

Supplementary Information

Table S1. Cell Properties Comparison

Property	Coin Cell	Single-Layer Pouch	Multi-layer Pouch
Cathodes / mm	φ14.8	68 x 48	68 x 48
Cathodes / cm ²	1.72	33.2	600
Capacity / A hr	0.0045	0.080	1.40
Electrolyte / g	0.10	0.65	7.0
Electrolyte / g (A hr) ⁻¹	22.2	8.1	5.0

Table S2. Averaged cell results

Coating	Cell Type	Coating / gsm	Porosity / %	0.2C / mA hr g ⁻¹	FCL / %	5C / 0.2C / %	Min ASId / S cm ⁻²
MIX18	Coin	170.4	38.0	152.5	1.8	61.2	0.051
MIX19D1	Coin	165.2	40.6	152.5	1.7	62.2	0.052
MIX20D2	Coin	169.5	38.0	152.5	1.8	59.6	0.048
MIX20D3	Coin	170.6	36.8	153.3	1.8	61.5	0.052
MIX20D2	Coin	170.4	37.0	142.6	10.3	45.3	0.042
MIX20D3	Coin	170.7	37.3	141.8	10.4	48.9	0.046
MIX20D2	SLP	165.3	38.8	144.7	8.2	48.1	0.051
MIX20D3	SLP	163.8	40.1	144.3	8.2	48.8	0.050
MIX20D3	MLP	164.1	N / A	145.3	8.4	63.8	0.039

TableS3. Summary of resistance measurements

Mix I.D.	Drying Conditions	Coating Thickness / um		Total Through-Plane / Ω cm ²	
		Uncalendered	Cal485	Uncalendered	Cal485
MIX18	Lab	142	80	1.30	0.13
MIX19	D1	128	88	0.76	0.07
MIX20	D2	132	85	2.45	0.12
MIX20	D3	133	76	2.44	0.10
MIX20 (A)	D3	139	77	2.69	0.27
MIX20 (B)	D3	139	77	2.54	0.28

NB. Coating thicknesses of double-sided coating assumed to be symmetrical (cal. and uncal.).

Table S4. Averaged resistivity measurements (uncalendered)

Mix I.D.	Drying Conditions	Volumetric Resistivity / Ω cm		Interface Resistance / Ω cm ²	
		Mean	S.D.	Mean	S.D.
MIX18	Lab	5.36	0.20	1.22	0.114
MIX19	D1	4.97	0.04	0.70	0.037
MIX20	D2	7.71	0.10	2.35	0.295
MIX20	D3	7.03	0.09	2.34	0.270
MIX20 (A)	D3	6.90	0.31	2.59	0.208
MIX20 (B)	D3	7.13	0.19	2.44	0.228

Table S5. Averaged resistivity measurements (calendered, 500 μ m)

Mix I.D.	Drying Conditions	Volumetric Resistivity / Ω cm		Interface Resistance / Ω cm ²	
		Mean	S.D.	Mean	S.D.
MIX19	D1	3.53	0.06	0.06	0.002
MIX20	D2	6.01	0.10	0.17	0.013
MIX20	D3	5.80	0.13	0.13	0.013

Table S6. Averaged resistivity measurements (calendered, 495 μ m)

Mix I.D.	Drying Conditions	Volumetric Resistivity / Ω cm		Interface Resistance / Ω cm ²	
		Mean	S.D.	Mean	S.D.
MIX19	D1	3.58	0.11	0.04	0.003
MIX20	D2	5.62	0.23	0.14	0.016
MIX20	D3	5.48	0.13	0.11	0.009

Table S7. Averaged resistivity measurements (calendered, 485 μ m or “Zero Gap”)

Mix I.D.	Drying Conditions	Volumetric Resistivity / Ω cm		Interface Resistance / Ω cm ²	
		Mean	S.D.	Mean	S.D.
MIX18	Lab	3.90	0.34	0.10	0.013
MIX19	D1	3.84	0.21	0.03	0.002
MIX20	D2	4.80	0.10	0.08	0.006
MIX20	D3	5.01	0.11	0.07	0.003
MIX20 (A)	D3	4.98	0.10	0.23	0.024
MIX20 (B)	D3	4.26	0.10	0.25	0.031

Table S8. Model input parameters

Code	Feature	Actual		Coded		Mix18	
		Min.	Max.	Low	High	Actual	Coded
A	LFP	94.03	98.50	0.00	1	97.03	0.671
B	KS6L	0.70	2.80	0.00	0.480	0.87	0.038
C	SWCNT	0.05	0.20	0.00	0.034	0.16	0.025
D	PVDF	0.75	3.00	0.00	0.503	1.94	0.266
E	Coat weight	152.0	188.5	- 1.00	1.00	170	-0.014
F	Porosity	36.5	43.5	- 1.00	1.00	38.2	-0.514

Table S9. Output ranges and desirability scores

Parameter		Minimum	Maximum
5 C discharge capacity	/ mA hr g _{coat} ⁻¹	0	100
Adhesion	/ kPa	150	600
Through plane resistance	/ S cm ⁻²	0	6
Area specific impedance	/ S cm ⁻²	0.02	0.06

Table S10. Multi-linear regression coefficients

Coeff.	5C / mA hr g _{coat} ⁻¹	Adhesion / kPa	Vol. conductivity / S cm ⁻¹	ASI (min) / Ω cm ²
A	12.4	149.9	0.41	69.3
B	82.9	-54.0	1.10	-40.3
C	2029.5	65.1	-25.35	-30168.3
D	21.4	988.2	0.91	98.8
A:B	108.5			
A:C				30814.3
A:D				-141.1
A:E	-29.1			37.7
A:F	-32.8			17.4
B:C	-2974.3		130.52	33054.2
B:D				
B:E				-81.2
B:F	69.2			
C:D			256.76	28824.5
C:E				
C:F				-863.3
D:E				
D:F	-103.7			32.6
E:F				
E ²				-24.2
F ²				

