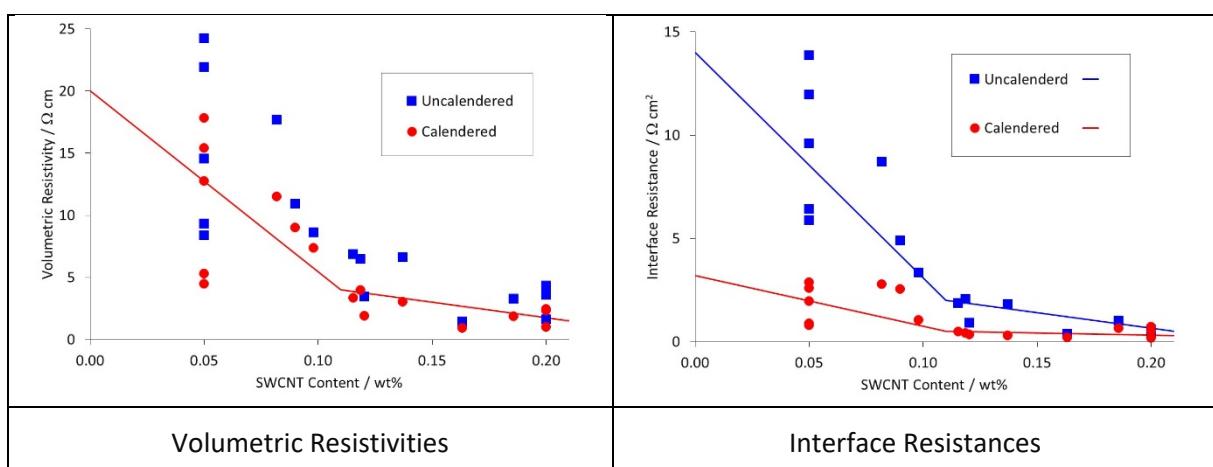
**Figure S3.** Variations In Volumetric Resistivity And Interface Resistance With SWCNT Content.

**Figure S4.** Experimentally Measured Variations In Coatings.



**Figure S5.** Results From Formation Cycle At ± C/20.



**Figure S6**. Results From Rate Tests.

| ASI Conductivity vs. SWCNT Content | ASI Conductivity vs. PVDF Content |
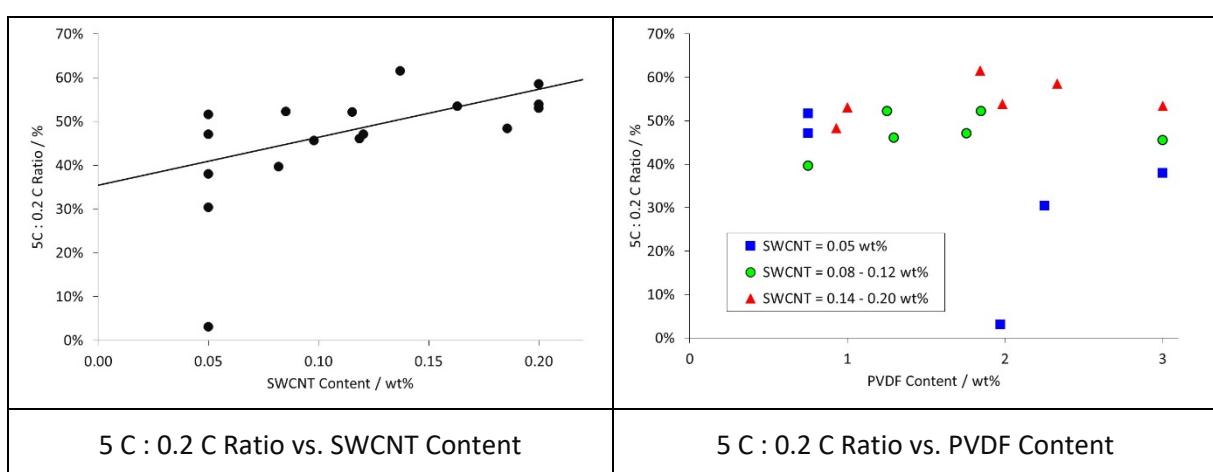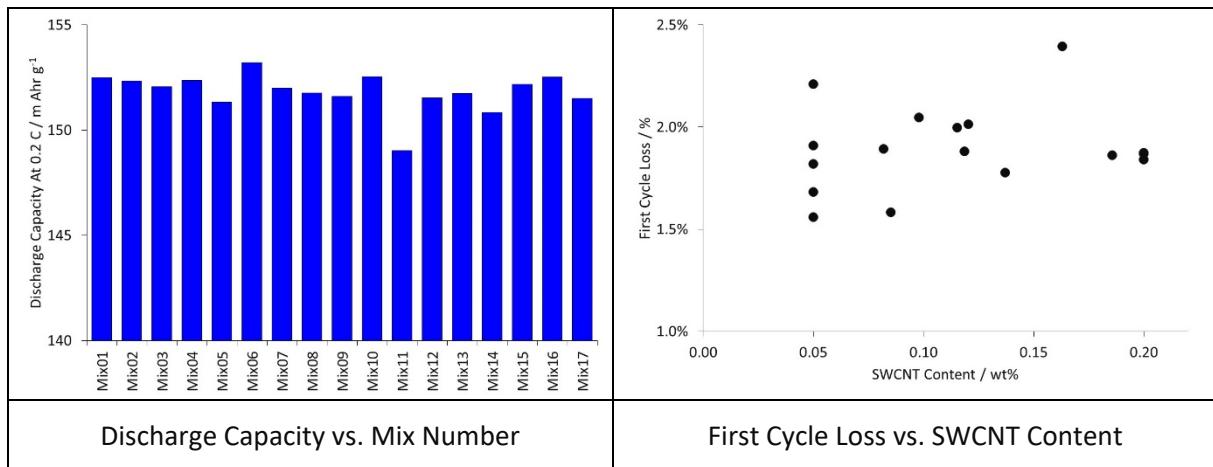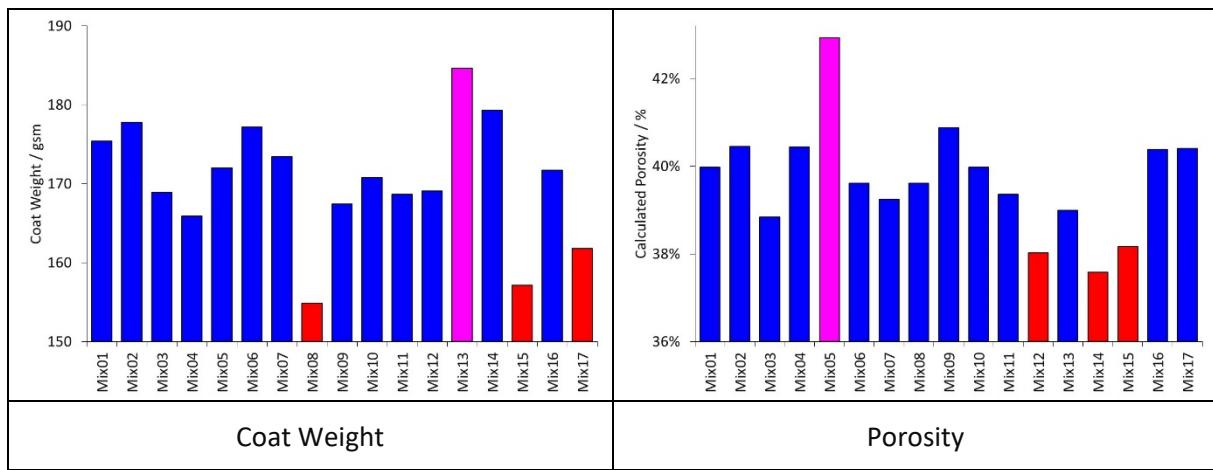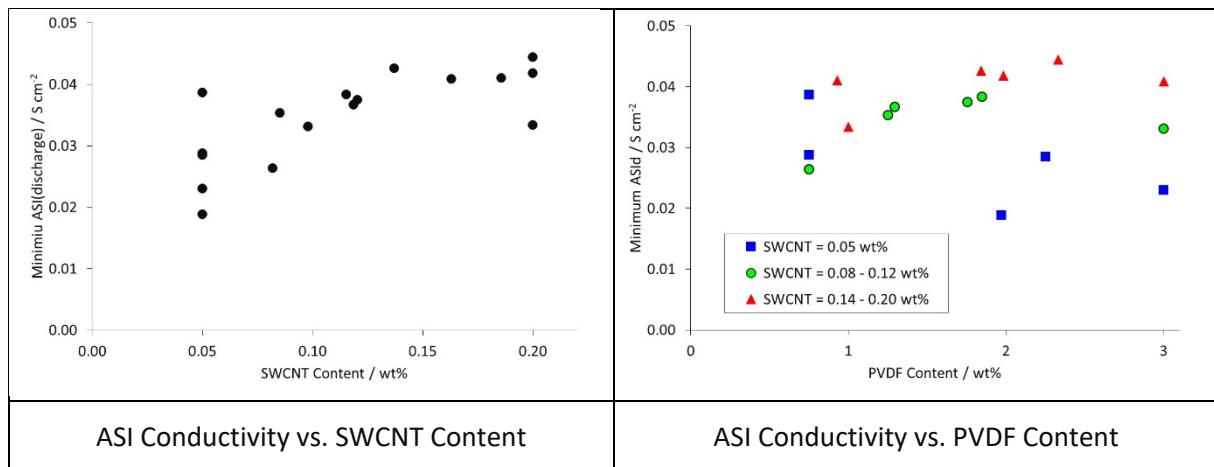
**Figure S7.** Results From ASI Measurements.



