Electronic Supplementary Material (ESI) for ChemComm.

Supporting Information for:

Designing Functional Li_2CuO_2-coated Separators from Cu Foil towards spent $LiFePO_4$ Cathode Regeneration

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Experimental Part

Materials Preparation

The used LFP/graphite battery was provided by a company based in Hunan, China. The commercial graphite and LFP (C-LFP) cathode were obtained from Guangdong Canrd New Energy Technology Co. Ltd. Additionally, coin cell battery components, including coin cells, Celgard 2500 separator, polyvinylidene fluoride (PVDF), carbon black, aluminum (Al) foil, and li metal were also purchased from the same company. The electrolyte, purchased from DoDochem, which was mixed by LiPF6 (1 M), ethylene carbonate (EC), diethyl carbonate (DEC), and dimethyl carbonate (DMC) in a volumetric ratio of 1:1:1.

Spent LFP Cathode/Cu Foil Material Harvesting

The spent LFP/graphite battery was immersed in a NaCl solution to completely discharge its power, followed by its disassembly to extract the spent cathode and anode sheets. To obtain LFP cathode materials, the spent cathode sheets were separated from the water, removing the Al foil. (500g LFP cathode sheets was soaked in 2L water) The obtained cathode sheets were then dried at 70°C for 3 hours to eliminate any residual water. Finally, a 10-minute grinding process was performed to transform the sheets into powder-like LFP samples, which were designated as spent-LFP (S-LFP). The same method was used to process the waste anode electrode plates to extract the copper foil.

Preparation of CuO and Li_2CuO_2

Cut the spent Cu foil into as small fragments as possible using scissors, later the as-cut Cu foil was sintered at 500℃ for 2h in the atmosphere to obtain CuO. Then, weigh a certain proportion of CuO and LiOH (molar ratio of LiOH: CuO = 2.2: 1) in the ball milling tank, later mixed them with a speed of 400rmp/min for 2h. The mixture was sintered at 800°C for 18h to obtain the Li_2CuO_2.

Preparation of Li_2CuO_2-based Functional Separator

Active materials (Li_2CuO_2), polyvinylidene difluoride (PVDF), and conductive carbon black particles were blended to obtain the slurry (active materials: conductive carbon black: PVDF binder = 80:10wt:10wt %). Later, the slurry was coated on the surface of the commercial PP separator to prepare Li_2CuO_2-based functional separator (named as Li_2CuO_2@PP). As-obtained Li_2CuO_2-based functional separator was dried at 50°C for 5 hours and transferred to a glove box filled with Ar. The loading mass of Li_2CuO_2 was about 0.8 mg.

Materials Characterization.

The composition of the as-prepared material was determined using X-ray diffraction (XRD) analysis. The morphology and architecture of the as-prepared materials were explored using scanning electron microscopy (SEM) and transmission electron microscopy (TEM). Additionally, Inductive coupled plasma emission spectrometer (ICP) was utilized to analyze the elements content of the spent-LFP (S-LFP).

Electrochemical Characterization.

The assembly of half cells involved utilizing spent LFP as the active material. CR2016-type half coin cells were assembled in an argon-filled glovebox. In these cells, the counter electrode was lithium metal, and the electrolyte consisted of 1 M LiPF6 in a mixture of ethylene carbonate, diethyl carbonate, and dimethyl carbonate (EC: DMC: DEC = 1:1:1). The Celgard 2400 membrane was used as the separator. However, for the electrochemical tests presented in Figure 5, the separator was replaced with Li_2CuO_2@PP. The active materials (spent LFP), polyvinylidene difluoride (PVDF), and conductive carbon black particles were mixed to create a slurry (with a ratio of active materials to conductive carbon black to PVDF binder of 8:10wt:10wt %). This slurry was then evenly distributed onto Al foil to prepare working electrodes. When assembling full cells, the anode was replaced with graphite. In parallel, the active materials (graphite), carboxymethyl cellulose (CMC, binder), and conductive carbon black particles (conductive agent, radius of 30 nm) were mixed to create a slurry (with a ratio of active materials to conductive carbon black to CMC binder of 95 wt%: 2 wt%: 3 wt%). In this study, a battery cycler (BT2000) was utilized to investigate the galvanostatic charge and discharge tests, rate performances, and electrochemical behaviors of the as-prepared samples.

