Homogenization Enabled Efficient Regeneration of Spent Ni-rich $LiNi_xCo_yMn_{1-x-y}O_2$ cathodes

Xiangjun Liu,[1,2†] Penglei Yan,[3†] Binglei Jiao,[3*] Guiling Wang,[1] Chunling Zhu,[1] Qiao Zhang,[3] Jinxing Chen,[3] Panpan Xu.[2*]

[1]Key Laboratory of Superlight Materials and Surface Technology of Ministry of Education, Department of Materials Science and Engineering, Harbin Engineering University, Harbin, Heilongjiang, 150001, China

[2]Suzhou Institute of Nano-Tech and Nano-Bionics, Chinese Academy of Sciences, Suzhou, 215123, China

[3]Institute of Functional Nano & Soft Materials (FUNSOM), Jiangsu Key Laboratory for Carbon-Based Functional Materials & Devices, Soochow University, Suzhou, 215123, Jiangsu, P. R. China

[†]X. L. and P. Y. contributed equally to this work.

Corresponding to bljiao@suda.edu.cn; panpanxu2021@sinano.ac.cn.

Figure S1. Digital photograph of the spent DNCM70 battery.

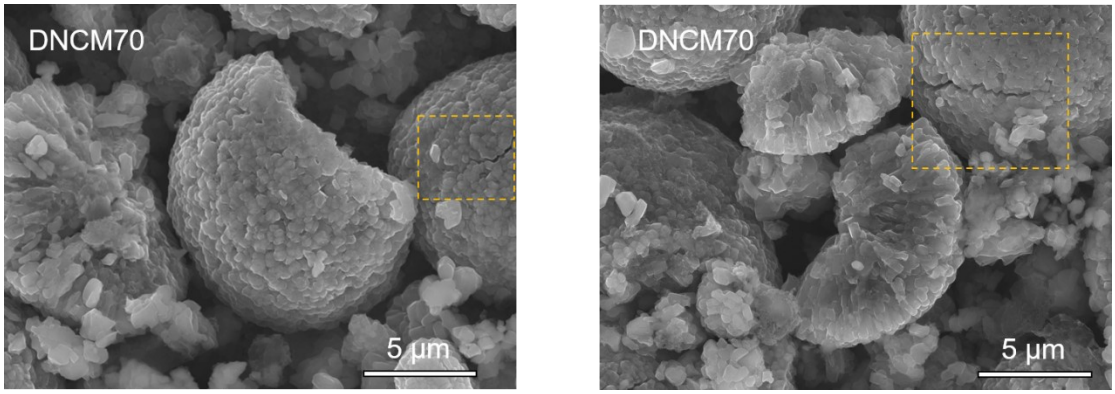

Figure S2. SEM images of DNCM70 before ball milling.

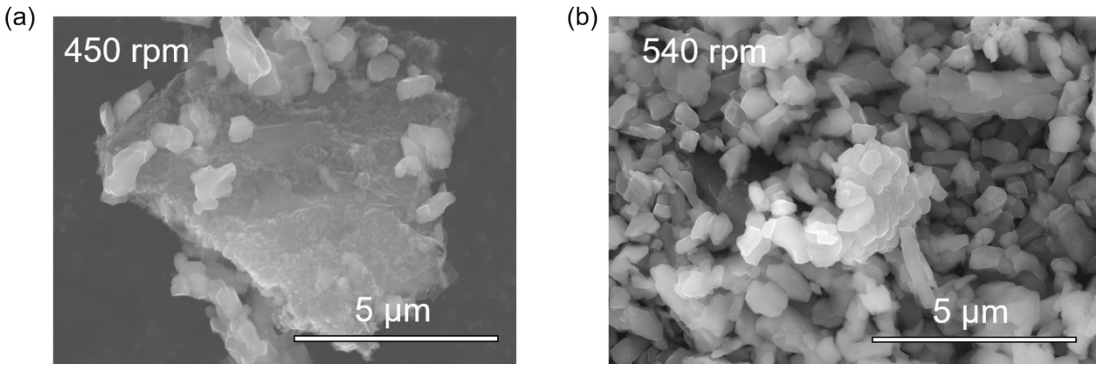

Figure S3. SEM images of HDNCM70 milled at speeds of (a) 450 rpm and (b) 540 rpm.