| $D_{51} : D_{00}$ Ratio vs. Coat Weight | $D_{50} : D_{01}$ Ratio vs. Coat Weight |
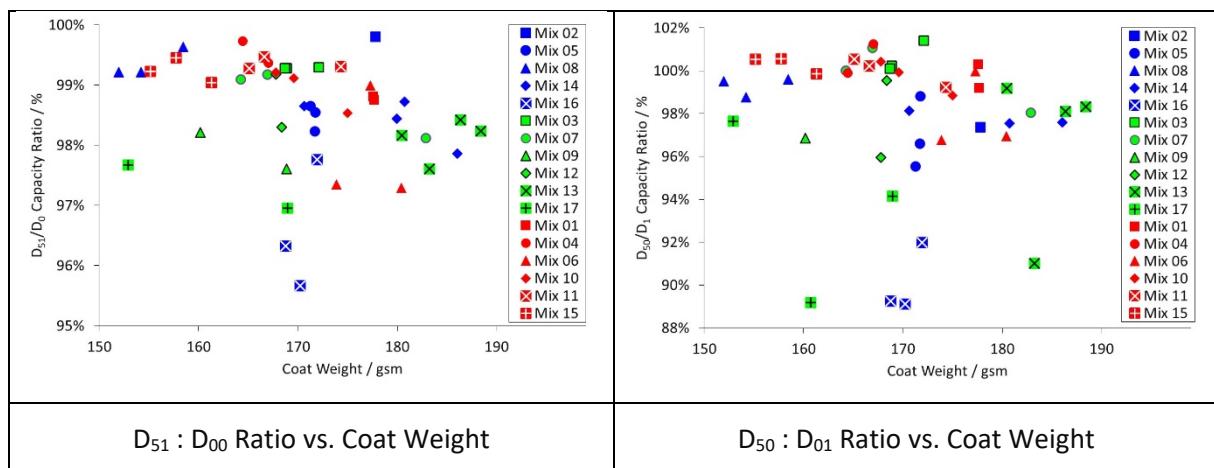
**Figure S8.** Results From Cell Cycling Measurements.



| Before Cycling | After Cycling |
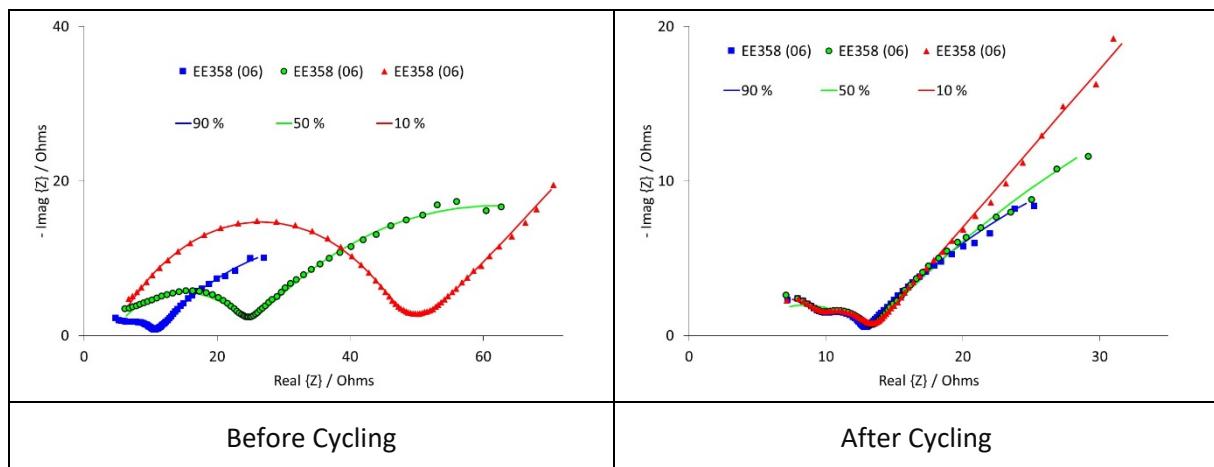
**Figure S9.** Impedance Spectra For Cell EE358.

