Electronic Supplementary Information (ESI)

Sustainable Lithium Recovery from Spent $LiFePO_4$ Cathode via $K_2S_2O_8$-Assisted Solid State Oxidation Through Mechanochemistry

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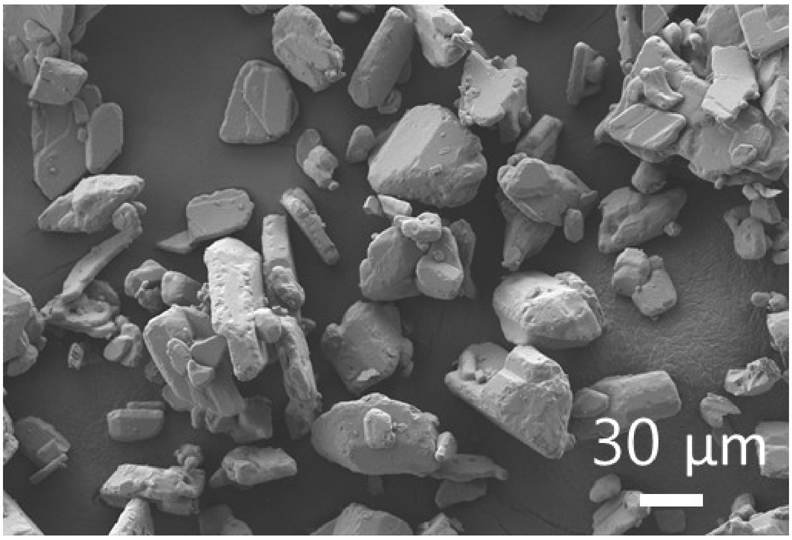

Fig. S1 SEM image of commercial potassium persulfate ($K_2S_2O_8$).