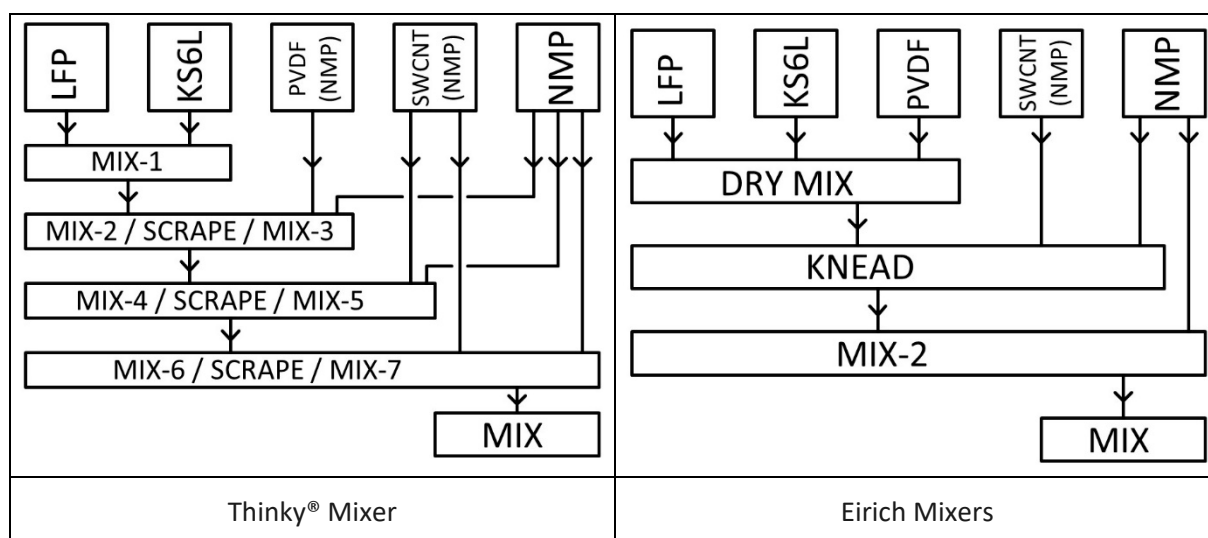


Figure S1. Mixing protocols in Thinky® and Eirich mixers

Thinky® mixing steps	MIX-1	1300 rpm, 5 minutes	Others	2000 rpm, 5 minutes
Thinky® solids contents	MIX-2	66.8 wt%	MIX-4	57.5 wt%
Eirich solids contents	Knead	~ 67 wt%	MIX-2	~ 52 wt%