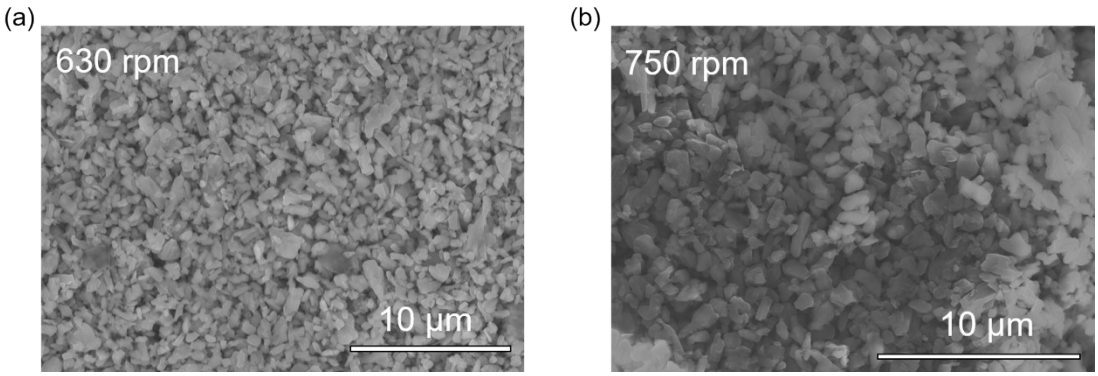

Figure S4. SEM images of HDNCM70 milled at speeds of (a) 630 rpm and (b) 750 rpm.

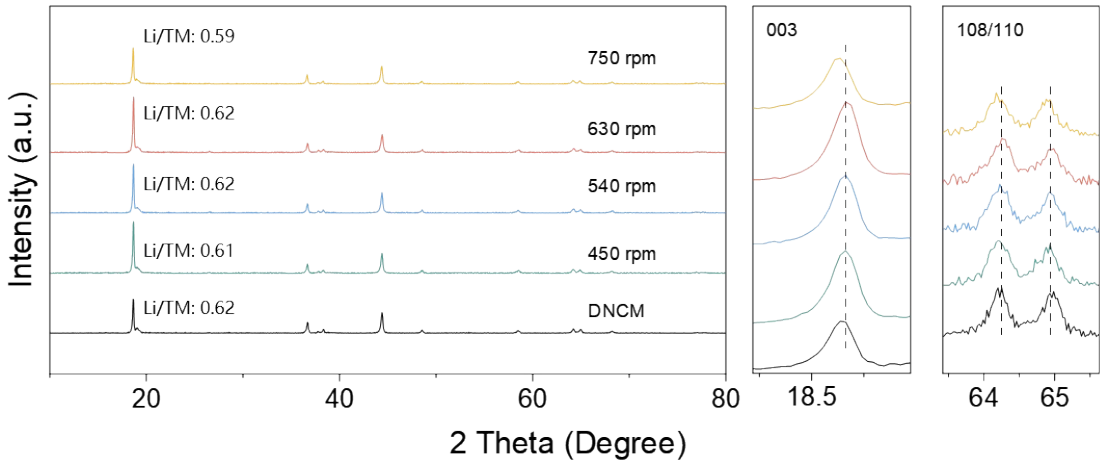

Figure S5. XRD patterns of DNCM70 and HDNCM70 milled at 450, 540, 630, and 750 rpm, with magnified views of the (003) peak and (108)/ (110) peaks.

