Electronic Supplementary Information

Increased residual lithium compounds guided design for green recycling of spent lithium-ion cathodes

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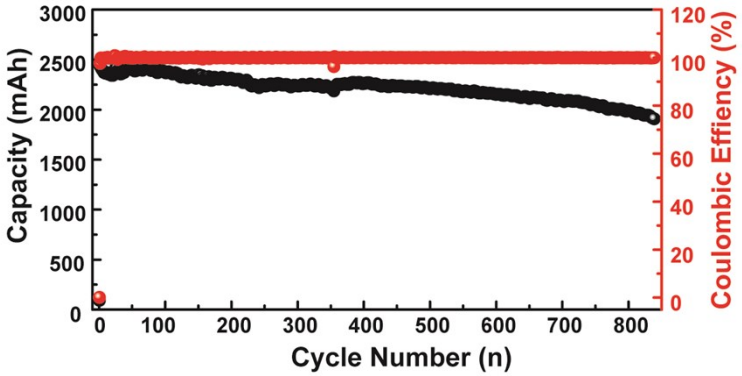

Figure S1. Cycling performance of commercial NCM523 pouch cell.

The performance of the pouch cell we choose to recycle is shown in Figure S1. The initial capacity of the pouch cell is 2430 mAh, and the electrochemical performance was gradually decaying with the cycle time increasing. The capacity degraded to 78.4% after 840 cycles, which is defined as reaching the end of life.

Table S1. Qualitative tests of residual lithium compounds of P-NCM523 and D-NCM523.

Sample	1	2	3	Mean	Standard deviation
pH of P-NCM523 eluent	7.13	7.07	7.08	7.09	3.2%
pH of D-NCM523 eluent	8.25	8.30	8.23	8.26	3.5%

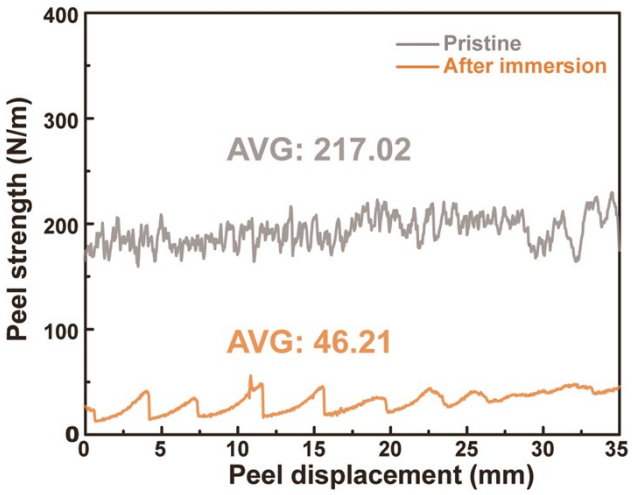

Figure S2. Peeling strength change of the electrode sheet under electrolyte immersion.

Table S2. Atomic ratio of different elements in the filtrate

Element	Li	Al	Ni+Co+Mn
Atomic ratio	0.977	0.021	＜0.002

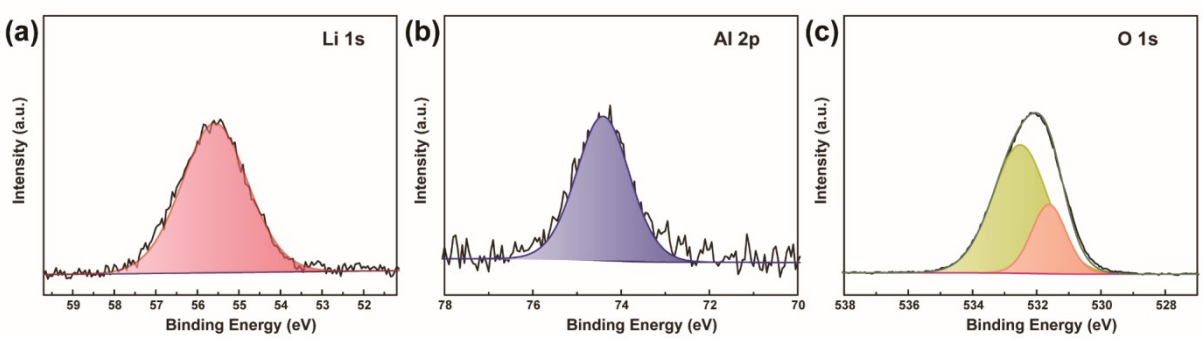

Figure S3. XPS spectra of (a) Li 1s, (b) Al 2p and (c) O1s in the residue.