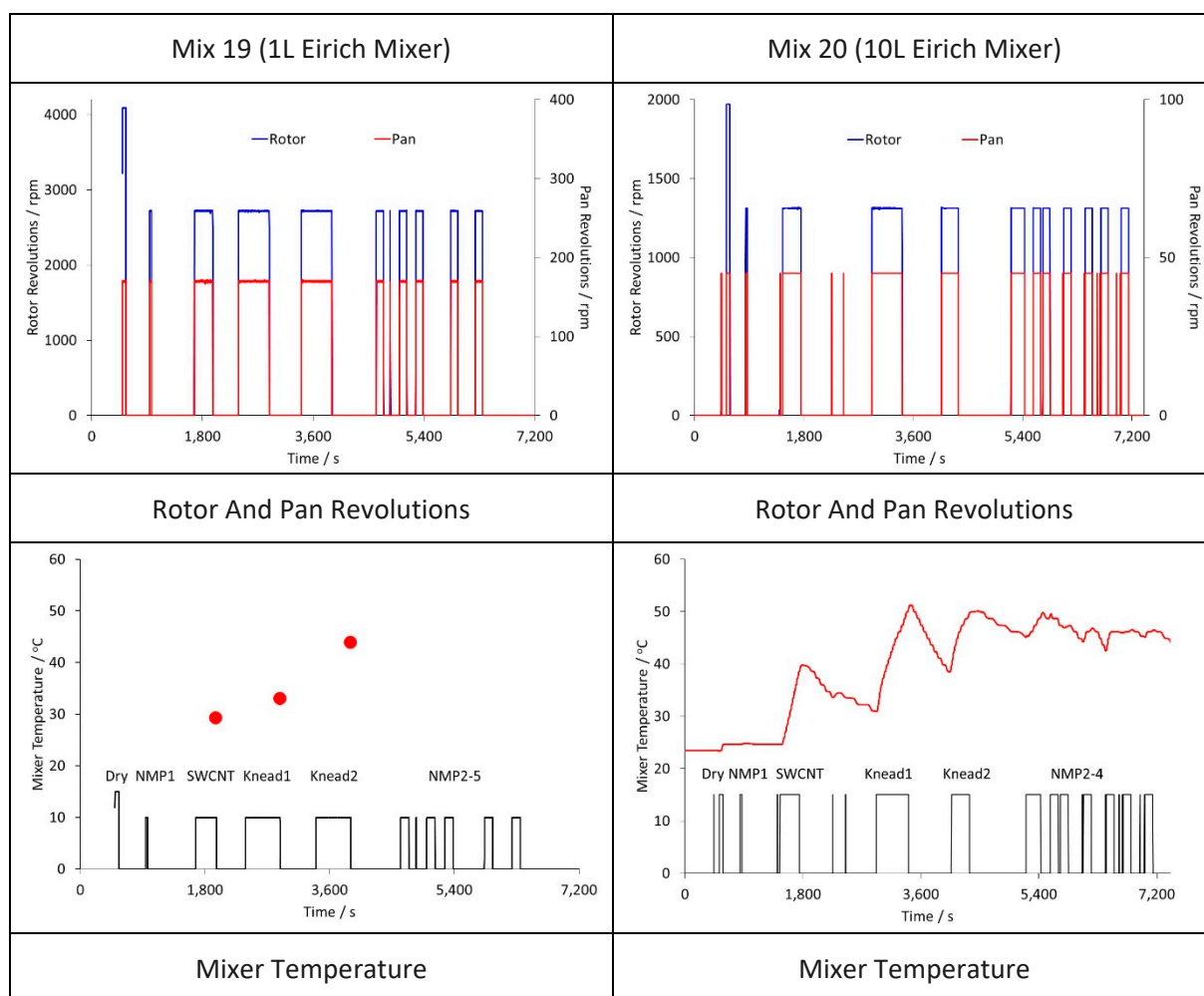


Figure S2. Data recorded during Eirich mixing processes

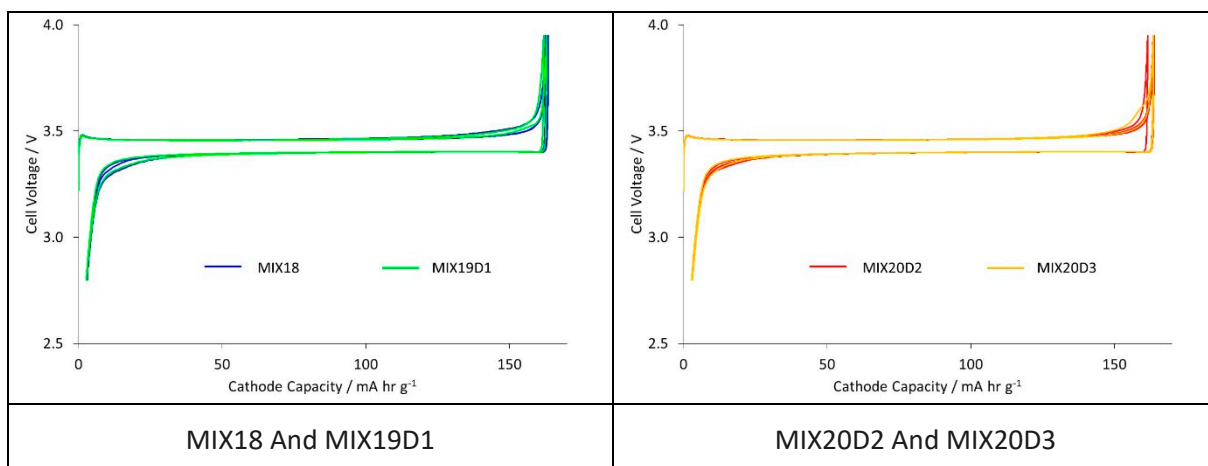


Figure S3. Half-cell coin cell voltages during formation cycle

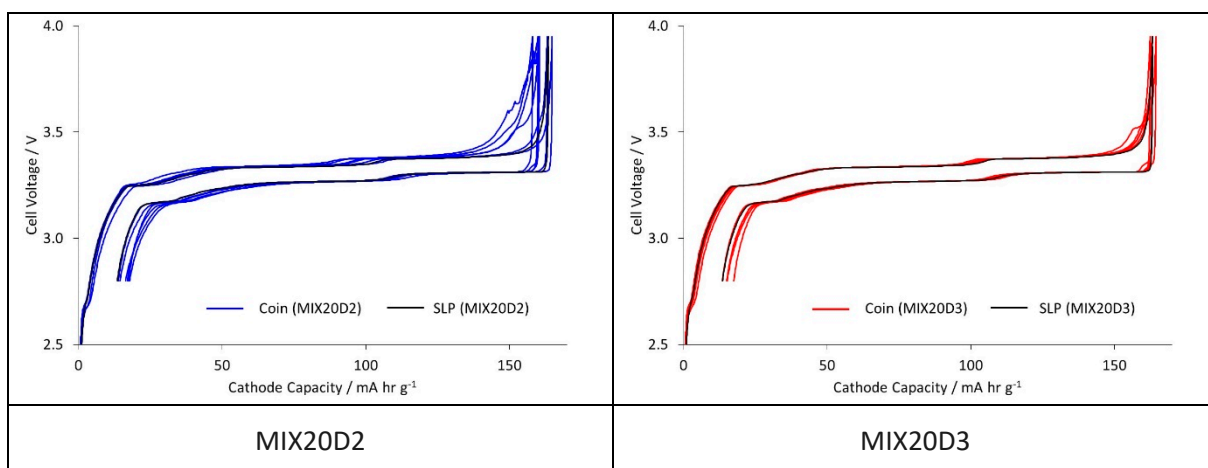


Figure S4. Full-cell coin cell voltages during formation cycle

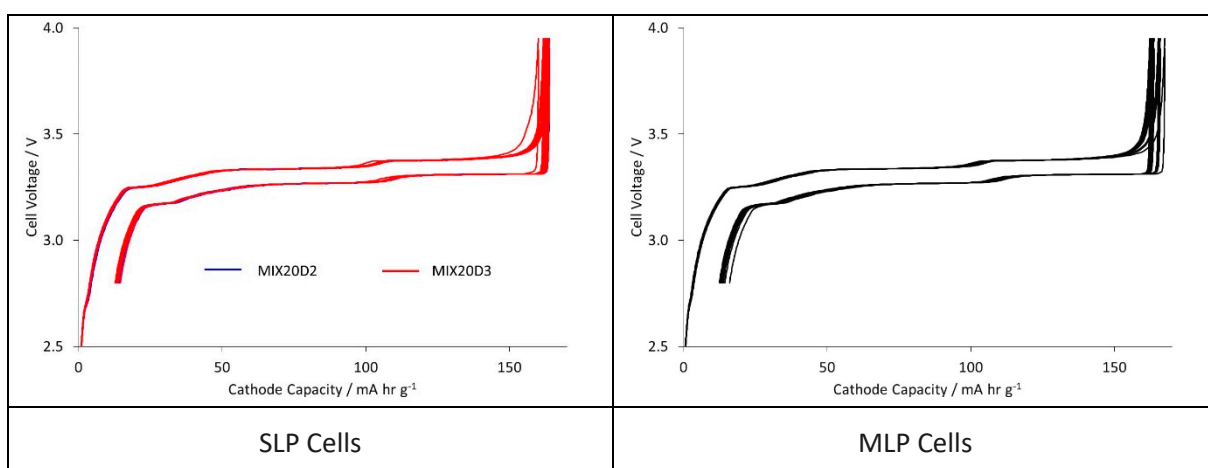