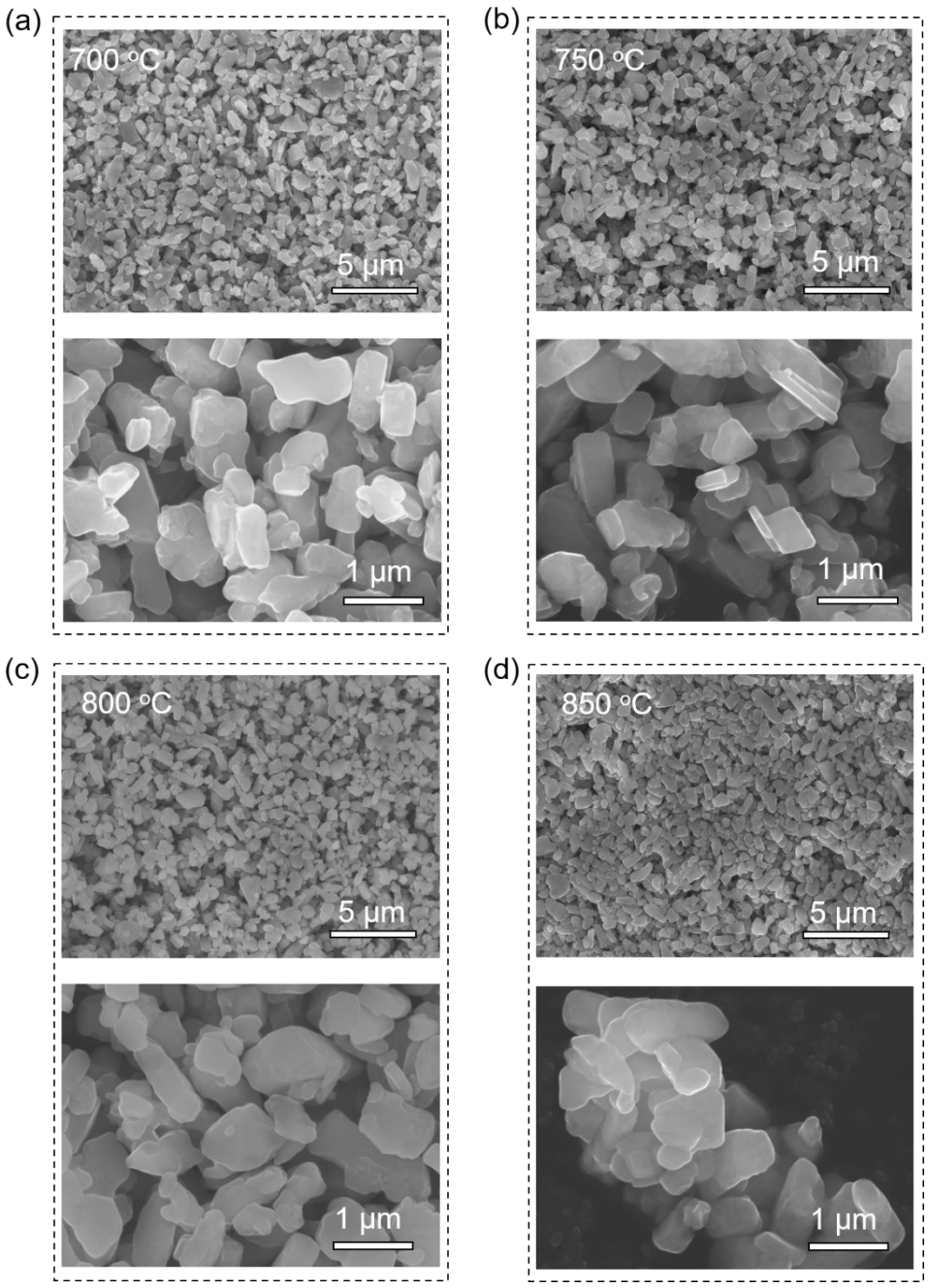

Figure S6. SEM images of RNCM70 sintered at various temperatures: (a) 700 °C, (b) 750 °C, (c) 800 °C, and (d) 850 °C.

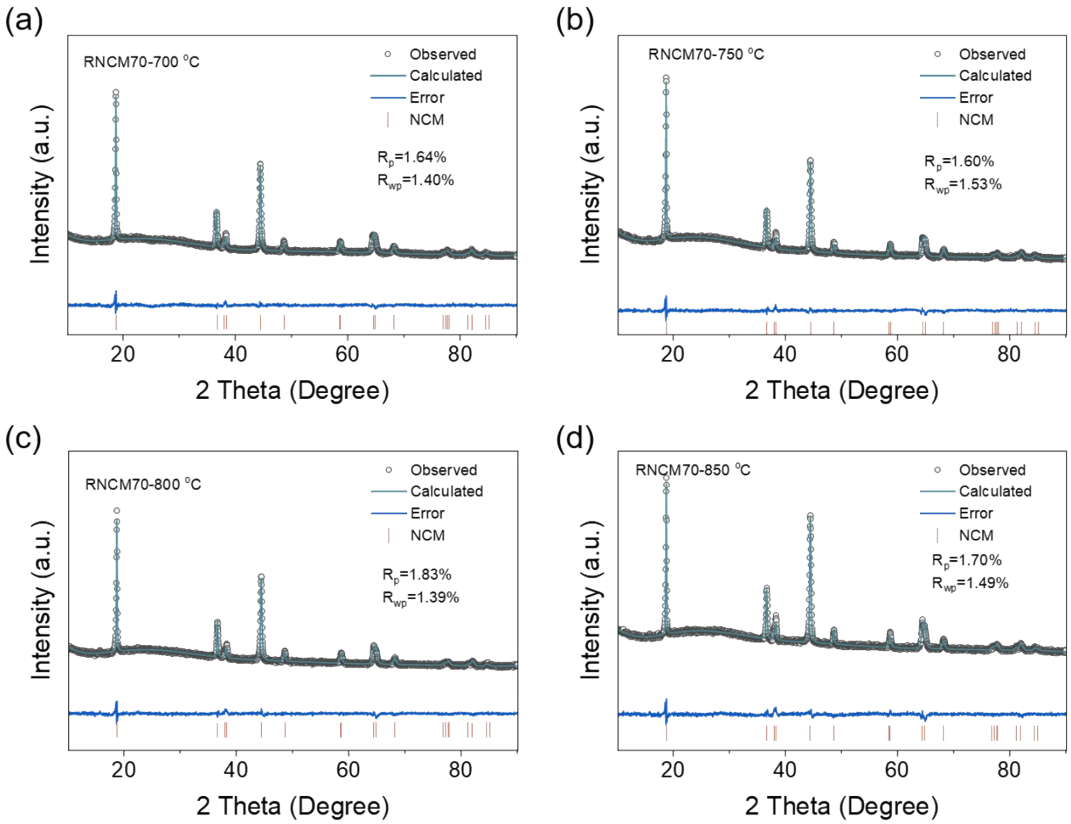

Figure S7. Rietveld XRD refinement results of RNCM70 sintered at various temperatures: (a) 700 °C, (b) 750 °C, (c) 800 °C, and (d) 850 °C.