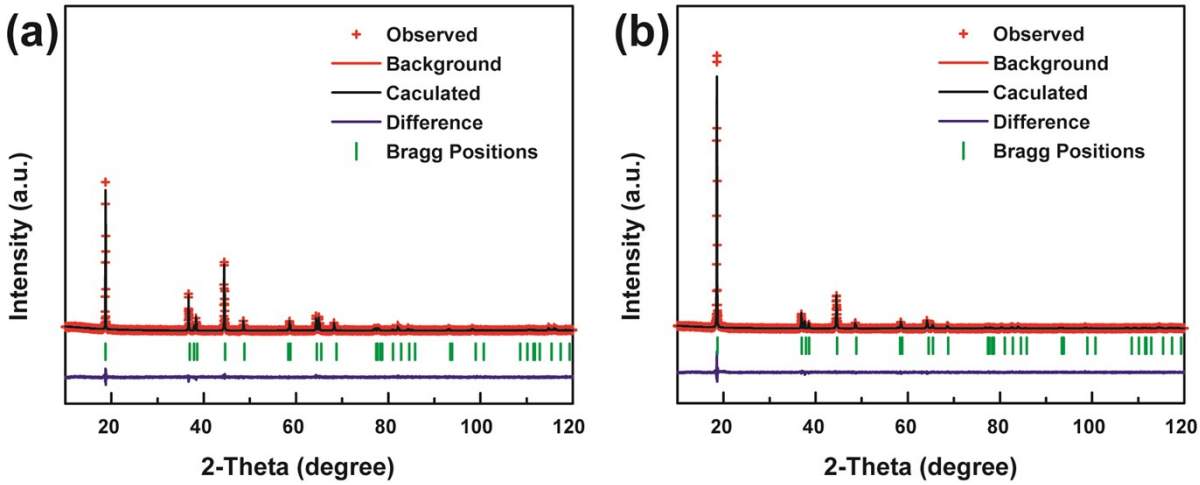

Figure S4. XRD patterns and Rietveld refinement plots of (a) the P-NCM523 and (b) the D-NCM523.

Table S3. Lattice parameters of P-NCM523, D-NCM523 and R-NCM523

Sample	a /Å	c/Å	Li/Ni mixing/%	R_{wp}/%	R_p/%
P-NCM523	2.8712(8)	14.2429(6)	3.00	3.28	2.52
D-NCM523	2.8539(6)	14.3335(4)	0.04	2.43	2.56
R-NCM523	2.8716(2)	14.2426(5)	2.2	3.61	2.76

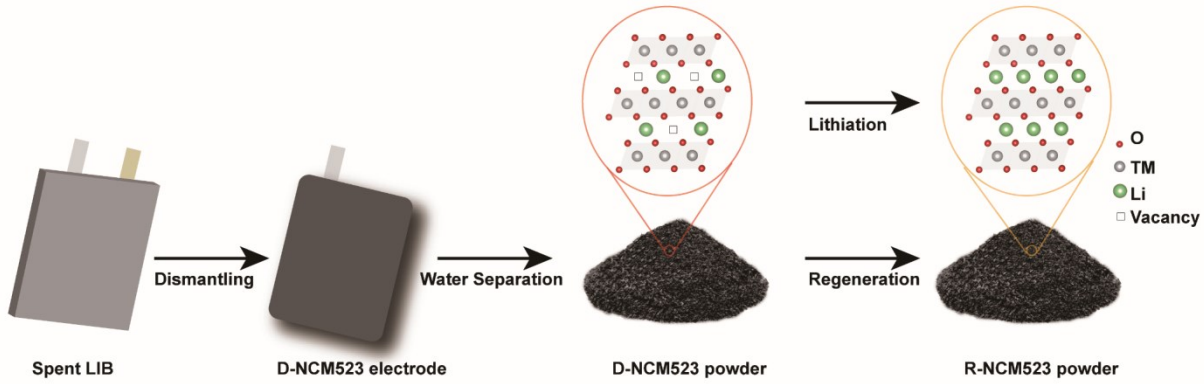

Figure S5. Schematic diagram of the recycling process in this work.