Figure S5. SLP and MLP cell voltages during formation cycle

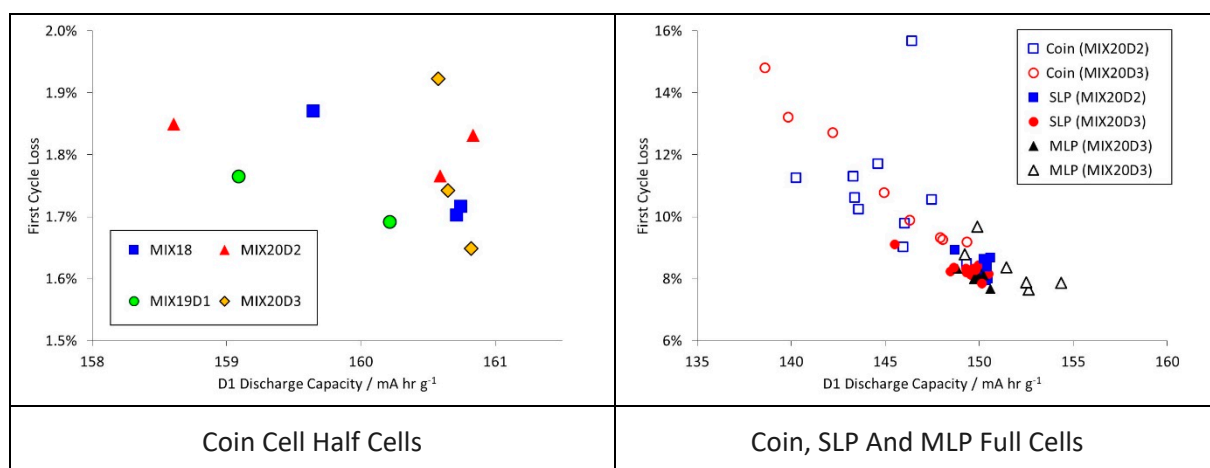


Figure S6. Results from formation cycles first cycle loss

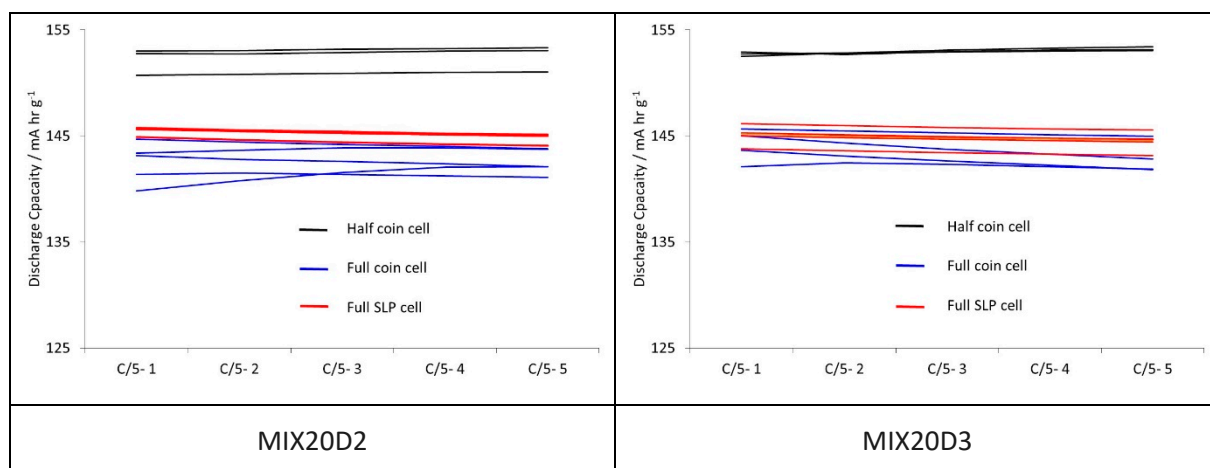


Figure S7. Coin capacities during conditioning cycles before rate tests

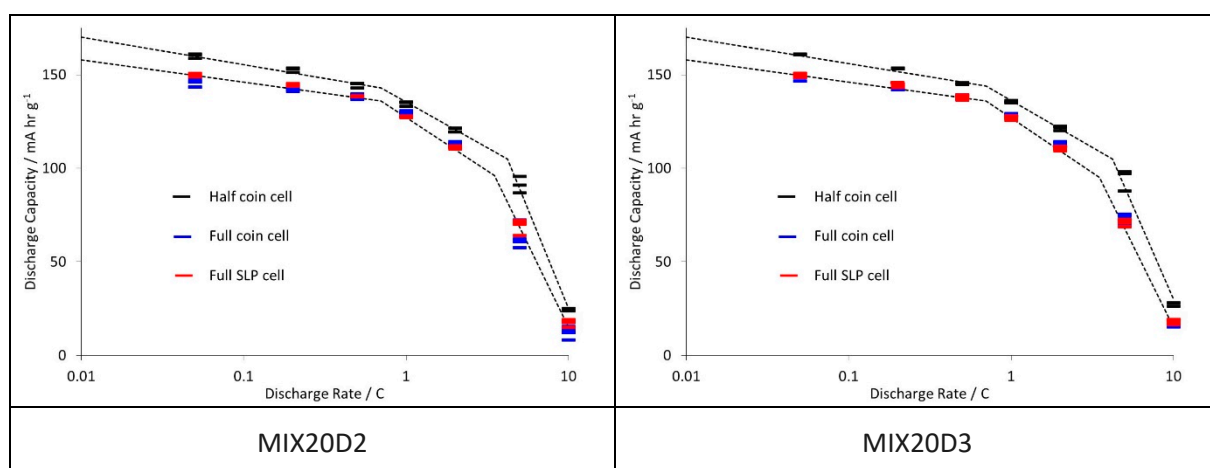


