Supporting Information

Selective Extraction of Lithium from a Spent Lithium Iron Phosphate Battery by Mechanochemical Solid-phase Oxidation

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Content

Table S1

Table S1 The metal impurities of the obtained Li_3PO_4 product (wt.%)

Element	Percentage (wt.%)
Li_3PO_4	98.50
Na	1.20
Fe	0.21
Al	0.07
Cu	0.02

Figure S1

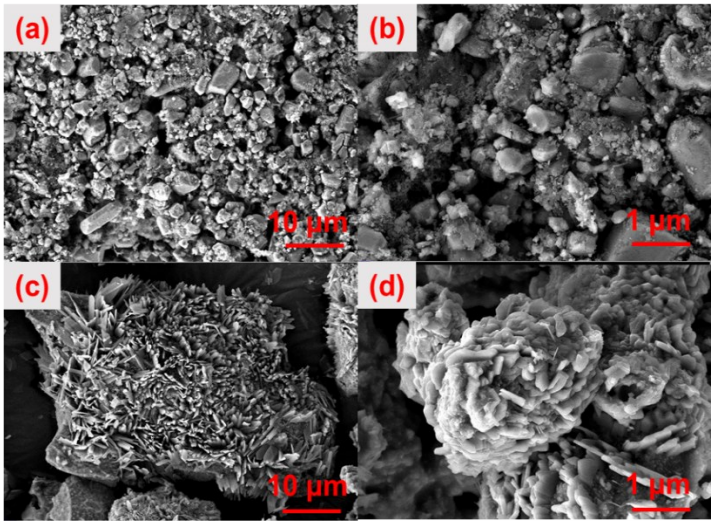

Figure S1 SEM results of $Na_2S_2O_8$/$LiFePO_4$ samples (a-b) before and (c-d) after mechanochemical reaction (reaction time of 5.0 min, $Na_2S_2O_8$/$LiFePO_4$ mass ratio of 2:1 and rotary speed of 600 rpm).

Figure S2

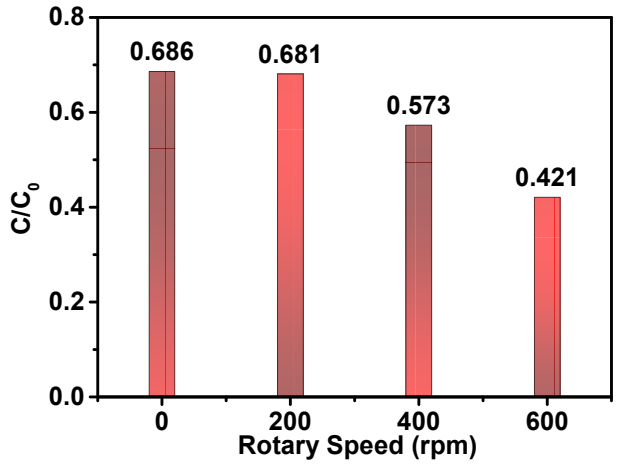

Figure S2 Residual ratio of the $Na_2S_2O_8$ after mechanochemical reaction at different speeds.

A chemical oxidation reaction occurs between $Na_2S_2O_8$ and the reduced lithium that has been released, resulting in the formation of $LiNaSO_4$ and the decomposition of $Na_2S_2O_8$. In the process of mechanochemical reaction, C species as a reducing agent can participate in the consumption reaction of the solid-phase oxidant $Na_2S_2O_8$ under the induced action of mechanical force. In addition, $Na_2S_2O_8$ is not stable in aqueous solutions, and the consumption reactions can occur. The reason is that a small amount of excess $Na_2S_2O_8$ will oxidize H_2O and lead to an acidification of solution, which can be illustrated by the following chemical equation:

$$2H_2O + 2Na_2S_2O_8 \rightarrow 2Na_2SO_4 + 2H_2SO_4 + O_2\uparrow \quad \text{(S1)}$$