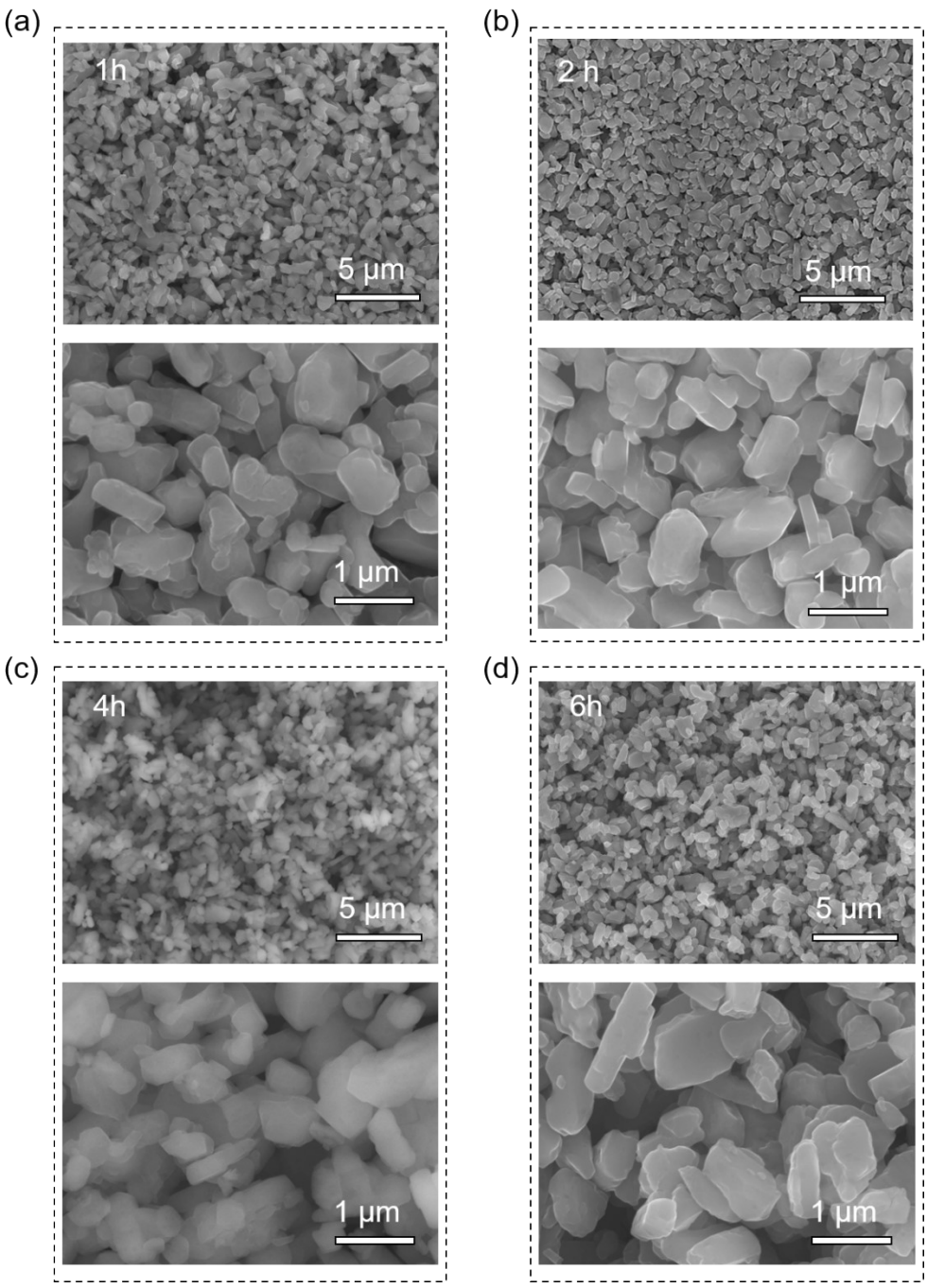

Figure S8. SEM images of RNCM70 sintered at 750 °C for durations of (a) 1 hour, (b) 2 hours, (c) 4 hours, and (d) 6 hours.