## Modelling

Pearson's Coefficient
$$\rho = \frac{\sum_i^n (x_i - \bar{x})(y_i - \bar{y})}{\sqrt{\sum_i^n (x_i - \bar{x})^2}\sqrt{\sum_i^n (y_i - \bar{y})^2}}$$
(S1)

In this equation, $x_i$ and $y_i$ are points from features x and y, and $\bar{x}$ and $\bar{y}$ are their respective averages.

Coefficient of determination
$$R_{adj}^2 = 1 - \left(\frac{\sum_i^n (y_i - \hat{y}_i)^2}{\sum_i^n (y_i - \bar{y})^2}\right)\left(\frac{df_{tot}}{df_{resid}}\right)$$
(S2)

In this equation, i is the index of the experiment, n is the total number of experiments, $y_i$ and $\hat{y}_i$ are the actual and predicted values of the ith experiment, $\bar{y}$ is the mean of all values of $y_i$, and $df_{resid}$ and $df_{tot}$ are the degrees of freedom of the residuals and total, respectively.

The mixture only model has four input parameters, designated A to D. The output y is defined by equation S.3, with regression fitting parameters $\beta$, and a power transformation exponent $\lambda$. There are no quadratic terms; curvature is covered by the cross terms.

$$y^\lambda = \beta_A A + \beta_B B + \beta_C C + \beta_D D + \beta_{AB} AB + \beta_{AC} AC + \beta_{AD} AD + \beta_{BC} BC + \beta_{BD} BD + \beta_{CD} CD \quad (S3)$$

The mixture + process model includes two further process related inputs, designated E and F. The output is defined by Equation S.4, which does include some quadratic terms.

$$y^\lambda = \beta_A A + \beta_B B + \beta_C C + \beta_D D + \beta_{AB} AB + \beta_{AC} AC + \beta_{AD} AD + \beta_{BC} BC + \beta_{BD} BD + \beta_{CD} CD \quad (S4)$$

$$+ \beta_{AE} AE + \beta_{AF} AF + \beta_{BE} BE + \beta_{BF} BF + \beta_{CE} CE + \beta_{CF} CF + \beta_{DE} DE + \beta_{DF} DF + \beta_{EF} EF + \beta_{E2} E^2 + \beta_{F2} F^2$$

The power transformation exponent $\lambda$ was fitted using a Box-Cox transformation, based on the method of maximum likelihood. For some of the data, ln (y) was used instead of $y^\lambda$. Over-fitting can be a problem, leading to a model with accurate fitting of the training data, but poor fitting to the more general test data. To avoid this, model reduction was done via minimizing the corrected Akaike Information Criterion (AICc).

For the single variate models, the effects of each input on the output were quantified using Pearson's coefficients (S.1). For a multi-variate model, this comparison would normally be done using the regression fitting parameters, $\beta$. However, in a mixture model, the components are highly correlated with each other, and this comparison is invalid. The alternative is to plot accumulated local effects (ALE), which show the marginal effects of each component. The data set was divided into several small bins, and the difference in the response when the feature was varied within the small bin was calculated. These differences were then accumulated together to form the ALE plots. The bin sizes ranged from 5 - 8 points per bin. The ALE values were calculated using the PyALE package in Python.

**Table S7.** Adjusted Regression Coefficients For Different Models And Output Parameters.

| Model | Parameter | $R^2_{adj}$ for different discharge rates | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | C/20 | C/5 | C/2 | C | 2C | 5C | 10C |
| Mixture only | Capacity | 0.40 | 0.39 | 0.36 | 0.45 | 0.56 | 0.50 | 0.46 |
| | Gravimetric (active) | 0.92 | 0.97 | 0.79 | 0.66 | 0.75 | 0.51 | 0.41 |
| | Gravimetric (coating) | 0.64 | 0.84 | 0.54 | 0.57 | 0.70 | 0.50 | 0.41 |
| Mixture+Process | Capacity | 0.99 | 0.99 | 0.99 | 0.96 | 0.78 | 0.81 | 0.72 |
| | Gravimetric (active) | 0.64 | 0.57 | 0.52 | 0.45 | 0.70 | 0.78 | 0.72 |
| | Gravimetric (coating) | 0.97 | 0.89 | 0.83 | 0.58 | 0.67 | 0.77 | 0.72 |

**Table S8.** Multi-variate Regression Fitting To Adhesion And Resistance Output Parameters.

| Response | Adhesion | | Volumetric resistivity | | Interface resistance | |
|---|---|---|---|---|---|---|
| | Pre-cal | Post-cal | Pre-cal | Post-cal | Pre-cal | Post-cal |
| $R^2_{adj}$ | 0.80 | 0.79 | 0.87 | 0.81 | 0.83 | 0.68 |
| A | 74.2 | 149.9 | 3.2 | 3 | 3.8 | 1.4 |
| B | 60.4 | -54 | 1.7 | 1.2 | 3 | -0.4 |
| C | -1836.1 | 65.1 | -33.3 | -52.3 | -89.5 | -46.8 |
| D | 791.4 | 988.2 | 2.4 | 1.6 | 1.1 | -0.4 |
| A:B | | | | | | |
| A:C | | | | | | |
| A:D | | | | | | |
| B:C | | | | | | |
| B:D | | | | | | |
| C:D | | | -67.2 | | | |
| λ | 1 | 1 | ln | ln | ln | ln |

**Table S9.** Multi-variate Regression Fitting Coefficients (Capacity).

| Parameter | C/20 | C/5 | C/2 | C | 2C | 5C | 10C |
|---|---|---|---|---|---|---|---|
| A | 4.74 | 4.51 | 4.22 | 3.65 | 2.64 | 0.85 | - 0.05 |
| B | 3.92 | 3.74 | 3.51 | 3.47 | 3.11 | 1.76 | - 0.03 |
| C | 9.23 | 8.97 | 9.36 | 9.78 | 15.86 | 38.44 | 12.45 |
| D | 4.22 | 4.01 | 3.81 | 3.01 | 2.27 | 1.31 | 0.3 |
| A:B | | | | | | | |
| A:C | | | | | | | |
| A:D | | | | 1.85 | 3.09 | | |
| B:C | | | | | | | |
| B:D | | | | | | | |
| C:D | | | | | | | |
| λ | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 |