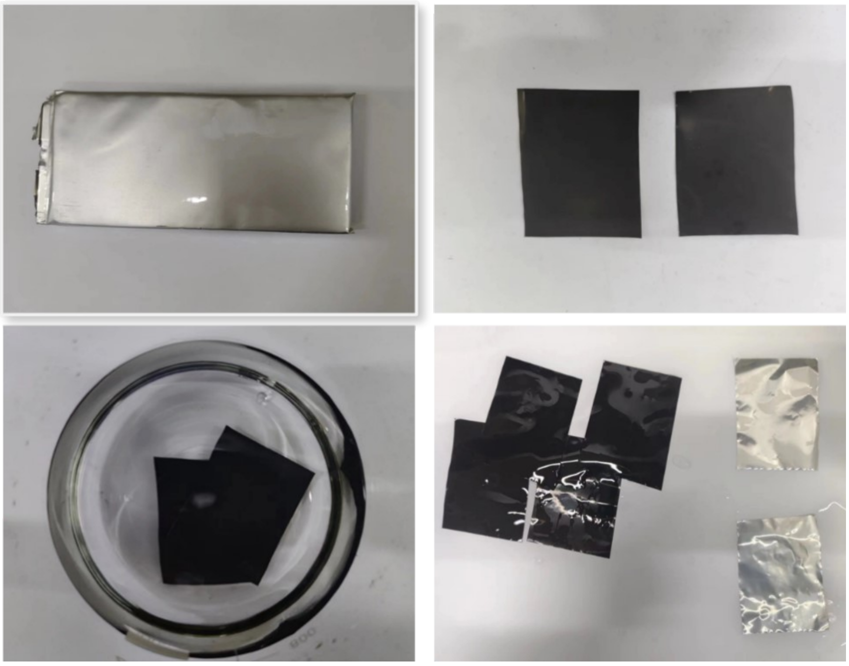

Figure S1. The images of cathode-separation process in this work.

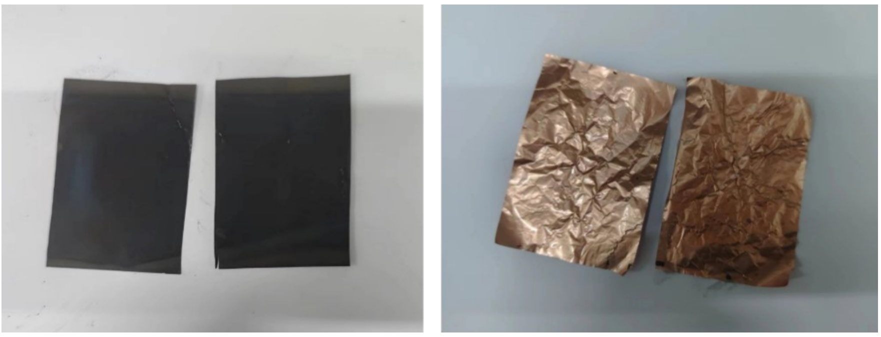

Figure S2. The images of spent anode sheets and Cu foil.