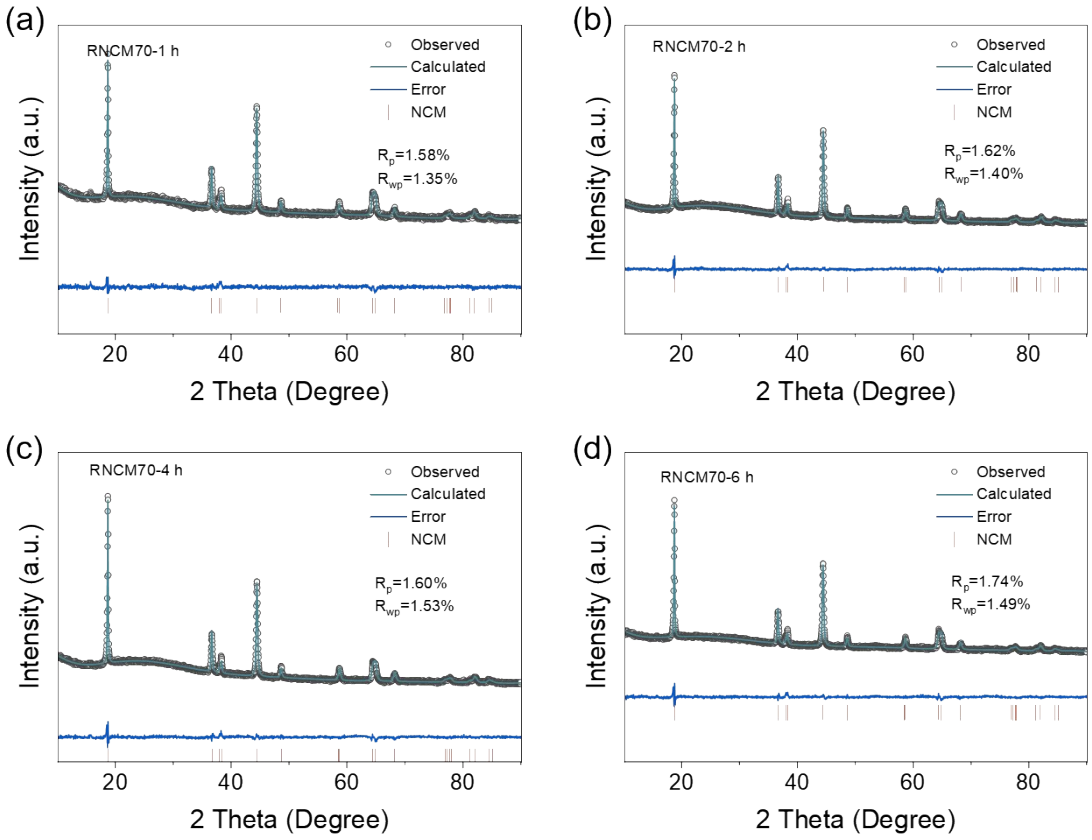

Figure S9. Rietveld XRD refinement results of RNCM70 sintered at 750 °C for durations of (a) 1 hour, (b) 2 hours, (c) 4 hours, and (d) 6 hours.

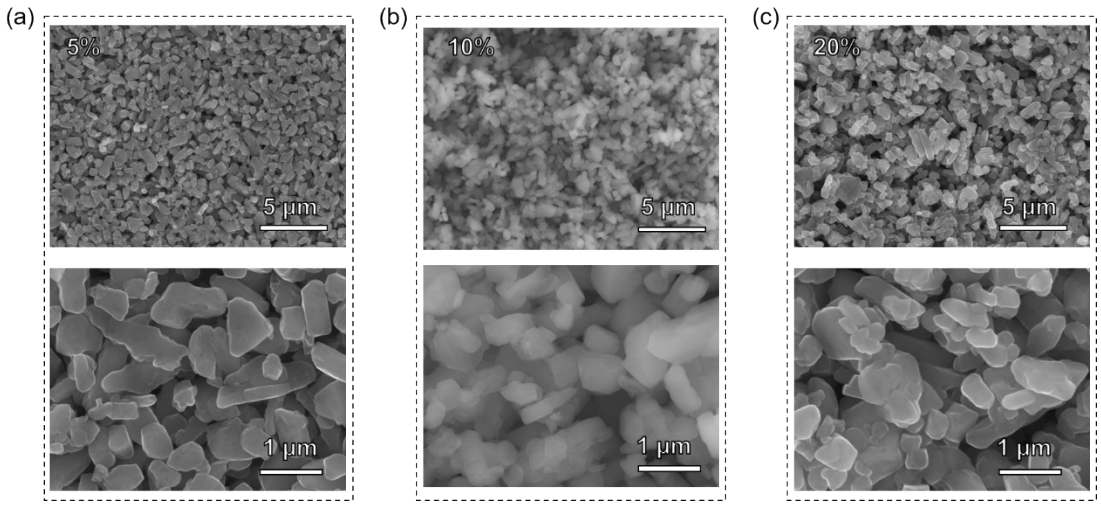

Figure S10. SEM images of RNCM70 sintered at 750 °C for 2 hours with excess Li_2CO_3 contents of (a) 5%, (b) 10%, and (c) 20%.