**Table S10.** Multi-variate Regression Fitting Coefficients (Capacity).

| Parameter | C/20 | C/5 | C/2 | C | 2C | 5C | 10C |
|---|---|---|---|---|---|---|---|
| A | 4.6 | 4.4 | 4.1 | 3.7 | 2.7 | 0.5 | 0.1 |
| B | 4.4 | 4.1 | 4.2 | 3.6 | 4 | 3.8 | -2.6 |
| C | 5.2 | 7 | 5 | 8.3 | 483.2 | 143.7 | 8.7 |
| D | 4.3 | 4.1 | 3.6 | 3.5 | 2.1 | -3.8 | -2.9 |
| A:B | | | -0.4 | | | | 4 |
| A:C | | | | | -486 | -154.3 | |
| A:D | | | 0.6 | 0.5 | 2.4 | 8.1 | 3.4 |
| A:E | 0.5 | 0.5 | 0.4 | 0.4 | | -1.3 | |
| A:F | | | | | | | -0.5 |
| B:C | | | | | -512 | -151.1 | |
| B:D | | | | | | | 10.1 |
| B:E | 0.3 | 0.3 | 0.4 | 0.4 | 1.2 | 1.6 | -1 |
| B:F | | | | | | 2.1 | 0.5 |
| C:D | | -6.5 | | -7.2 | -472 | | 29.7 |
| C:E | | | 2 | | | 54 | |
| C:F | | | | | 8.4 | | |
| D:E | 0.5 | 0.5 | 0.3 | 0.4 | | -3.3 | |
| D:F | | | | | -0.7 | -5 | |
| E:F | | | | | | | |
| E² | | | 0 | 0.1 | 0.4 | 0.7 | -0.2 |
| F² | | | | | | | |
| λ | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 |

**Table S11.** Multi-variate Regression Fitting Coefficients (Gravimetric, active).

| Parameter | C/20 | C/5 | C/2 | C | 2C | 5C | 10C |
|---|---|---|---|---|---|---|---|
| A | 158.7 | 150.7 | 140.3 | 126.1 | 88.6 | 27.1 | - 2.2 |
| B | 158.6 | 153.2 | 134.8 | 125.2 | 110.7 | 70.6 | - 0.5 |
| C | 274.9 | 276.0 | - 75.6 | 437.3 | 773.2 | 1299.8 | 441.3 |
| D | 138.1 | 133.2 | 119.3 | 141.3 | 114.4 | 50.6 | 11.1 |
| A:B | -14.7 | -12.4 | | | | | |
| A:C | | | 415.7 | | | | |
| A:D | 33.3 | 29.4 | 41.0 | | 61.9 | | |
| B:C | | -72.9 | 380.6 | | | | |
| B:D | 45.3 | 36.00 | 66.9 | | | | |
| C:D | -291.8 | -262.5 | | -690.6 | -1207.2 | | |
| λ | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 |

**Table S12.** Multi-variate Regression Fitting Coefficients (Gravimetric, active).

| Parameter | C/20 | C/5 | C/2 | C | 2C | 5C | 10C |
|---|---|---|---|---|---|---|---|
| A | 161 | 152.5 | 143.1 | 127.8 | 94 | 12.1 | -1.5 |
| B | 159.5 | 151.6 | 143.9 | 133.8 | 146.1 | 87.7 | 6.8 |
| C | 6540.8 | 5537.2 | 4804 | 221.6 | 16814.4 | 2093.1 | 69.6 |
| D | 138.4 | 132.6 | 125.2 | 124.9 | 94.9 | 23.4 | -36 |
| A:B | | | | | | 110.3 | |
| A:C | -6566.6 | -5497 | -4717.3 | | -16745 | | |
| A:D | 21.8 | 22.5 | 22.3 | 16.4 | 53.7 | | 66.7 |
| A:E | | -0.9 | -1.1 | | -23.9 | -30 | |
| A:F | | -1.3 | -1.7 | | -16.3 | -32.9 | |
| B:C | -6701.3 | -5644.8 | -4887.1 | | -18046 | -3026.4 | |
| B:D | 42 | 39.4 | 36.5 | | | | |
| B:E | | | | | 40 | | |
| B:F | | | | | | 71.1 | |
| C:D | -6510 | -5538.7 | -4802.5 | | -16635 | | 1788.4 |
| C:E | | | | | | | -417.4 |
| C:F | | | | | 520.8 | | |
| D:E | | | | | | | -21.3 |
| D:F | -2.8 | | | | | -109 | |
| E:F | | | | | | | |
| $E^2$ | | | | 2.5 | 11.9 | | 7 |
| $F^2$ | | | | | | | |
| λ | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 |

**Table S13.** Multi-variate Regression Fitting Coefficients (Gravimetric, coating).

| Parameter | C/20 | C/5 | C/2 | C | 2C | 5C | 10C |
|---|---|---|---|---|---|---|---|
| A | 154.8 | 148.0 | 137.8 | 123.9 | 87.1 | 27.1 | - 2.0 |
| B | 145.5 | 155.7 | 130.2 | 117.1 | 104.1 | 66.5 | - 0.6 |
| C | 291.5 | 96.6 | 323.4 | 435.0 | 756.2 | 1252.8 | 423.0 |
| D | 154.0 | 130.9 | 140.3 | 133.8 | 107.6 | 47.4 | 10.5 |
| A:B | | - 27.1 | | | | | |
| A:C | | 184.1 | | | | | |
| A:D | | 21.0 | | | 62.0 | | |
| B:C | | | | | | | |
| B:D | | | | | | | |
| C:D | -394.5 | | -488.3 | -714.0 | -1207.5 | | |
| λ | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 |

**Table S14.** Multi-variate Regression Fitting Coefficients (Gravimetric, coating).

| Parameter | C/20 | C/5 | C/2 | C | 2C | 5C | 10C |
|---|---|---|---|---|---|---|---|
| A | 158.2 | 149.1 | 139.8 | 125.1 | 92.1 | 12.4 | -1.4 |
| B | 148.2 | 146.1 | 142.4 | 124.5 | 138.1 | 82.9 | 6.5 |
| C | 3990.5 | 125.8 | 135.8 | 279 | 15075.5 | 2029.5 | 66.8 |
| D | 131.3 | 128.2 | 124 | 119.2 | 88.5 | 21.4 | -35.1 |
| A:B | | -8.6 | -13.3 | | | 108.5 | |
| A:C | -3964.3 | 92.5 | 77.6 | | -14952 | | |
| A:D | 22.1 | 20.5 | 18.2 | 18.3 | 55.1 | | 65.4 |
| A:E | | | | -2 | -23.1 | -29.1 | |
| A:F | | | | | -15.7 | -32.8 | |
| B:C | -3969.4 | | | | -16216 | -2974.3 | |
| B:D | 33.8 | 18.8 | | | | | |
| B:E | -4 | -2.2 | | | 38.3 | | |
| B:F | -2 | | | | | 69.2 | |
| C:D | -3914.8 | | | -230.7 | -14899 | | 1711.9 |
| C:E | 35.6 | | | | | | -393.6 |
| C:F | | | | | 516 | | |
| D:E | | | -2.7 | | | | -21 |
| D:F | | | | | | -103.7 | |
| E:F | | | | | | | |
| E² | | | 0.8 | 2.5 | 11.6 | | 6.7 |
| F² | | | | | | | |
| λ | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 |