Figure S8. Comparison of rate test capacities in different cell formats

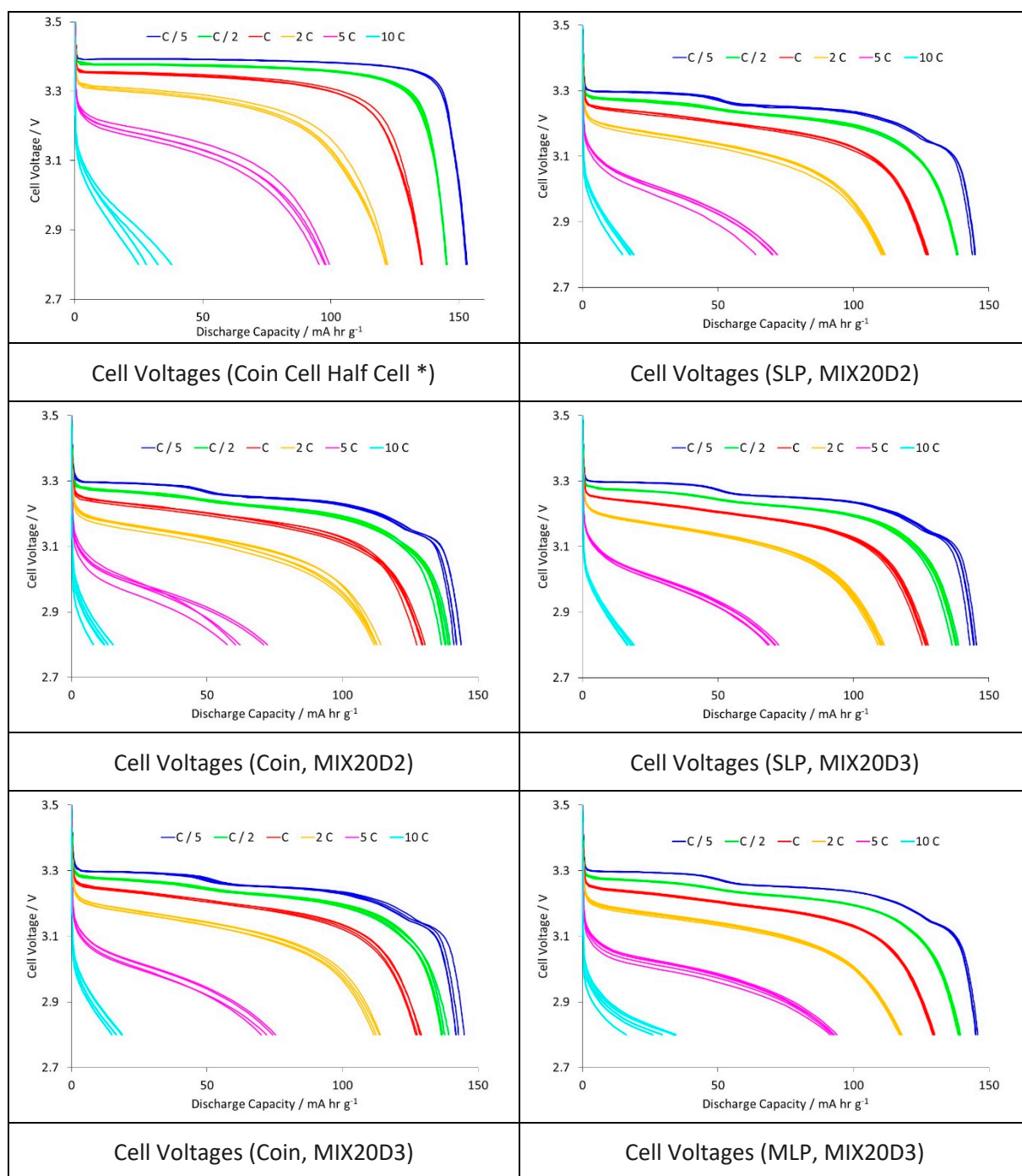


Figure S9. Cell voltages during rate tests

* For the half-cell coin cells, the values shown are for the best cell from each coating.

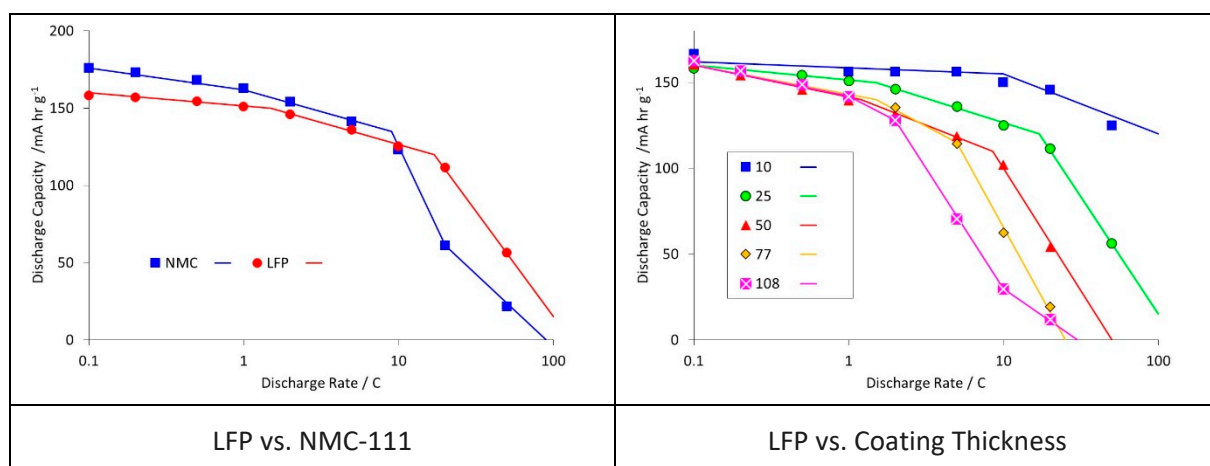


Figure S10. Published rate test results (Electrochim. Acta **71** (2012) p. 258)

