Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A

Supporting Information

Octahedral and truncated high-voltage spinel cathodes: the role of morphology and surface planes on electrochemical properties

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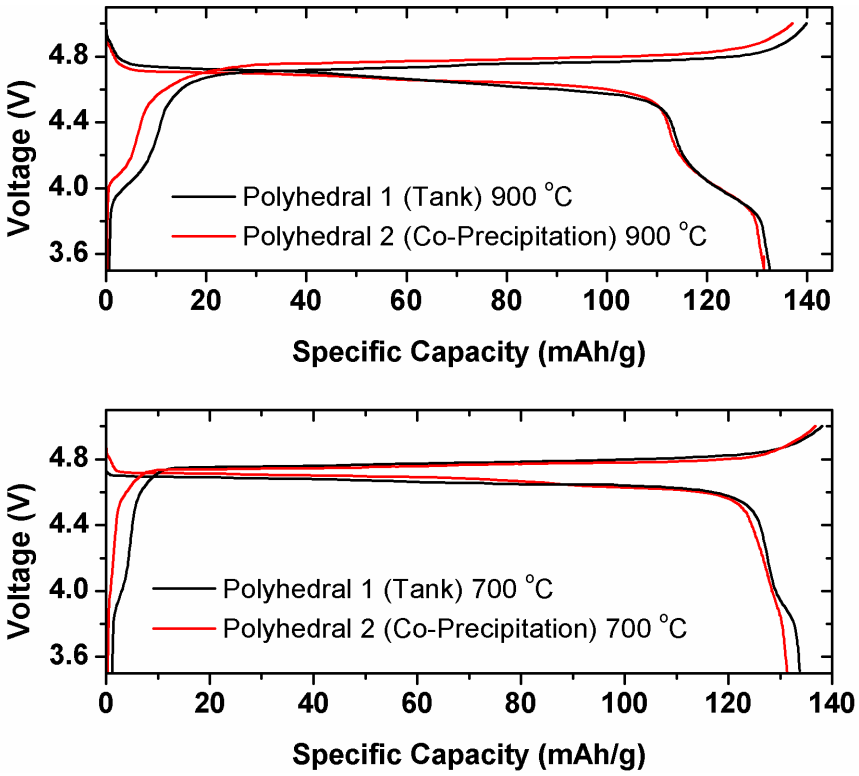

Figure S1: First-charge/discharge profiles of Polyhedral 1 and Polyhedral 2 samples before and after post-annealing process at 700 °C, demonstrating the elimination of Mn^{3+} after the annealing process.

