Supplementary Information

Spray pyrolysis technology-based closed-loop for regenerating single-

crystal cathodes from spent lithium-ion batteries

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Fig. S1 Photos of (a) spent LIBs, (b) cathodes, anodes, and separators of the batteries and (c) cathode powders after exfoliating Al foils.



Fig. S2 Photo of the spray pyrolysis device, mainly including a vertical furnace reactor, ultrasonic atomizer, and a powder collector (nickel foam).



Fig. S3 The process of powder collection.



Fig. S4 Photos of LiCl and CoCl₂ mixture sintered at (a) 500°C; (b) 525°C; (c) 550°C.



Fig. S5 SEM images of pyrolyzed powders at (a) 650°C; (b) 750°C; (c) 850°C.



Fig. S6 EDS diagram of (a) pyrolysis product and (b) water leaching residue at 750°C; EDS diagram of (c) pyrolysis product and (d) water leaching residue at 850°C.



Fig. S7 TG/DTA curves of LiCl in air.



Fig. S8 (a) Photo of the regenerated Li_2CO_3 ; (b) XRD pattern of the regenerated

Li₂CO₃.



Fig. S9 (a) XRD pattern and (b) SEM image of the cycled R-LCO electrode.

Temperature/°C	650	750	850
Li concentration/ (mg L ⁻¹)	1.60	2.05	1.12

Table S1 The Li concentration in the sample obtained at 650°C, 750°C, and 850°C.*

*Note: Solution preparation for ICP measurement: 100.0 mg Co_3O_4 residue was dissolved in HCl (7.9 mol L⁻¹) and the volume was fixed at 50 mL. Then the solutions were measured without diluting.