Supplementary Information (SI) for ChemComm.

Supporting Information for:

Upcycling of Spent $LiCoO_2$: Engineering the Coordination-Trapping Behaviors towards Conversion-type Anode for Advanced Li-storage

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Experiment Part

1. Chemicals and materials.

In this experiment, all the chemicals, including Sodium diethyldithiocarbamate (DDTC-Na), HCl, Na_2SO_4, phytic acid solution was brought from Macklin Company. And, all of the chemicals were utilized directly in the materials preparation without further purification. The component of button battery, including the LX-031electrolyte, CR 2016 battery can and PP diaphragm were purchased from Guangdong Canrd New Energy Technology Co.,Ltd.

2. Battery disassembling.

The spent $LiCoO_2$(LCO)/graphite pouch battery was discharging in the 1 mol/L NaCl solutions until the voltage was decreased into the 1.0V, and manually disassembled the pouch battery. Later, separated the cathode\anode sheet and diaphragm to recovery the spent LCO cathode sheet. And, for separating the spent LCO and Al foil, the spent LCO cathode sheet was soaked in the phytic acid solution for 10 min. Supported by the complexing ability between the Al ions and phytic acid molecules, the spent LCO could be effective removed from the cathode sheets. As-obtained LCO was dried with 70 °C for 10 h to remove the water, and later grind the powders to obtained the spent LCO.

3. Leaching of Spent LCO

1.5 g of spent LCO containing 59.67 wt% Co was reacted with 50 mL diluted HCl, prepared by mixing 6 mL of HCl (37 wt.%) with 46 mL of deionized water (DI). The reaction was carried out under heat at 60 °C, during which H_2O_2 was added to expedite the dissolution of LCO. The reaction was considered complete when no further solid remained. The resulting solution was then adjusted to a pH around 6.8 using aqueous ammonia, yielding a $CoCl_2$ solution also containing LiCl. For comparison, a control $CoCl_2$ solution was prepared by dissolving 2 g of $CoCl_2$ in 50 mL of DI.

4. Preparation of CoS@NSC anode.

Preparation of Precursor Ni(DDTC)$_2$. First, an aqueous solution was prepared with a concentration of 5 g/50 mL of sodium diethyl dithiocarbamate (denoted as solution A). Co^{2+} solutions, which derived from the spent LCO or $CoCl_2$ solutions, denoted as solution B. Second, a dropper was used to drop solution B into solution A, which was being stirred, with the mixture continuing to be stirred for 30 min after dripping; then, it was left to settle for 10 h at room temperature. Lastly, the mixture was rinsed three times with deionized water, and the solution was filtered after rinsing. The solid obtained after suction filtration was freeze-dried after 12 h to obtain precursor $Co(DDTC)_2$.

Preparation of CoS@NSC. $Co(DDTC)_2$ and Na_2SO_4 were mixed in a mass ratio of 5:1, and the mixture was ground using a mortar until a uniform morphology was achieved (grinding duration: 30 minutes). The resulting mixture was transferred to the constant-temperature zone of a high-temperature tubular furnace, heated to 600 °C at a rate of 3 °C min^{-1} under an argon protective atmosphere, and maintained at this temperature for 2 hours. Upon cooling to room temperature, the product was washed three times with deionized water to remove impurities and subsequently dried in a vacuum oven at 80 °C, yielding *CoS@NSC*.

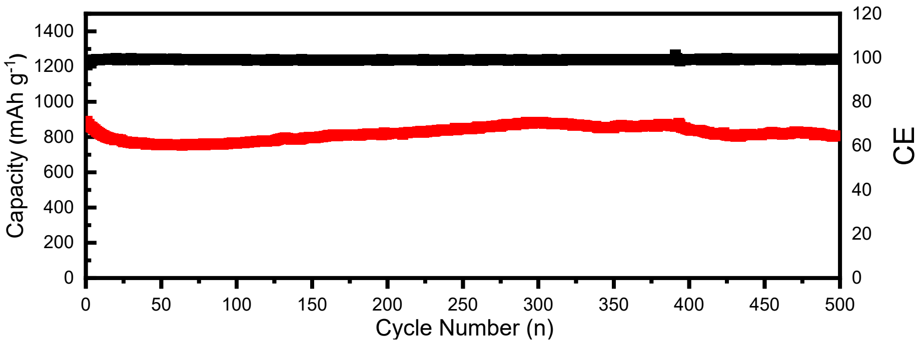

Figure S1. The long-term cycling stability of LCO6-CoS@NSC.

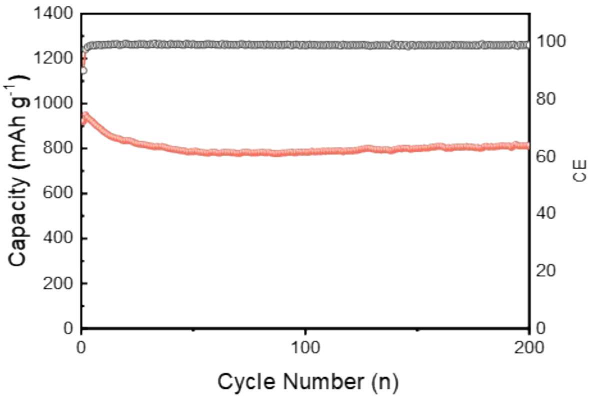

Figure S2. The cycling performance of CoCl6-CoS@NSC.

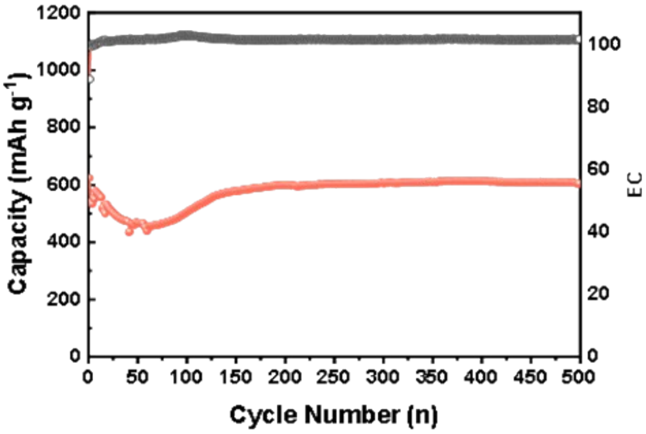

Figure S3. The Na-storage performance at 1.0 C of LCO6-CoS@NSC.

Table S1. elemental analysis of the LCO6-CoS@NSC.

Al	B	Ba	Ca
0.504342%	0.013120%	0.004823%	0.029039%
Co	Cr	Fe	K
62.97811%	0.006110%	0.035442%	0.018206%
Li	Mg	Mn	Na
0.015341%	0.031474%	0.064104%	0.858526%
Ni	S	Sr	Zn
0.020347%	35.054960%	0.002363%	0.024378%
Si	Ti	Zr	
0.056157%	0.015605%	0.099774%	