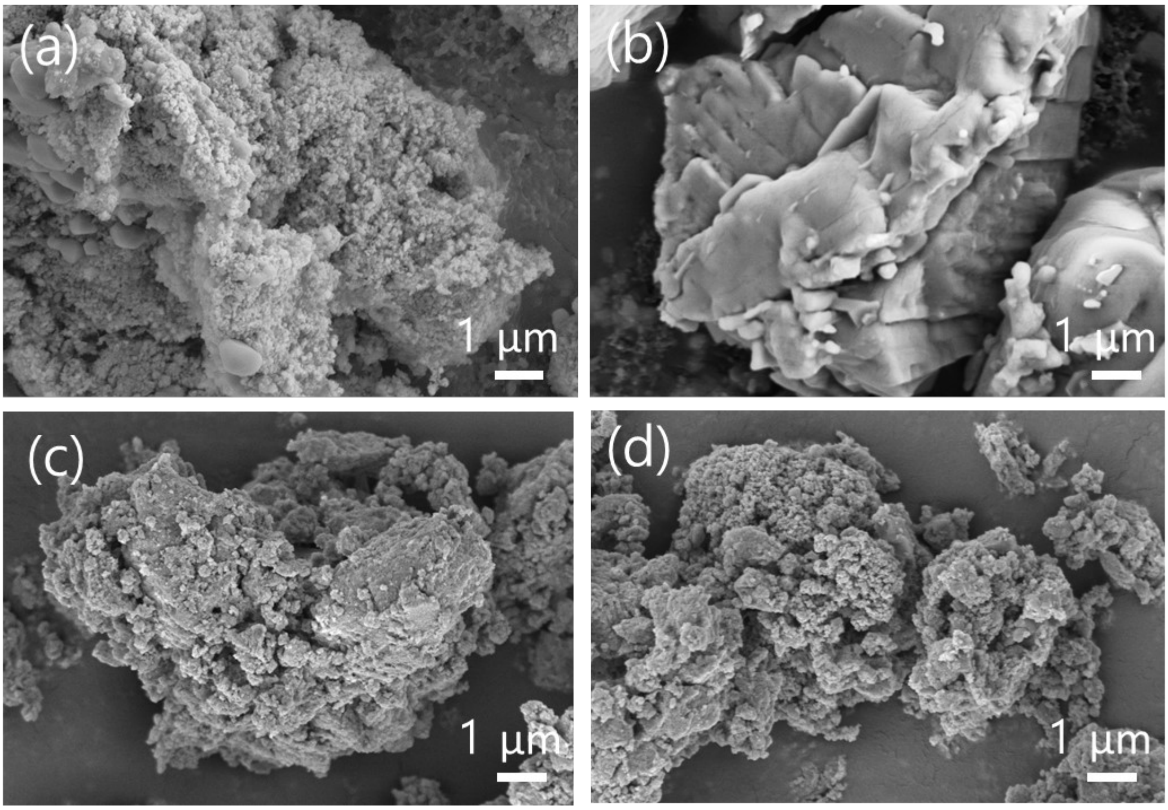

Fig. S2 SEM images of (a) black powder, (b) $K_2S_2O_8$, as-milled powders with milling process time of (c) 10 min and (d) 20 min, respectively.