**Table S15.** Multi-objective Optimisation Electrode Formulations.

| Title | LFP / wt% | PVDF / wt% | KS6L / wt% | SWCNT / wt% |
|---|---|---|---|---|
| Mixture only, 2C discharge, mA hr $g_{coat}$ | 95.8 | 1.2 | 2.8 | 0.20 |
| Mixture + Process, 5C discharge, mA hr $g_{coat}$ | 96.7 | 1.1 | 2.0 | 0.20 |
| Mixture + Process, 5C discharge, mA hr | 94.9 | 2.1 | 2.8 | 0.20 |
| Mix04, experimental | 94.7 | 2.3 | 2.8 | 0.20 |
| Mix11, experimental | 94.9 | 3.0 | 1.9 | 0.16 |
| Multi-objective optimisation model | 95.34 | 2.85 | 1.63 | 0.18 |

**Table S16.** Multi-objective Optimisation Desirability Scores.

| Title | Discharge 5 C | Adhesion (cal) | $R_{thru}$ (total) | ASI (disch) | Weighted Average |
|---|---|---|---|---|---|
| Priority weighting | (3) | (2) | (2) | (1) | N / A |
| Mixture, 2C discharge, mA hr $g_{coat}$ | 0.86 | 0.00 | 0.51 | 0.47 | 0.00 |
| M + P, 5C discharge, mA hr $g_{coat}$ | 0.89 | 0.09 | 0.30 | 0.51 | 0.35 |
| M + P, 5C discharge, mA hr | 0.78 | 0.26 | 0.83 | 0.54 | 0.57 |
| Mix04, experimental | 0.89 | 0.17 | 0.92 | 0.60 | 0.57 |
| Mix11, experimental | 0.80 | 0.73 | 0.78 | 0.53 | 0.74 |
| Mix04, modelled responses | 0.76 | 0.32 | 0.90 | 0.56 | 0.62 |
| Mix11, modelled responses | 0.69 | 0.76 | 0.74 | 0.59 | 0.71 |
| Multi-objective optimisation model | 0.78 | 0.78 | 0.68 | 0.70 | 0.74 |

The total desirability D is calculated from the weighted geometric mean of the individual desirability factors $d_i$, using the equations [36] :-

$$d_i(Y_i) = \begin{cases} 0 & if\ Y_i \leq Y_{i_*} \\ \left(\frac{Y-Y_{i_*}}{Y_i^*-Y_{i_*}}\right) & if\ Y_{i*} < Y_i < Y_i^* \\ 1 & if\ Y_i^* \leq Y_i \end{cases}, \qquad D = \sqrt[\Sigma k_j]{\prod_{j=1}^n d_j^{k_j}} \qquad (S5)$$

In this equation, i is a formulation in the range 1 to n, $Y_i$ is the modelled value for a formulation's response, $Y_{i_*}$ and $Y_i^*$ are the lower acceptable and target bounds of the response, $k_j$ is the priority weighting for response j. Thus, for the multi-objective optimisation model :-