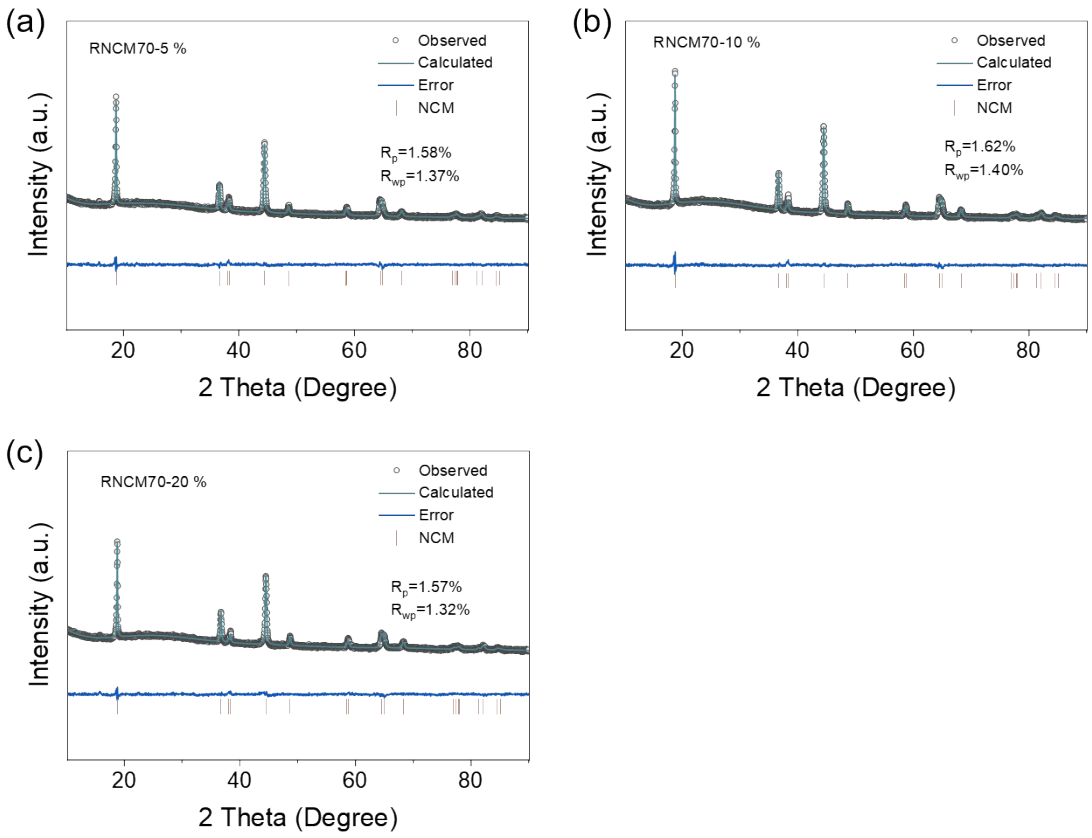

Figure S11. Rietveld XRD refinement results of RNCM70 sintered at 750 °C for 2 hours with excess Li_2CO_3 contents of (a) 5%, (b) 10%, and (c) 20%.