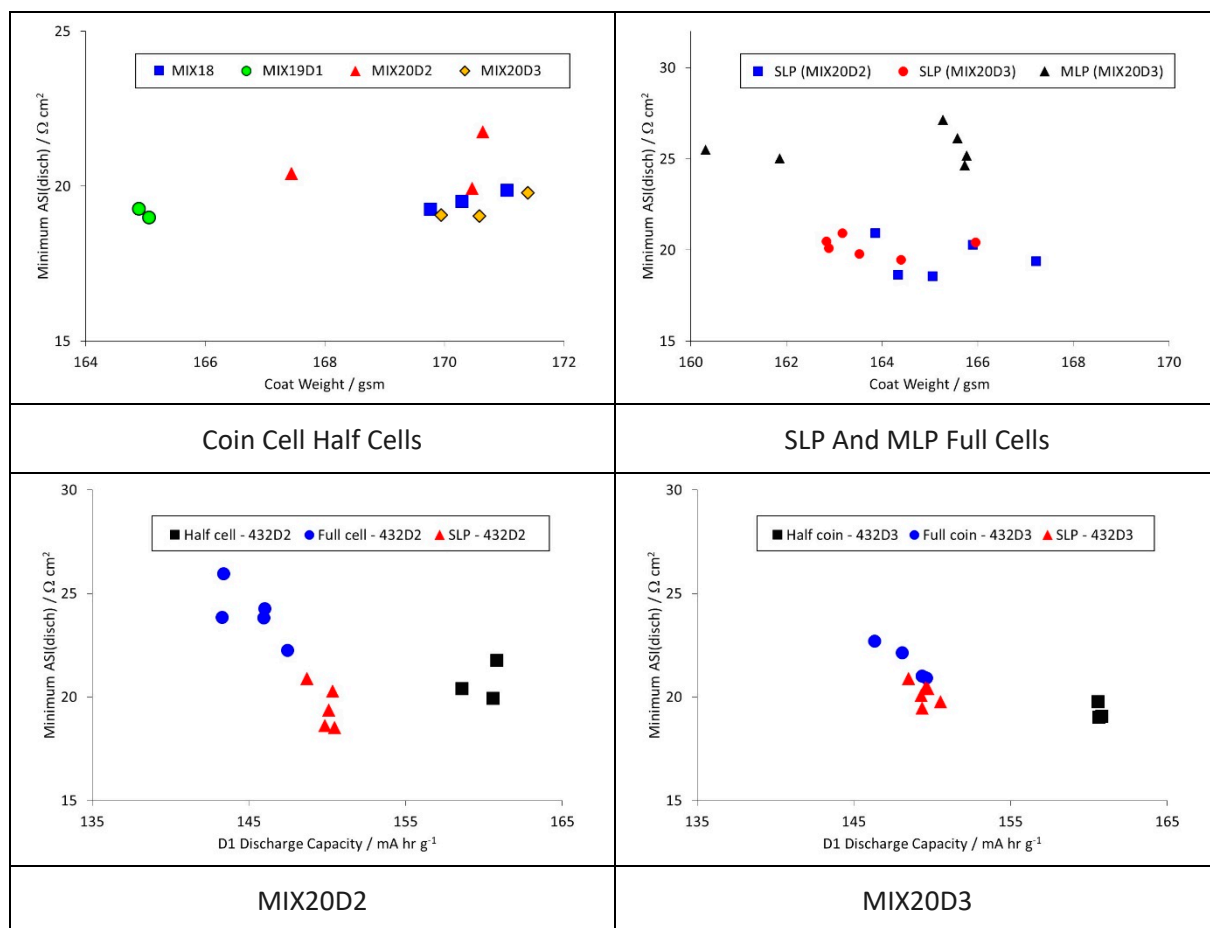


Figure S11. ASI measurements during discharge pulses

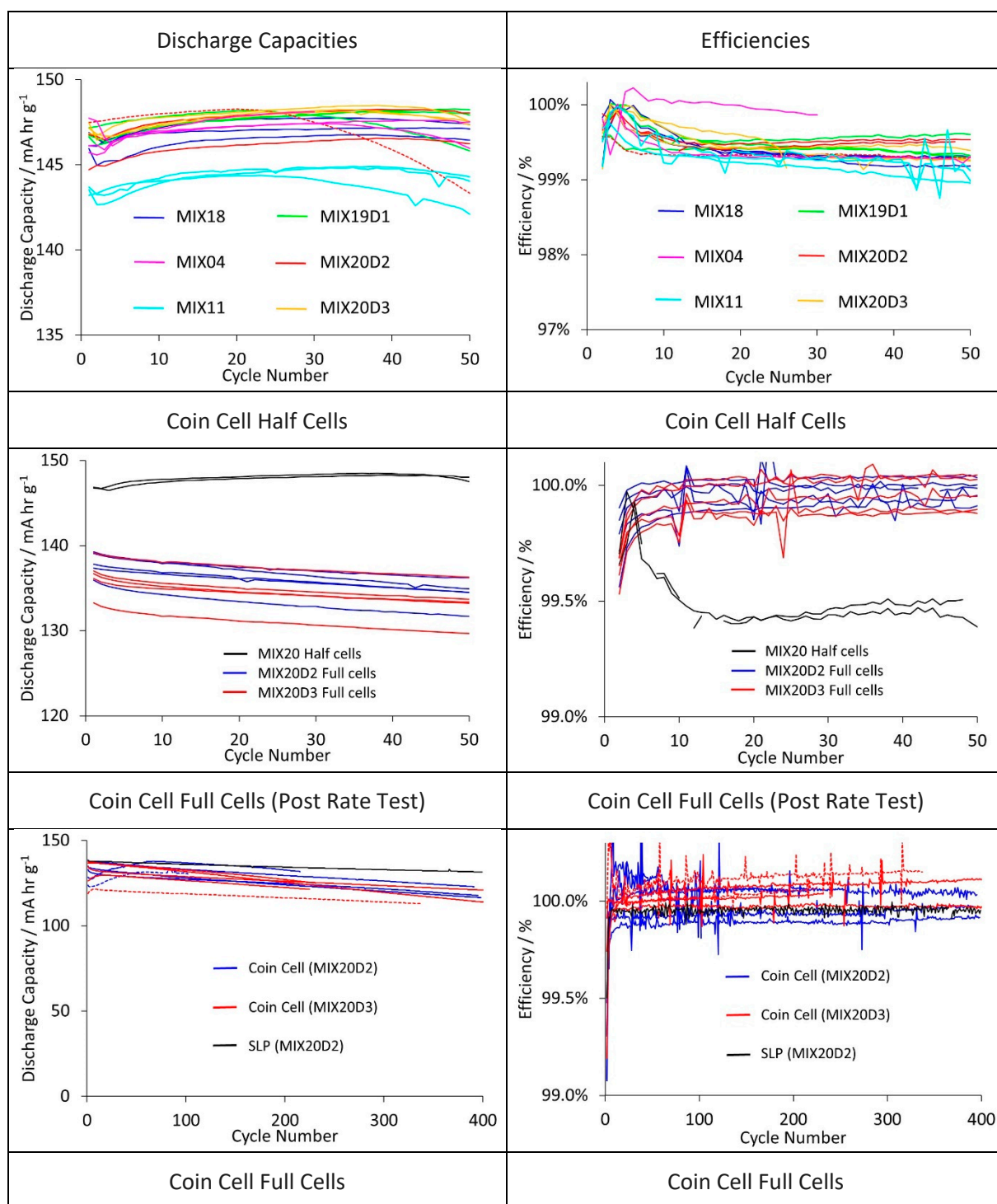


Figure S12. Discharge capacities and efficiencies during coin cell cycling tests
(One SLP full cell is included, for comparative purposes)