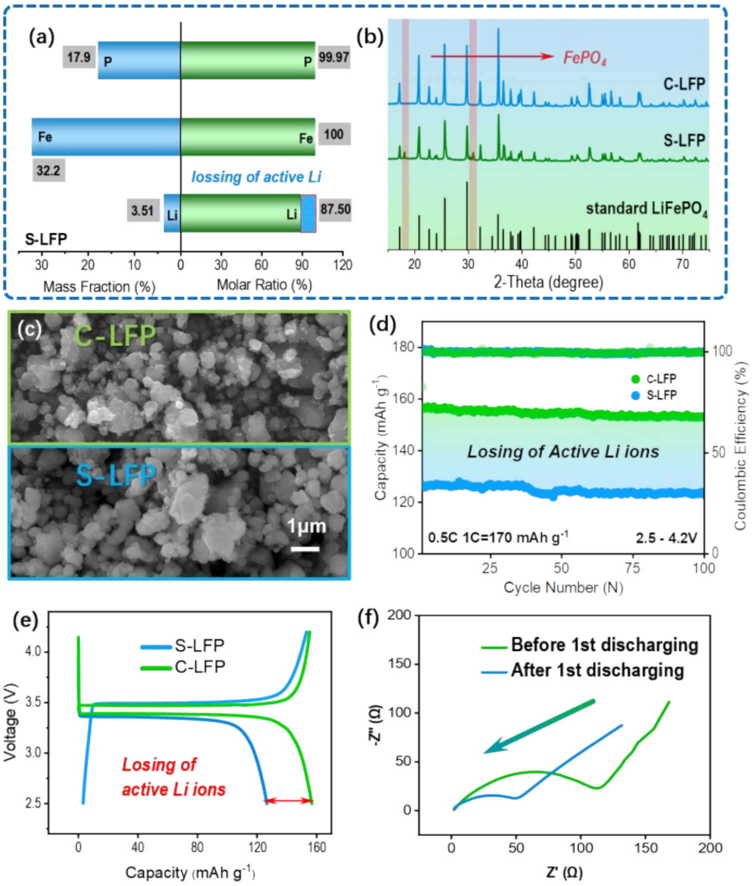

Figure S3. (a) The mass fraction and molar ratio of S-LFP. (b) The x-ray diffraction (XRD) results of C-LFP and S-LFP. (c) The SEM images of C-LFP and S-LFP. (d) The electrochemical comparison of C-LFP and S-LFP at 0.5 C (1.0 C = 170 mAh g^{-1}). (e) The first charging/discharging curves of C-LFP and S-LFP at 0.5 C (1.0 C = 170 mAh g^{-1}). (f) The Nyquist plots before first discharging process and after first discharging process of S-LFP.

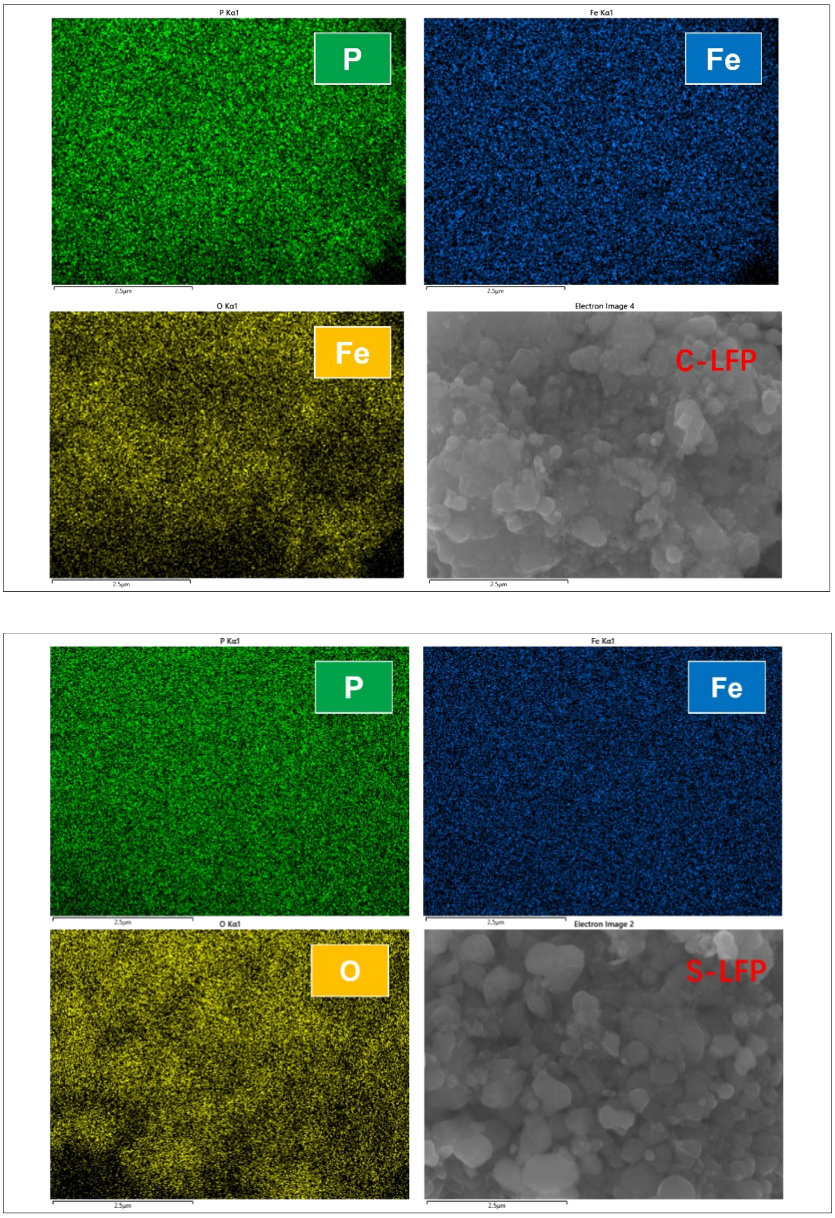

Figure S4. The EDS mapping images of C-LFP and S-LFP.