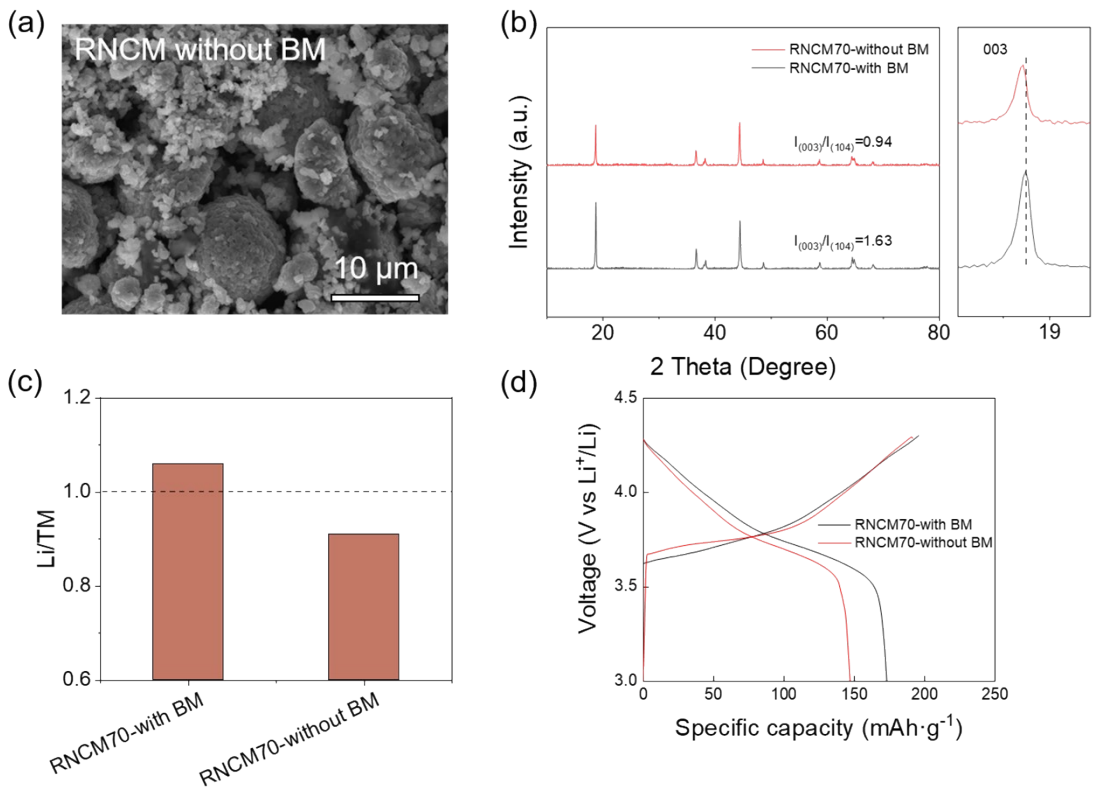

Figure S12 (a) SEM image of RNCM70 sintered at 750 °C for 2 hours with 10% excess Li_2CO_3 without ball milling. (b) XRD patterns, including the amplified (003) peak, comparing regenerated RNCM70 with and without ball milling under the same sintering conditions, (c) corresponding Li/TM content determined by ICP, and (d) corresponding initial charge-discharge profile.

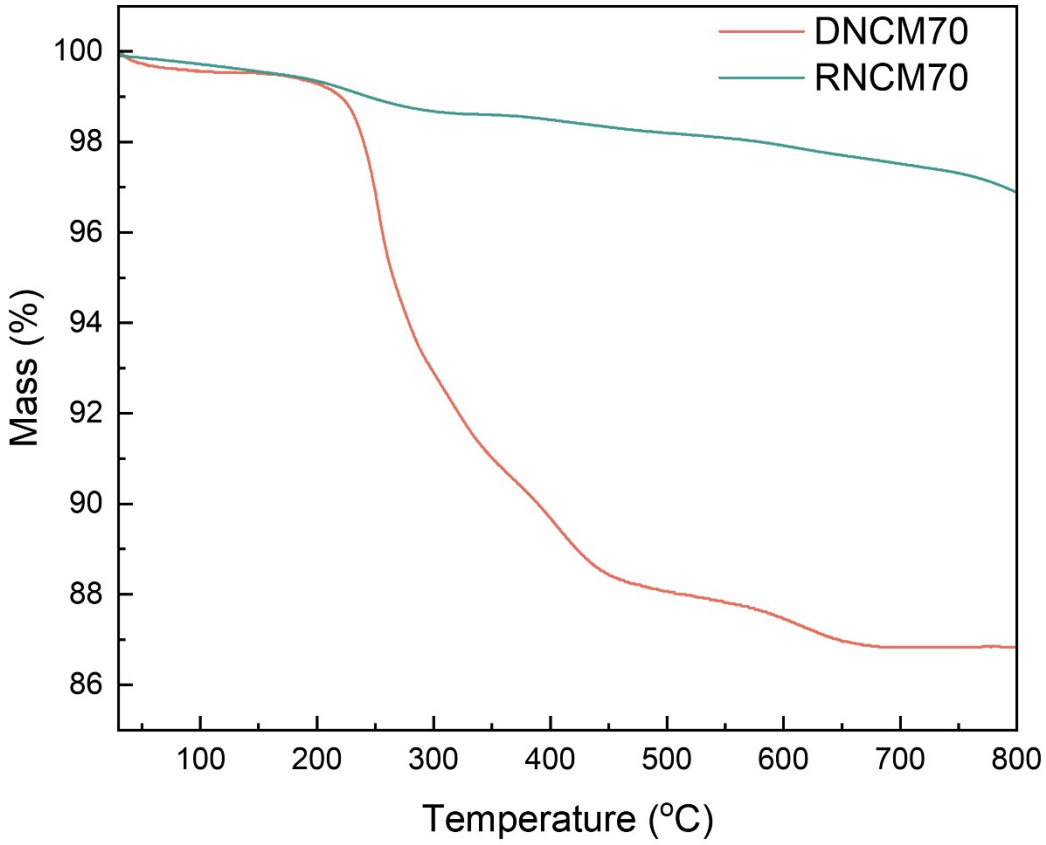

Figure S13. TG curves of DNCM70 and RNCM70 in an air atmosphere.

Table S1. Rietveld XRD refinement results for RNCM70 sintered at 700 °C.

RNCM70-700 ºC (R-3m Space Group)				
Lattice Parameters	a / Å	c / Å	V / Å	c /a
	2.872003	14.1966	101.375	4.943
Li/Ni Antisite	Li/Ni Antisite Ratio			
	5.14%			
Agreement Factors				
R_p	R_{wp}	χ^2		
1.64%	1.40%	1.97		

Table S2. Rietveld XRD refinement results for RNCM70 sintered at 750 °C.