D  =  $[ (0.78)^3 \times (0.78)^2 \times (0.68)^2 \times (0.70)^1 ]^{1/8}$  =  0.743
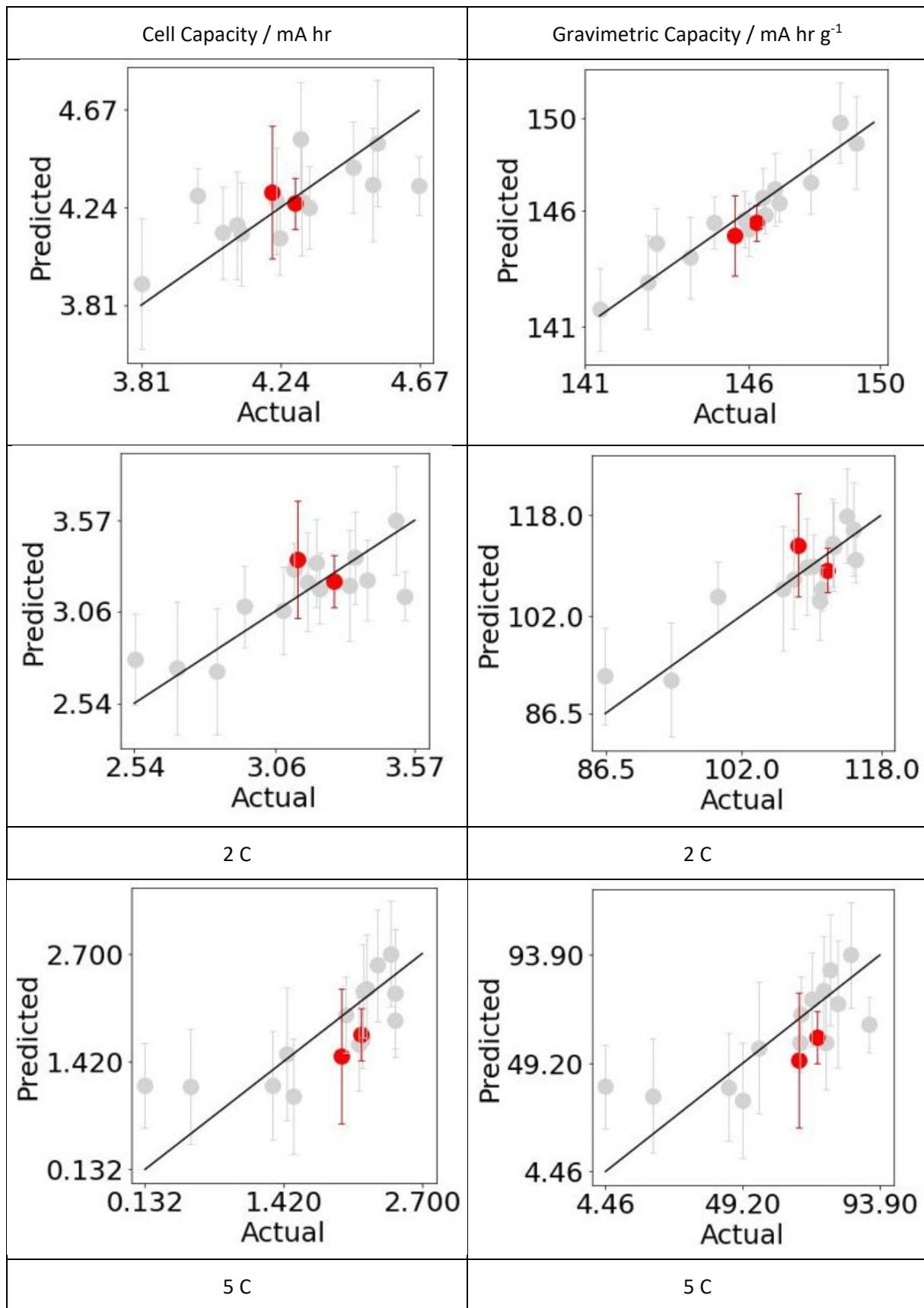
**Figure S10.** Goodness Of Fit Plots (Mixture Only).
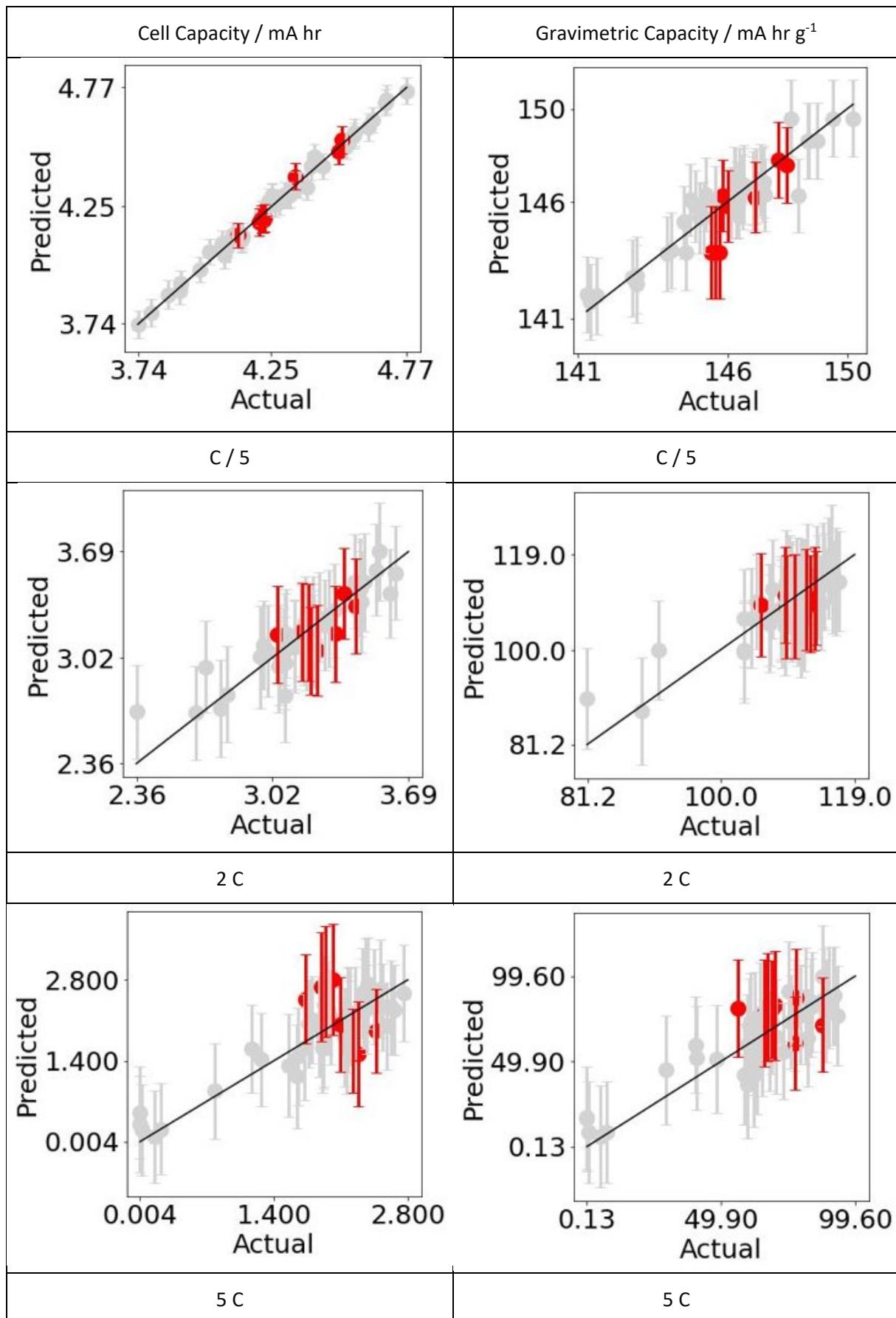
**Figure S11.** Goodness Of Fit Plots (Mixture + Process).
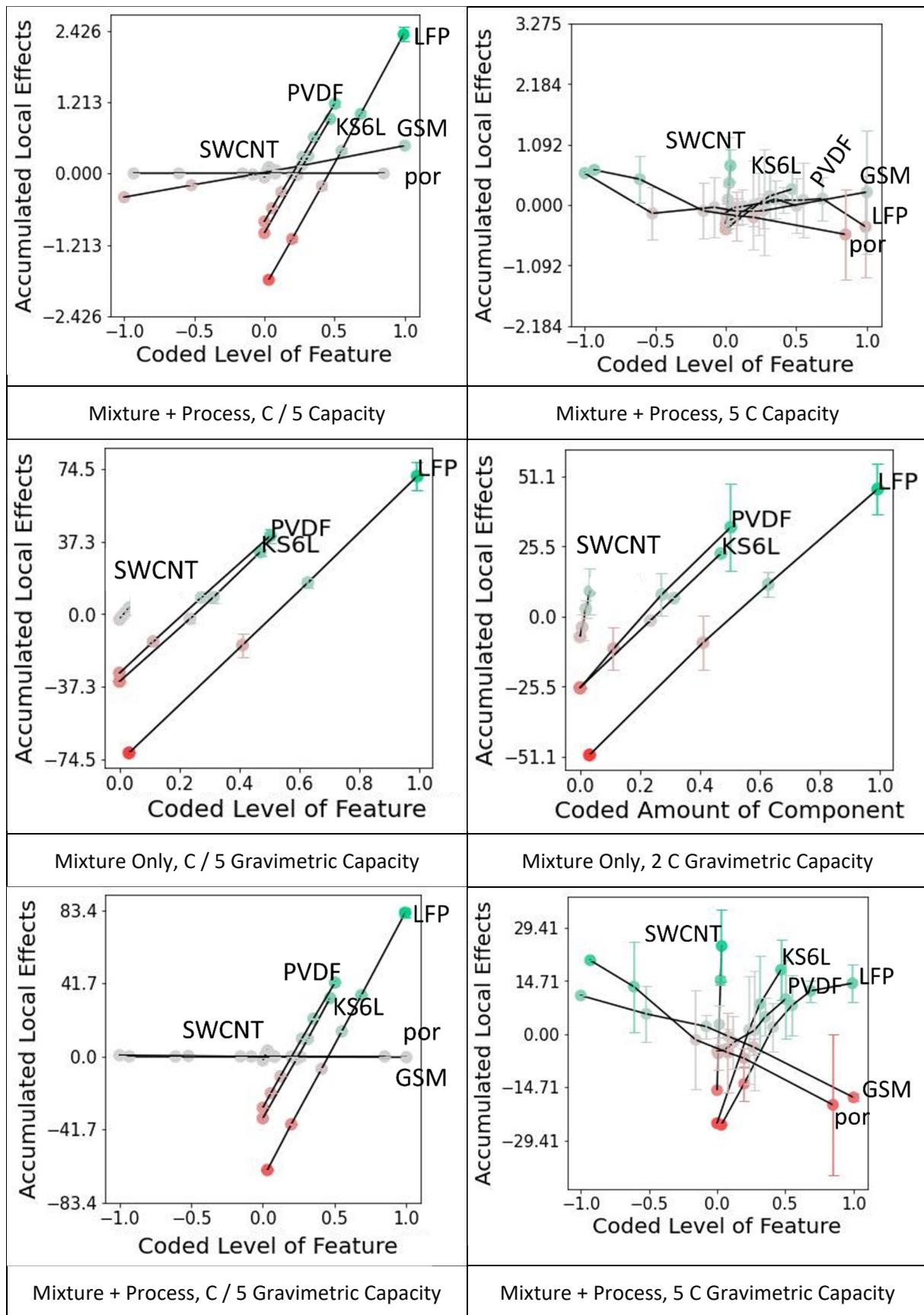
**Figure S12.** 1D ALE Plots On Capacity.