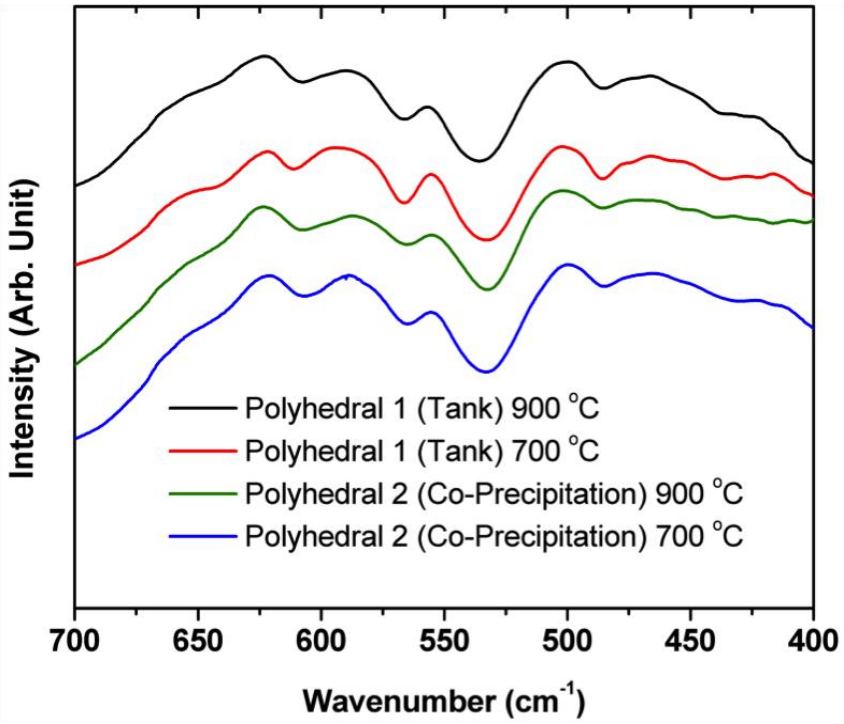

Figure S2: FTIR data indicating an increase in the degree of cation ordering after annealing at 700 °C.

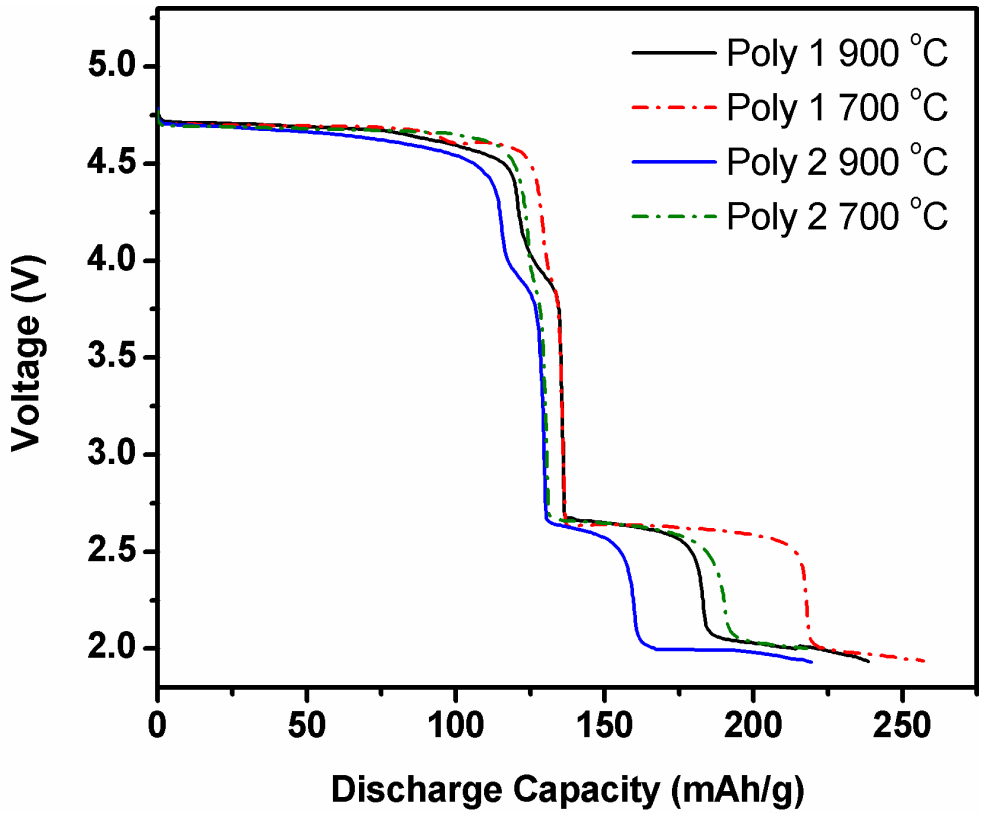

Figure S3: Deep discharge to 2.0 V to compare the degree of cation ordering before and after annealing at 700 °C, where longer plateaus at ~ 2.7 V correspond to higher degree of cation ordering.