Figure S3

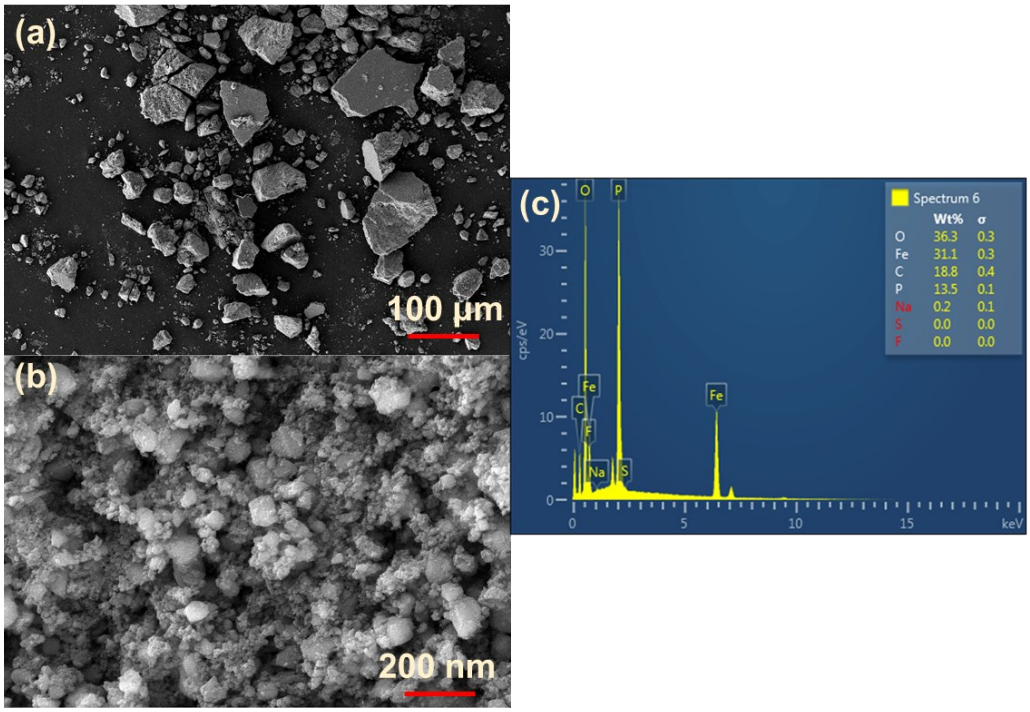

Figure S3 SEM-EDAX result of the obtained $FePO_4$ product.

Figure S4

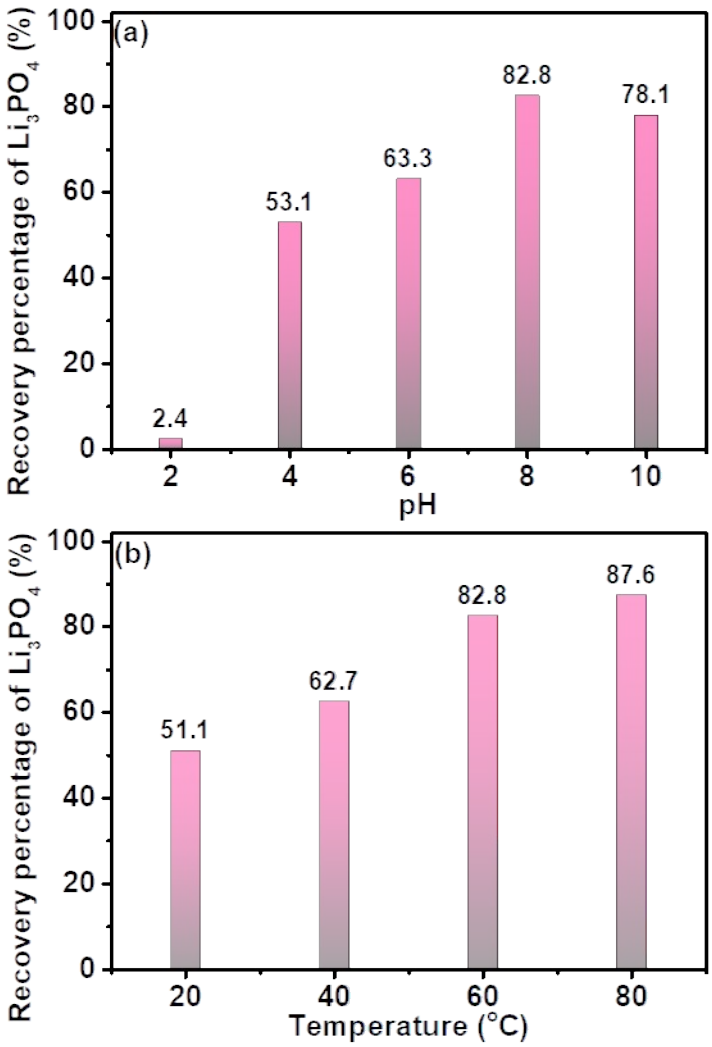

Figure S4 The influence of different factors: (a) pH and (b) temperature on the recovery percentage of Li_3PO_4. (Conditions: (a) Na_3PO_4:Li_2SO_4 molar ratio = 3:4, temperature = 60 °C; (b) Na_3PO_4:Li_2SO_4 molar ratio=3:4, pH = 8).

Experimental procedure:

To accurately quantify the lithium extraction process, a simulated solution of Li_2SO_4 (31.7 g/L) and Na_2SO_4 (154.5 g/L)) was used to calculate the recovery percentage of Li_3PO_4. The solution was first heated at the set temperature and adjusted the pH using

NaOH or H_2SO_4 solution. Subsequently, Na_3PO_4 was added into the simulated solution according to a molar ratio of 3:4 with a stirring time of 1h. The recovery percentage of lithium was calculated based on the actual precipitation percentage and theoretical precipitation percentage of Li_3PO_4.