| | Mixture + Process, C / 5 Capacity | Mixture + Process, 5 C Capacity |

| Mixture Only, C / 5 Gravimetric Capacity | Mixture Only, 2 C Gravimetric Capacity |

| Mixture + Process, C / 5 Gravimetric Capacity | Mixture + Process, 5 C Gravimetric Capacity |
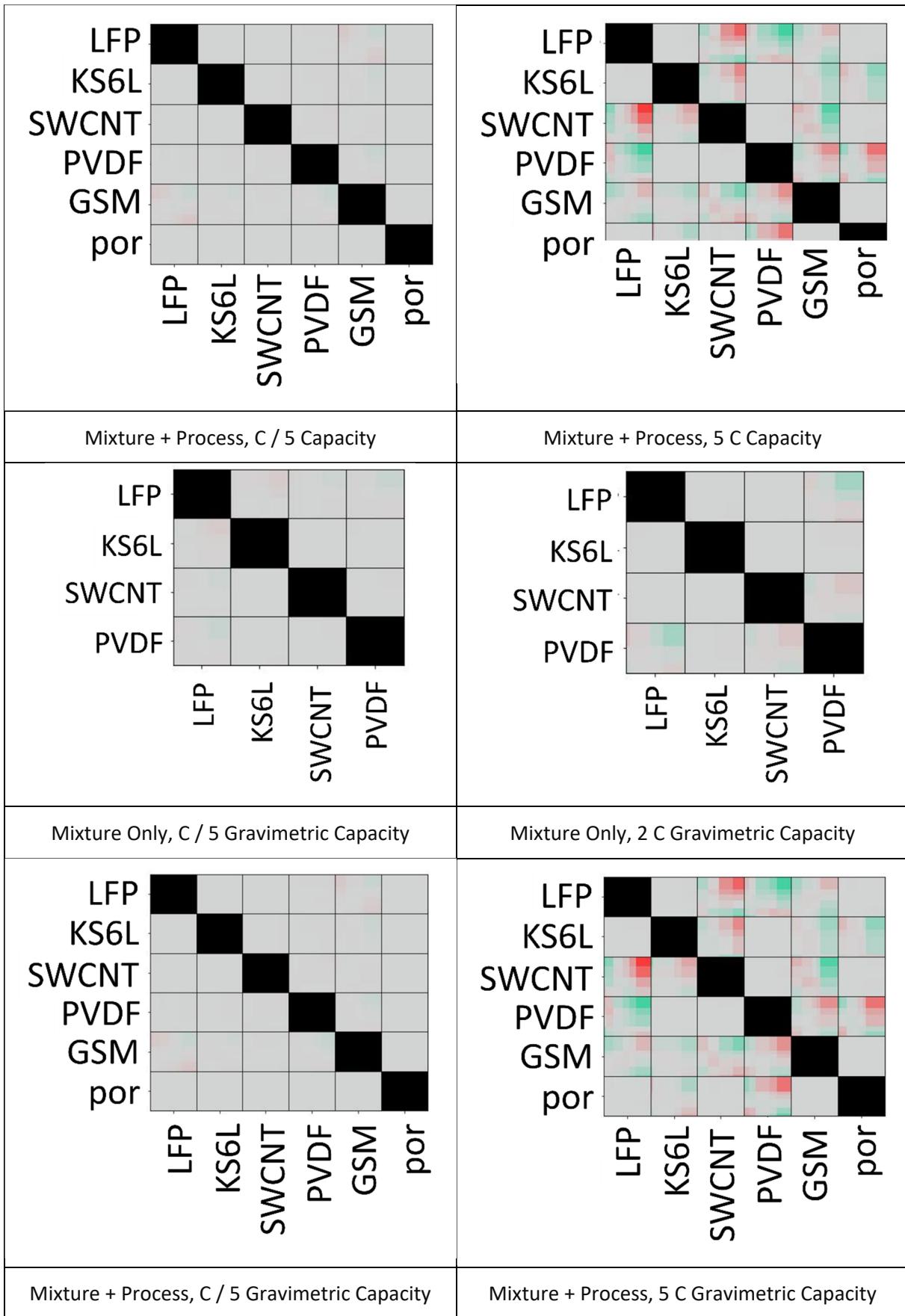
**Figure S13.** 2D ALE Plots On Capacity.
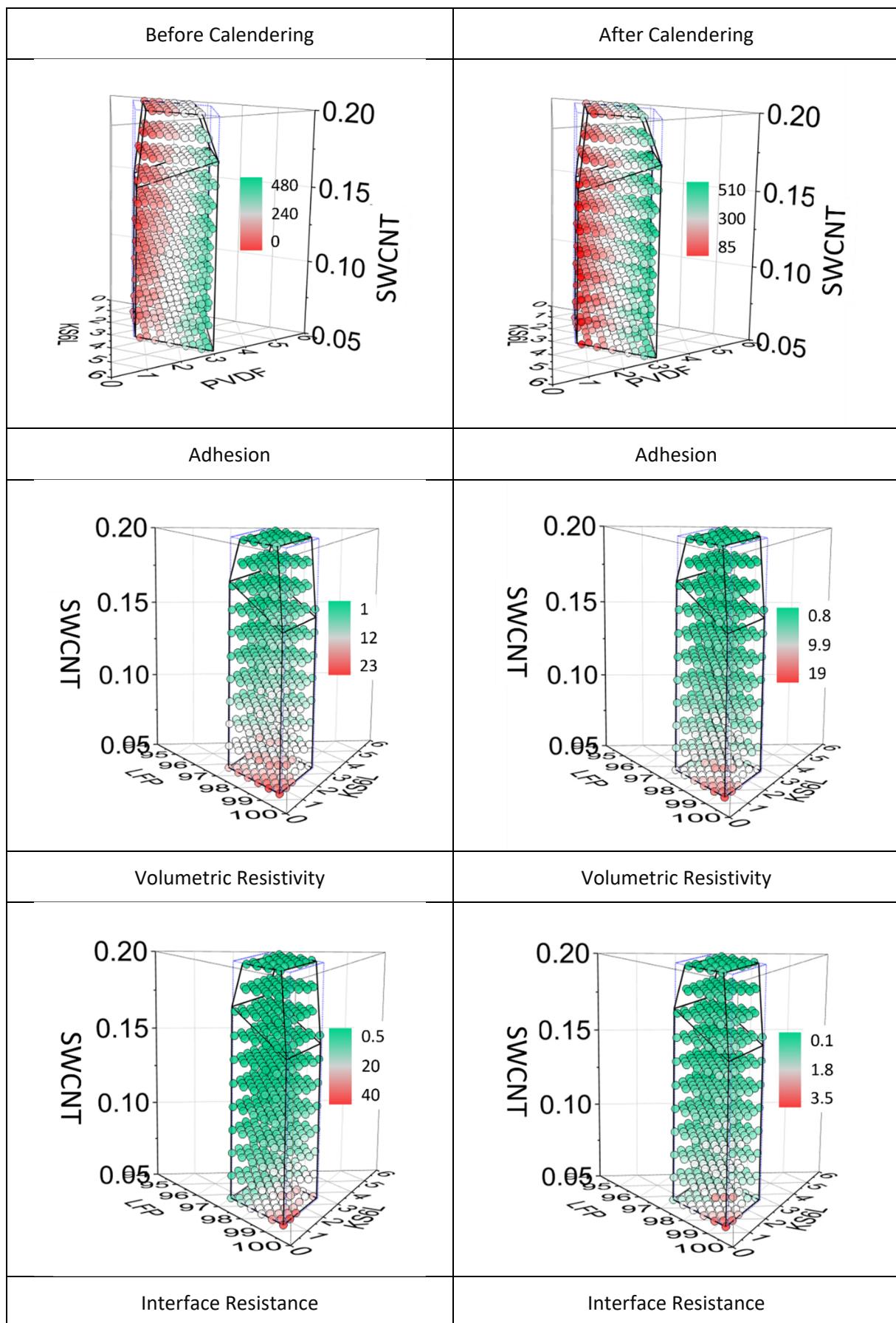
**Figure S14.** Model Predictions For Adhesion And Resistances.

