## Supporting Information

## Effective Separation of LiNi<sub>0.5</sub>Co<sub>0.2</sub>Mn<sub>0.3</sub>O<sub>2</sub> Cathode Materials via

## **Digestion of PVDF Enabling a Close-loop Recycle**

Zuoyu Qin,<sup>a</sup> Jiaqi Li,<sup>a</sup> Tao Zhang,<sup>a</sup> Zuxin Wen,<sup>a</sup> Zhicheng Zheng,<sup>a</sup> Ying Zhang,<sup>b</sup> Ning Zhang,<sup>a</sup> Chuankun Jia,<sup>c</sup> Xiaohe Liu<sup>b</sup> and Gen Chen<sup>a</sup>\*

<sup>a</sup> School of Materials Science and Engineering, Key Laboratory of Electronic Packaging and Advanced Functional Materials of Hunan Province, Central South University, Changsha 410083, China

<sup>b</sup> Zhongyuan Critical Metals Laboratory and School of Chemical Engineering, Zhengzhou University, Zhengzhou, Henan 450001, P. R. China

<sup>c</sup> College of Materials Science and Engineering, Changsha University of Science & Technology, Changsha, 410114, China

\* Correspondence: geenchen@csu.edu.cn (G. Chen)



**Figure S1** Digital photos of cathode strips after ST pretreatment at different temperature: (a) 120 °C, (b) 130 °C, (c) 140 °C, (d) 150 °C.



Figure S2 The corresponding EtOH solution after solvothermal treatments at different temperature.



**Figure S3** The collecting solution after solvothermal treatments using different solvents: (a) DMC, (b) DMF and (c) DMAC



**Figure S4** The molecular structure tests of collecting solution after ST treatments with different cooking time: (a) UV spectra. Inset shows the collecting solution after ST treatment with the pristine PVDF at 1 h. (b) Raman spectra.



**Figure S5** Digital photos of different powders gathered after pretreatment of DT, ST and SN.



Figure S6 The EDS results of DT, ST and SN.



Figure S7 (a) XRD pattern and (b) Rietveld refinements of R-NCM.



Figure S8 Deep cycling of the commercial NCM523 cylindrical cell.

	_		-		
Solution	Operation	Temperatur e	Mechanism	Solid:liquid	Ref.
H <sub>2</sub> O	Ultrasound + stir	/	Shearing force	>1:1	Li et al.[25]
AlCl <sub>3</sub> -NaCl	Tube furnace + N <sub>2</sub>	160 °C	Heat melting	10:1	Wang et al. [37]
(CH <sub>2</sub> OH) <sub>2</sub>	Sieving + stir	160 °C	Competitive inhibition	10:1	Bai et al. [40]
CH <sub>3</sub> CH <sub>2</sub> OH	/	150 °C	Chemical reaction	1:1	This work

**Table S1**. The detailed comparison information among this work and other three works.

		,	1	
Samples	a (Å)	c (Å)	Li/Ni mixing	
DT	2.8797	14.2679	14.7%	
ST	2.8678	14.2624	8.8%	
SN	2.8664	14.3114	10.1%	

Table S2. The result of Rietveld refinement of DT, ST and SN pretreatment.

			,	
Samples	F (At%)	C (At%)	O (At%)	Al (At%)
DT	0.8	16.7	4.0	78.5
ST	0.5	18.0	1.4	80.1
SN	1.2	43.5	3.0	52.2

Table S3. The EDS results of DT, ST and SN.

Samples	a (Å)	c (Å)	Li/Ni mixing
R-NCM	2.8739	14.2502	4.0%

Table S4. The result of Rietveld refinement of R-NCM.