RNCM70-750 °C (R-3m Space Group)				
Lattice Parameters	a / Å	c / Å	V / Å	c /a
	2.87129	14.19695	101.372	4.944
Li/Ni Antisite	Li/Ni Antisite Ratio			
	4.28%			
Agreement Factors				
R_p	R_{wp}	χ^2		
1.60%	1.53%	2.53		

Table S3. Rietveld XRD refinement results for RNCM70 sintered at 800 °C.

RNCM70-800 °C (R-3m Space Group)				
Lattice Parameters	a / Å	c / Å	V / Å	c /a
	2.87311	14.20492	101.548	4.944
Li/Ni Antisite	Li/Ni Antisite Ratio			
	5.38%			
Agreement Factors				
R_p	R_{wp}	χ^2		
1.83%	1.39%	1.93		

Table S4. Rietveld XRD refinement results for RNCM70 sintered at 850 °C.

RNCM70-850 °C (R-3m Space Group)				
Lattice Parameters	a / Å	c / Å	V / Å	c /a
	2.87560	14.21754	101.783	4.9442
Li/Ni Antisite	Li/Ni Antisite Ratio			
	8.33%			
Agreement Factors				
R_p	R_{wp}		χ^2	
1.70%	1.49%		2.23	

Table S5. Rietveld XRD refinement results for RNCM70 sintered at 750 °C for a duration of 1 hour.

RNCM70-1 h (R-3m Space Group)				
Lattice Parameters	a / Å	c / Å	V / Å	c /a
	2.87313	14.20589	101.557	4.9444
Li/Ni Antisite	Li/Ni Antisite Ratio			
	7.84%			
Agreement Factors				
R_p	R_{wp}	χ^2		
1.58%	1.35%	1.82		

Table S6. Rietveld XRD refinement results for RNCM70 sintered at 750 °C for a duration of 2 hours.

RNCM70-2 h (R-3m Space Group)				
Lattice Parameters	a / Å	c / Å	V / Å	c /a
	2.86939	14.19487	101.321	4.947
Li/Ni Antisite	Li/Ni Antisite Ratio			
	4.28%			
Agreement Factors				
R_p	R_{wp}	χ^2		
1.62%	1.40%	1.96		

Table S7. Rietveld XRD refinement results for RNCM70 sintered at 750 °C for a duration of 4 hours.

RNCM70-4 h (R-3m Space Group)				
Lattice Parameters	a / Å	c / Å	V / Å	c /a
	2.87129	14.19695	101.372	4.944
Li/Ni Antisite	Li/Ni Antisite Ratio			
	4.72%			
Agreement Factors				
R_p	R_{wp}		χ^2	
1.60%	1.53%		2.53	

Table S8. Rietveld XRD refinement results for RNCM70 sintered at 750 °C for a duration of 6 hours.

RNCM70-6 h (R-3m Space Group)				
Lattice Parameters	a / Å	c / Å	V / Å	c /a
	2.874255	14.20457	101.627	4.942
Li/Ni Antisite	Li/Ni Antisite Ratio			
	5.05%			
Agreement Factors				
R_p	R_{wp}	χ^2		
1.74%	1.49%	2.21		

Table S9. Rietveld XRD refinement results for RNCM70 sintered at 750 °C for a duration of 2 hour with 5% excess Li_2CO_3.

RNCM70-5% (R-3m Space Group)				
Lattice Parameters	a / Å	c / Å	V / Å	c /a
	2.87305	14.20084	101.515	4.942775
Li/Ni Antisite	Li/Ni Antisite Ratio			
	7.44%			
Agreement Factors				
R_p	R_{wp}		χ^2	
1.58%	1.37%		1.87	

Table S10. Rietveld XRD refinement results for RNCM70 sintered at 750 °C for a duration of 2 hour with 10% excess Li_2CO_3.

RNCM70-10% (R-3m Space Group)				
Lattice Parameters	a / Å	c / Å	V / Å	c /a
	2.86939	14.19487	101.321	4.947
Li/Ni Antisite	Li/Ni Antisite Ratio			
	4.28%			
Agreement Factors				
R_p	R_{wp}		χ^2	
1.62%	1.40%		1.96	

Table S11. Rietveld XRD refinement results for RNCM70 sintered at 750 °C for a duration of 2 hour with 20% excess Li_2CO_3.

RNCM70-20% (R-3m Space Group)				
Lattice Parameters	a / Å	c / Å	V / Å	c /a
	2.87045	14.19743	101.307	4.94606
Li/Ni Antisite	Li/Ni Antisite Ratio			
	4.45%			
Agreement Factors				
R_p	R_{wp}		χ^2	
1.57%	1.32%		1.75	