Table S1: First discharge capacity values at different voltage regions

Sample	Discharge Capacity (mAh/g)				
	Total	Capacity Above 3 V	Capacity Below 3 V		
			Total	~ 2.7 V	~2.1 V
Poly 1 900 ^{o}C	239	136	103	46	57
Poly 1 700 ^{o}C	252	136	116	81	35
Poly 2 900 ^{o}C	216	130	86	31	55
Poly 2 700 ^{o}C	226	130	95	59	36

Table S2: Rietveld fitting data of the XRD of the Polyhedral 1 sample prepared at 900 ^{o}C

State of Charge	Bragg R-factor			R_f factor			χ^2
	Phase 1	Phase 2	Phase 3	Phase 1	Phase 2	Phase 3	
0	17.94			10.71			1.99
20	13.54			9.54			2.34
40	13.56	21.70		10.57	15.81		2.46
60	9.47	12.11	15.08	6.28	11.84	12.76	2.28
80	18.35	6.45	16.57	13.47	4.32	8.01	2.37
100			16.97			11.99	2.89

Table S3: Rietveld fitting data of the XRD of the Polyhedral 1 sample annealed at 700 ^{o}C

State of Charge	Bragg R-factor			R_f factor			χ^2
	Phase 1	Phase 2	Phase 3	Phase 1	Phase 2	Phase 3	
0	18.45			10.74			3.07
20	18.74			10.45			3.11
40	17.00	20.07		8.10	12.14		2.31
60	17.64	12.52		10.60	8.68		3.16
80	17.02	24.13	19.77	7.47	9.67	9.04	2.61
100		16.25	11.79		9.58	8.28	2.54

Table S4: Rietveld fitting data of the XRD of the Polyhedral 2 sample prepared at 900 ^{o}C

State of Charge	Bragg R-factor			R_f factor			χ^2
	Phase 1	Phase 2	Phase 3	Phase 1	Phase 2	Phase 3	
0	19.51			10.45			2.74
20	21.27			11.40			2.49
40	18.61	14.46		12.12	7.49		2.66
60	15.98	12.68		10.41	8.10		3.08
80		17.17	16.69		9.46	9.28	2.57
100		18.65	14.75		13.15	8.94	2.95

Table S5: Rietveld fitting data of the XRD of the Polyhedral 2 sample annealed at 700 ^{o}C

State of Charge	Bragg R-factor			R_f factor			χ^2
	Phase 1	Phase 2	Phase 3	Phase 1	Phase 2	Phase 3	
0	16.60			9.66			2.73
20	16.12	18.88		8.79	14.31		2.35
40	19.48	18.64		9.48	10.93		2.58
60	20.88	16.35	19.70	11.43	8.71	16.94	2.39
80	21.95	20.19	14.65	14.27	10.04	9.33	2.41
100			16.54			9.88	3.29

For Tables S2-S5, the space group refined was assumed to be *Fd-3m*, with Li in the 8a site, Mn:Ni in a 3:1 ratio in 16d sites, and O in 32e sites.
Phase 1 represents a lithiated phase, Phase 2 represents and intermediate phase, and Phase 3 represents a delithiated phase.

Table S6: Phase compositions based on Rietveld refinement of Poly 1 samples

State of Charge	Prepared at 900 ^{o}C			Annealed at 700 ^{o}C		
	wt% of Each Phase			wt% of Each Phase		
	Phase 1	Phase 2	Phase 3	Phase 1	Phase 2	Phase 3
0	100	0	0	100	0	0
20	100	0	0	100	0	0
40	79.7	20.3	0	73.2	26.8	0
60	31.2	54.5	14.3	38.6	61.4	0
80	9.9	67.2	22.9	5.8	77.6	16.6
100	0	0	100	0	26.9	73.1

Table S7: Phase composition based on Rietveld refinement of Poly 2 samples

State of Charge	Prepared at 900 ^{o}C			Annealed at 700 ^{o}C		
	wt% of Each Phase			wt% of Each Phase		
	Phase 1	Phase 2	Phase 3	Phase 1	Phase 2	Phase 3
0	100	0	0	100	0	0
20	100	0	0	78.6	21.4	0
40	77.2	22.8	0	48.7	51.3	0
60	72.5	27.5	0	26.5	62.2	11.3
80	0	71.4	28.6	13.9	62	24.1
100	0	27.0	73.0	0	0	100