Supporting	Information
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Lithium-ion Batteries Recycling Technology Based on Controllable Product Morphology and Excellent Performance

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Figure S1 The separated Li₂SO₄ solution and Co₃O₄ powder.



Figure S2 XRD patterns of the roasted powder.



Figure S3 The XRD patterns of the Li_2CO_3 powder.



Figure S4 SEM images of Co₃O₄ particles



Figure S5 Particle size analysis results of the Co₃O₄ particles



Figure S6 (a)SEM image and (b)XRD patterns of commercial Co3O4 powder.



Figure S7 Cycle performance of the nano-Co₃O₄ electrode at a current density of 50 mA g⁻¹.



Figure S8 (a) Cycle performance of the re-synthesized LiCoO₂ powder at 1C (1C=150 mA h/g), (b) Discharge curves for the re-synthesized LiCoO₂ powder at different current densities



Figure S9 The XRD patterns of scrap CoSO₄·7H₂O powder. (The powder was obtained by heating CoSO4·7H2O to 150°C for 20min.)



Figure S10 The XRD patterns of scrap CoSO₄·7H₂O powder. (The powder was obtained by heating CoSO4·7H2O to 260°C for 20min.)



Figure S11 The XPS survey spectra of the $Li_{1-x}CoO_2$ and Co_3O_4 .

NO.	Sample	Initial capacity (mA h/g)	Charge/discharge condition	Performance (mA h/g)	Ref.
1	nanoparticles	1140	50 mA/g, 100 cycles	987.2	This work
2	Co ₃ O ₄ @Carbon Nanotube	1250	100 mA/g, 60 cycles	781	[1]
3	nanotube	928	50 mA/g, 80 cycles	380	[2]
4	nanoparticles	1109	50 mA/g, 30 cycles	970	[3]
5	hollow microspheres	1087.2	50 mA/g, 30 cycles	792.7	[4]
6	nanoparticles	1118.5	100 mA/g, 200 cycles	955.5	[5]
7	hollow-structured	1107	50 mA/g, 50 cycles	880	[6]
8	porous nanoflaked	1108	0.2 C, (1 C = 890 mA/g) ,100 cycles	908	[7]
9	MOF	1200	200 mA/g, 100 cycles	924.1	[8]

Table S1 A brief review of published laboratory work on Co_3O_4

Table S2 The fit result of EIS parameters

Samples			I	Element (Ω)				
	R _s	R _{ct}	CPE ₁ -T	CPE ₁ -P	W ₁ -R	W ₁ -T	W ₁ -P	
			_					

Nano-Co ₃ O ₄	3.175	66.78	0.000003.4241	0.76968	137.8	0.27638	0.58672
Commercial Co ₃ O ₄	1.95	146.2	0.00015207	0.56753	306.1	0.52531	0.69768

Table S3 Refined lattice parameters of all samples

Samples	a	c	volume	c/a	$I_{(003)}/I_{(104)}$	$I_{(006)} + I_{(012)} / I_{(104)}$
Li _{1-x} CoO ₂	2.8144	14.0408	96.3149	4.9889	3.0347	0.9275
C0 ₃ O ₄ →LiC0O ₂	2.8143	14.0451	96.3356	4.9907	5.0975	0.4035
Commercial LiCoO2	2.8159	14.0499	96.4798	4.9895	12.1334	0.7169

D		Peak binding	energy	
Pe	ак ——	Li _{1-x} CoO ₂	Co ₃ O ₄	
		284.8	284.8	
C 1s		285.97	286.25	
		288.66	288.3	
		529.21	529.87	
0	1s	529.75	531.37	
		531.4	532.54	
	Co _I P3	779.27	780.04	
Co 2n	Co _I P1	794.35	795.34	
C0 2p	Co _{II} P3	780.62	781.51	
	Co _{II} P1	795.86	796.87	

Table S4 Peak binding energies for all deconvoluted C 1s, O 1s and Co 2p peaks from XPS.

Co ion		Atomic or	bital (3d)		
Co ³⁺	1	1	1	1	
Co ²⁺	1	1	1	1	1

Table S5 Electronic configuration of Co ion

Table S6 The bond length of the reactants

LiCoO ₂	Bond length/Å
Li-O	1.63147
Li-Co	1.61547
Co=O	1.56039
CoSO4	Bond length/Å
CoSO ₄ S=O	Bond length/Å
CoSO ₄ S=O S-O	Bond length/Å 1.42715 1.61271

Reference:

Gu, D.; Li, W.; Wang, F.; Bongard, H.; Spliethoff, B.; Schmidt, W.; Weidenthaler,
 C.; Xia, Y.; Zhao, D.; Schuth, F., Controllable Synthesis of Mesoporous Peapod-like
 Co3O4@Carbon Nanotube Arrays for High-Performance Lithium-Ion Batteries.
 Angew Chem Int Ed Engl 2015, 127, 7166-7170.

2. Lou, X. W.; Deng, D.; Lee, J. Y.; Feng, J.; Archer, L. A., Self-Supported Formation of Needlelike Co3O4 Nanotubes and Their Application as Lithium-Ion Battery Electrodes. *Advanced Materials* **2008**, *20* (2), 258-262.

3. Yan, N.; Hu, L.; Li, Y.; Wang, Y.; Zhong, H.; Hu, X.; Kong, X.; Chen, Q., Co3O4 Nanocages for High-Performance Anode Material in Lithium-Ion Batteries. *The Journal of Physical Chemistry C* **2012**, *116* (12), 7227-7235. Wang, J.; Yang, N.; Tang, H.; Dong, Z.; Jin, Q.; Yang, M.; Kisailus, D.; Zhao, H.; Tang, Z.; Wang, D., Accurate control of multishelled Co3O4 hollow microspheres as high-performance anode materials in lithium-ion batteries. *Angew Chem Int Ed Engl* 2013, *52* (25), 6417-6420.

5. Xu, G.-L.; Li, J.-T.; Huang, L.; Lin, W.; Sun, S.-G., Synthesis of Co3O4 nanooctahedra enclosed by facets and their excellent lithium storage properties as anode material of lithium ion batteries. *Nano Energy* **2013**, *2* (3), 394-402.

 Deli Wang, Y. Y., Huan He, Jie Wang, Weidong Zhou, and Hector D. Abru~na,, Template-Free Synthesis of HollowStructured Co3O4 Nanoparticles. *ACS nano* 2015, 9 (2), 1775-1781.

 Wen, J.; Xu, L.; Wang, J.; Xiong, Y.; Ma, J.; Jiang, C.; Cao, L.; Li, J.; Zeng, M., Lithium and potassium storage behavior comparison for porous nanoflaked Co3O4 anode in lithium-ion and potassium-ion batteries. *Journal of Power Sources* 2020, *474*.
 Y.-H.; Li, J.-H.; Xu, Z.-F.; Liu, J.-M.; Liu, S.-J.; Wang, R.-X., Metal–organic framework derived porous nanostructured Co3O4 as high-performance anode materials for lithium-ion batteries. *Journal of Materials Science* 2021, *56* (3), 2451-2463.