| | Before Calendering | After Calendering |
|---|---|---|
| | 1D | 1D |
| | 2D | 2D |
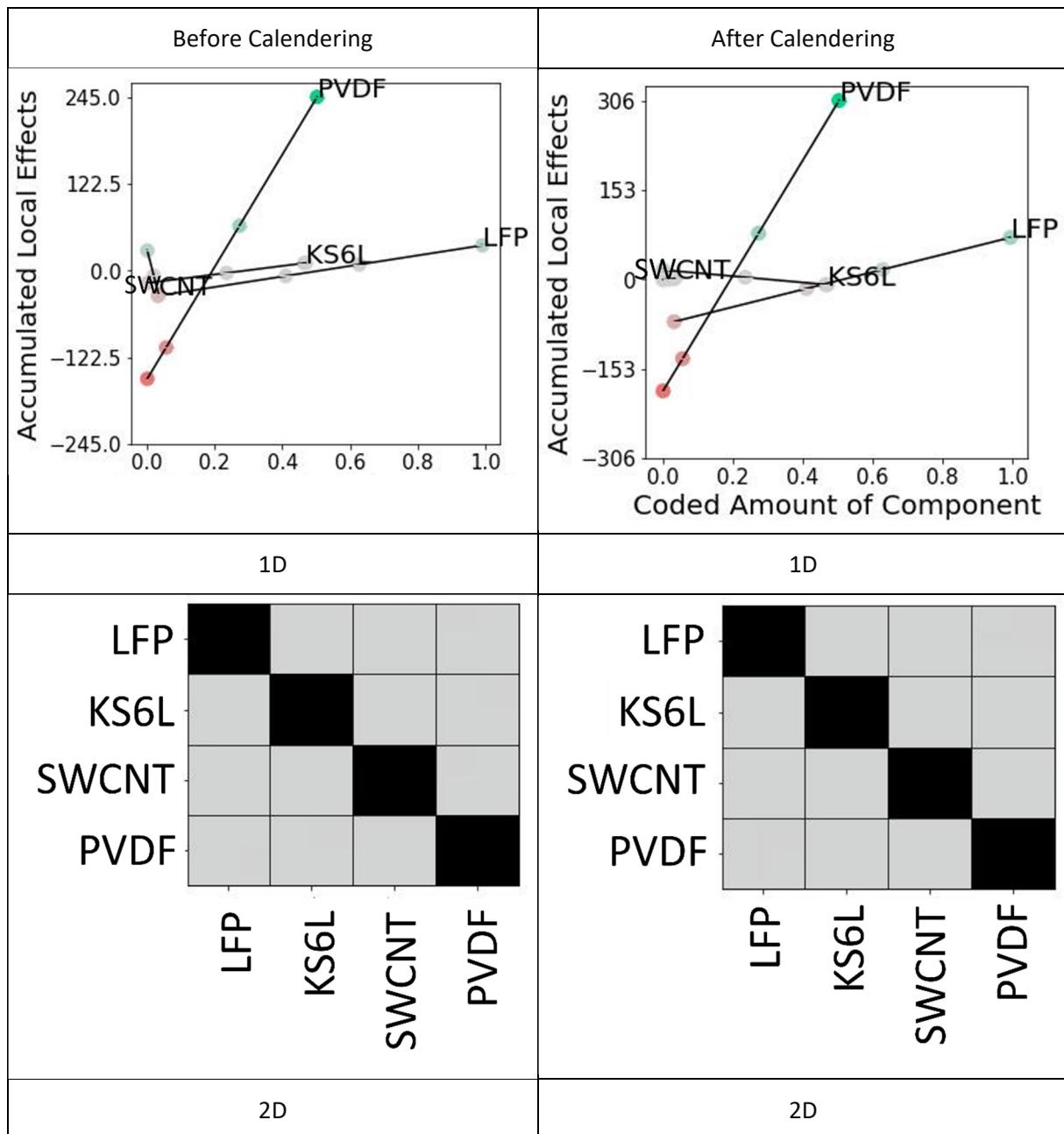
**Figure S15.** 1D and 2D ALE Plots For Adhesion.