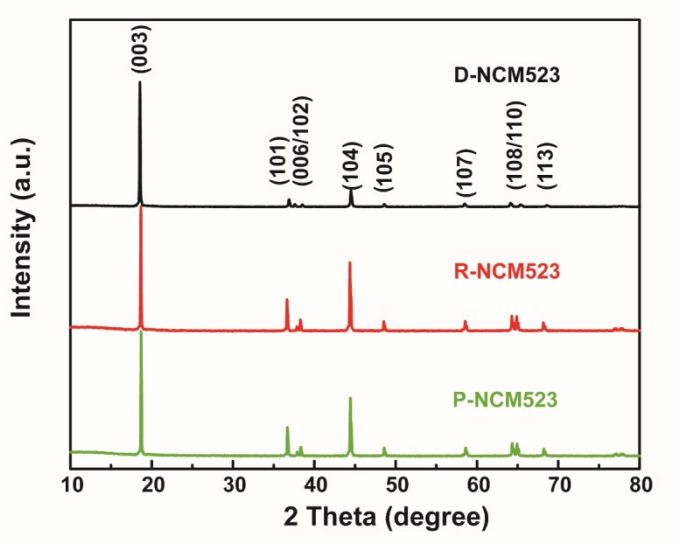

Figure S6. XRD patterns of the D-NCM523, R-NCM523 and P-NCM523

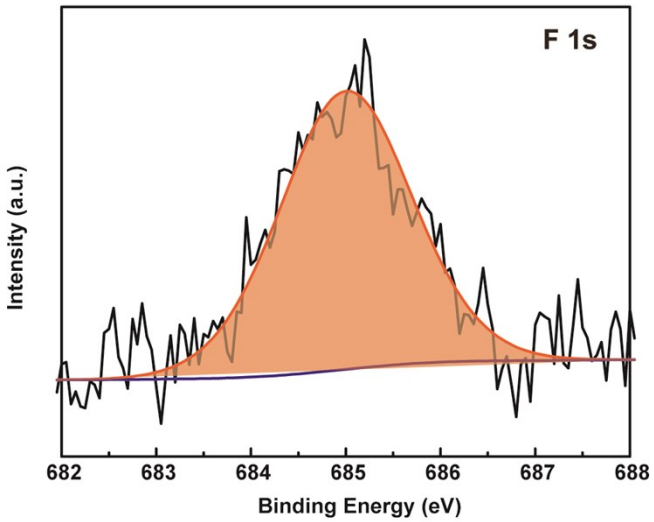

Figure S7. XPS spectrum of F 1s in R-NCM523.

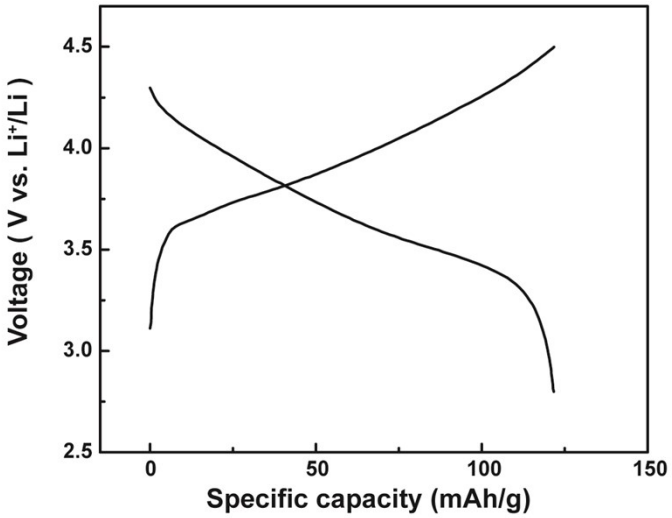

Figure S8. Charge/discharge profile of the last cycle of the pouch cell.