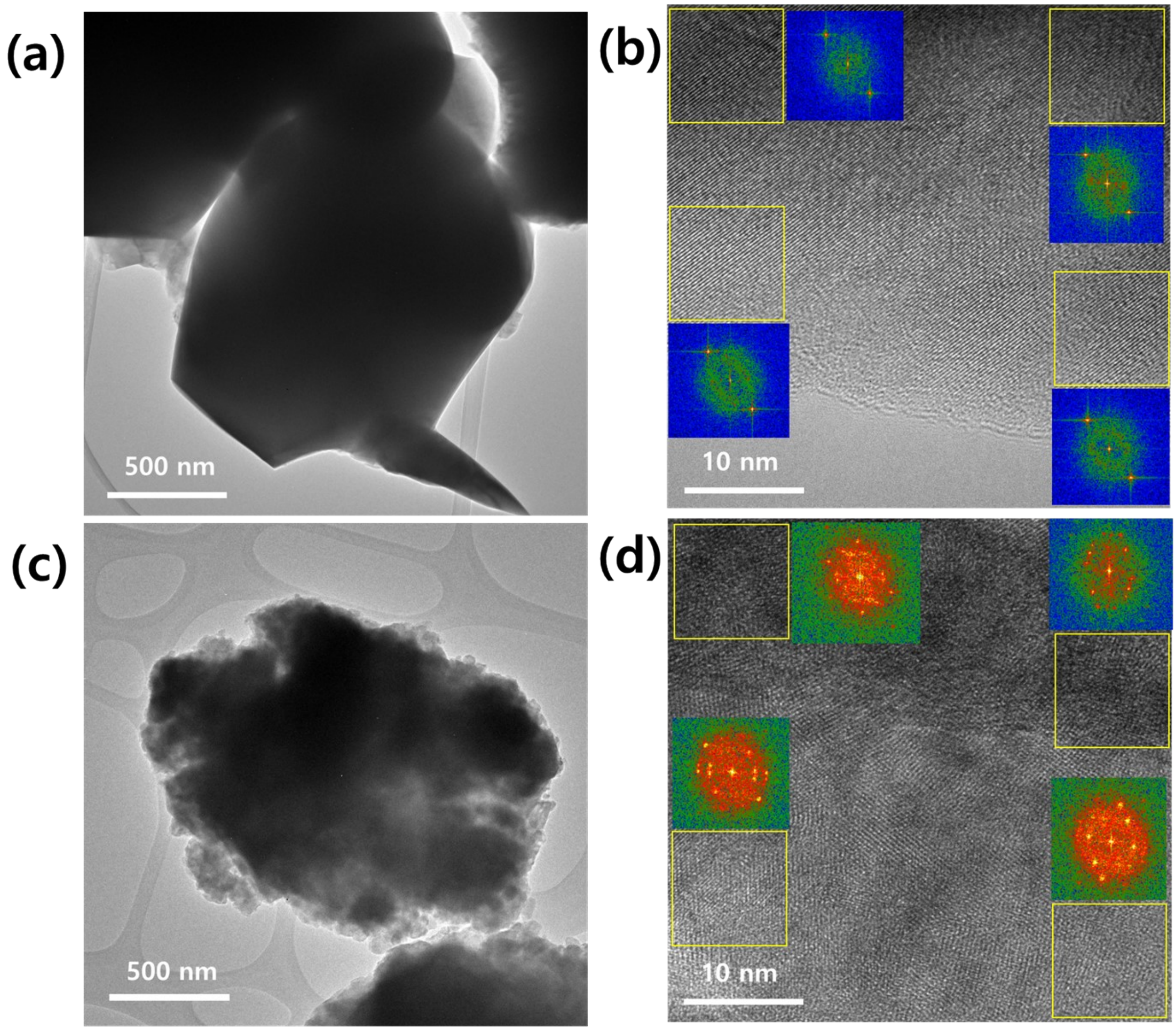

Fig. S3 (a,c) Low-magnification TEM images and (b,d) HR-TEM images with FFT patterns of before and after 30 min milling LFP particles, respectively.

Table. S1 Market price comparison of $Na_2S_2O_8$, $K_2S_2O_8$, Na_2SO_4, and K_2SO_4.

Category	$Na_2S_2O_8$	$K_2S_2O_8$	Na_2SO_4	K_2SO_4
Market price (USD kg^{-1})	0.955[1]	1.55[1]	0.223[2]	0.56[2]

[1] Oxidant prices were roughly estimated from Made-in-China supplier

[2] V. Y. C. Tatad, C. Tassel and J. Olchowka, ChemRxiv, 2025, DOI: 10.26434/chemrxiv-2025-lzph8-v2.

Table. S2 Comparison of environmental and circularity aspects of Na_2SO_4 and K_2SO_4.

Category	Parameters	Na_2SO_4	K_2SO_4
Environmental impact[3]	Aquatic toxicity (*L. minor*, Chronic EC50, 7d)	4.2 mS cm^{-1}	11 mS cm^{-1}
	Aquatic toxicity (*P. subcapitata*, Chronic EC50, 96h)	2.6 mS cm^{-1}	5.8 mS cm^{-1}
Resource circularity	Market demand	Accumulates as industrial waste due to low market demand	Directly reusable as an agricultural fertilizer

(Note: EC50 stands for Effective Concentration, representing the concentration that reduces 50% of the population. The unit mS cm^{-1} (milli Siemens per centimeter) indicates the electrical conductivity of the salt solutions. A lower EC50 value means that the substance exerts toxic effects at a lower concentration, indicating higher toxicity.)

Compared with Na_2SO_4, which exhibits relatively high toxicity to aquatic ecosystems, K_2SO_4 is significantly more environmentally benign. According to relevant ecotoxicological data[3], the chronic EC50 of Na_2SO_4 for the aquatic macrophyte Lemna minor and the green alga Pseudokirchneriella subcapitata are 4.2 mS cm^{-1} and 2.6 mS cm^{-1}, respectively. In contrast, K_2SO_4 shows a substantially lower toxicity, requiring much higher concentrations (11.0 mS cm^{-1} and 5.8 mS cm^{-1}, respectively) to induce the same detrimental effects. Furthermore, from an economic and resource circularity perspective, the conventional generation of Na_2SO_4 leads to severe waste accumulation issues due to the significant imbalance between battery industry growth and traditional market demand. Conversely, K_2SO_4 (valued at approx. USD 560 ton^{-1}) can be directly utilized as a high-value fertilizer for chloride-sensitive crops[2]. This indicates that the proposed process employing $K_2S_2O_8$ is not only technically efficient but also offers a highly sustainable closed-loop solution."

[3] J. A. Simmons, Environ. Toxicol. Chem., 2012, 31, 1370-1374.