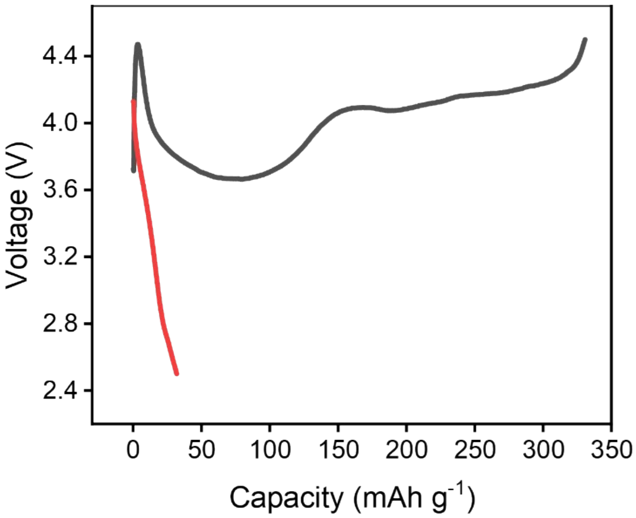

Figure S5. The 1st charging/discharging curves about Li_2CuO_2 at the current density of 1.0 C.

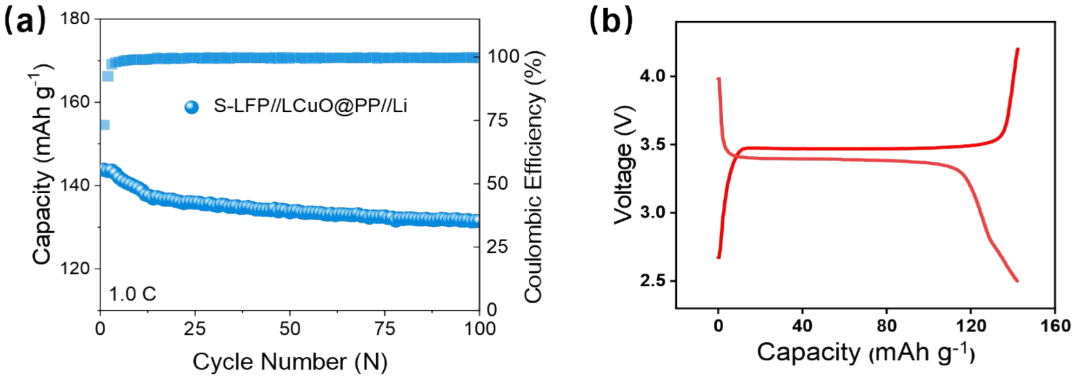

Figure S6. (a) The electrochemical performance of S-LFP//Li_2CuO_2@PP//Li battery at 1.0 C. (b) The 10th charging/discharging curves of S-LFP//Li_2CuO_2@PP//Li battery at 1.0 C.

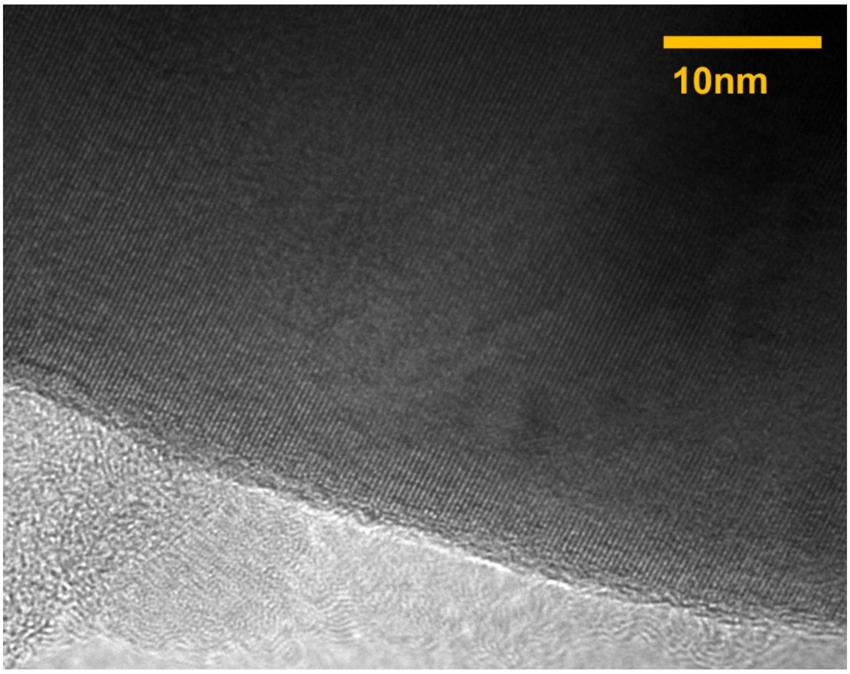

Figure S7. The TEM image of as-regenerated $LiFePO_4$.