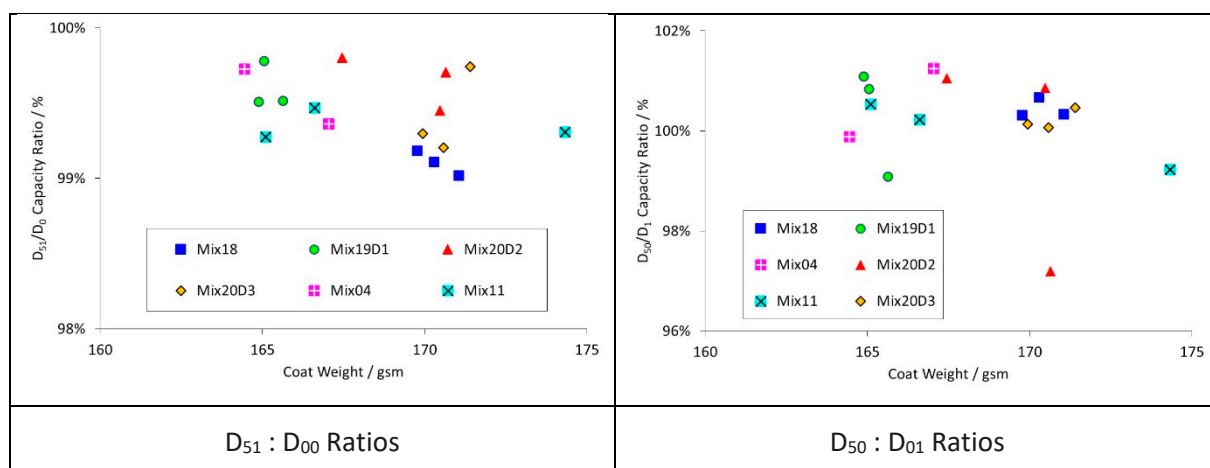


Figure S13. Capacity retention values for half-cell coin cell cycling tests

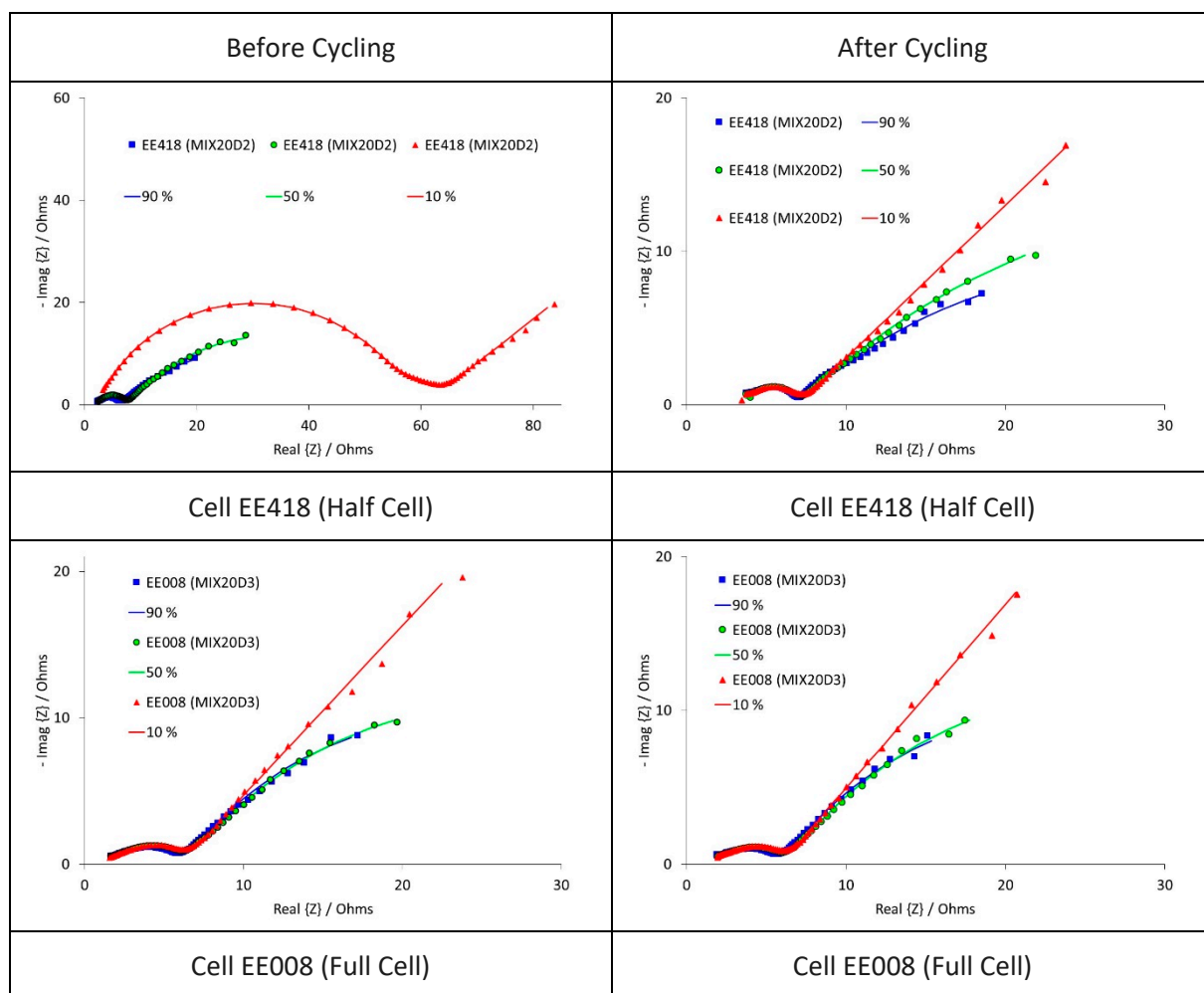


Figure S14. Impedance spectra for coin cells

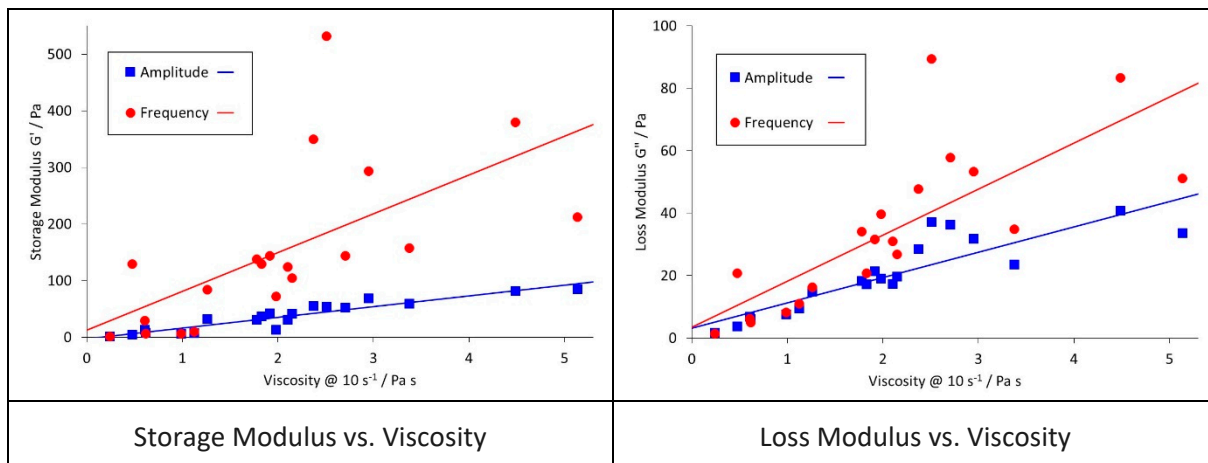


Figure S15. Viscosity measurement results (I)
(Amplitude: G' and G'' at 10 – 11 % strain, Frequency: G' and G'' at 10 rad s^{-1} , 0.1 % strain)

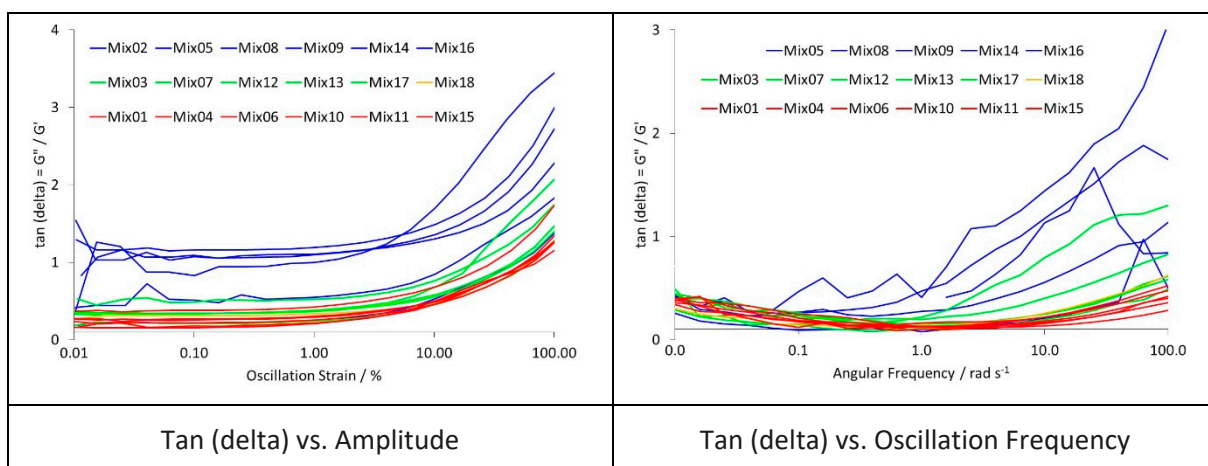


Figure S16. Viscosity measurement results (II)

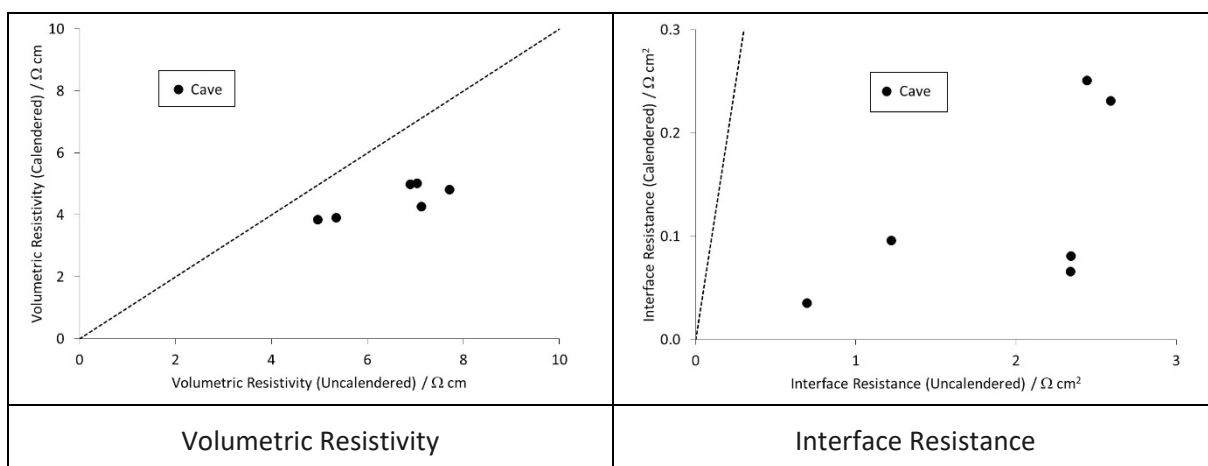


Figure S17. Comparison of resistance values before and after calendaring

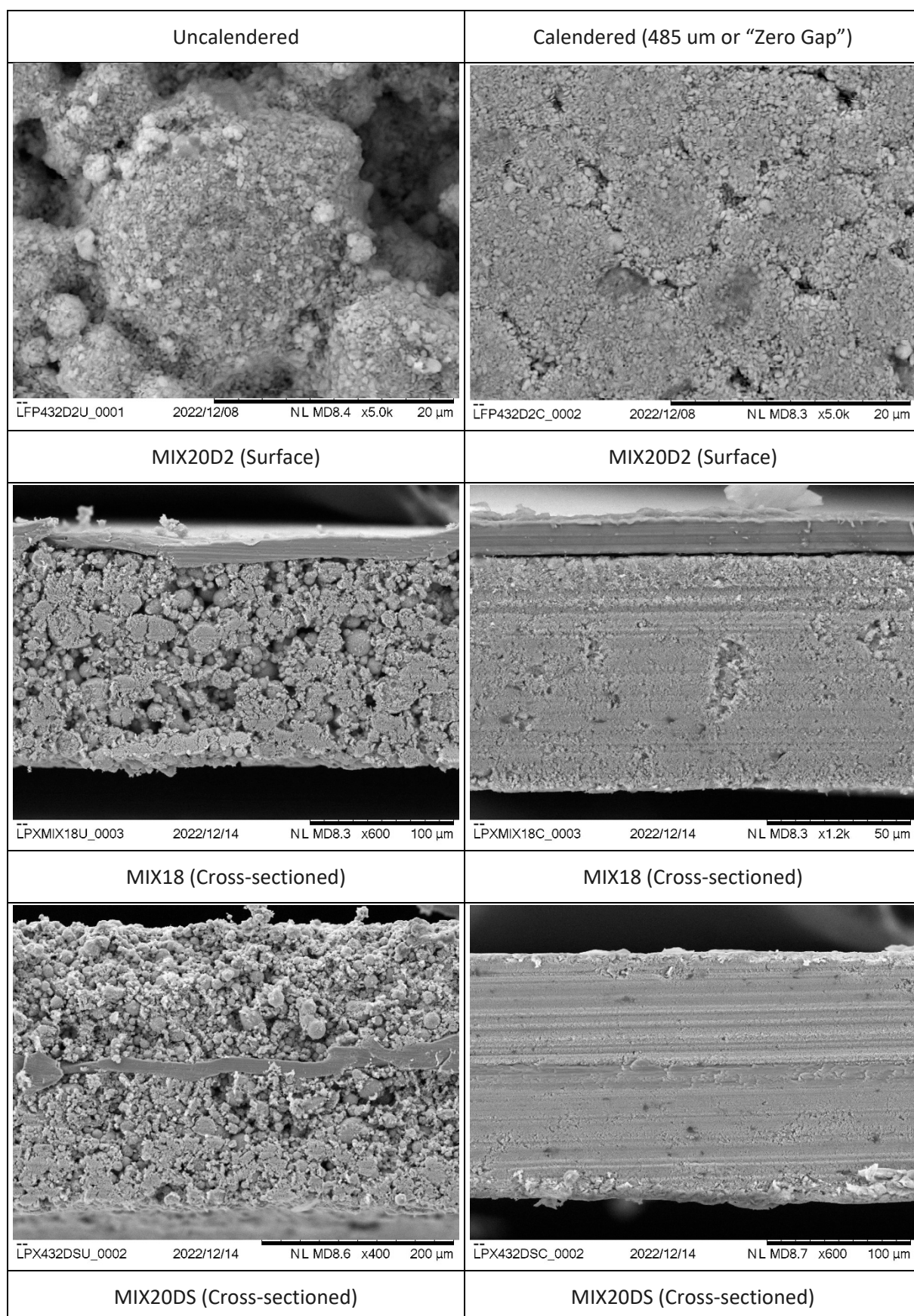


Figure S18. Selected SEM images of coatings