# **Supporting Information**

### Large-scale direct regeneration of LiFePO<sub>4</sub>@C based on spray

### drying

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**Fig. S1.** Photograph of (a) spent, (b) homogenized, (c) spray-dried and (d) regenerated LiFePO<sub>4</sub>



Fig. S2. The first turn charge and discharge curve of (a) S-LFP and (b) R-LFP



Fig. S3. The first turn charge and discharge curve of (a) R-LFP-without

homogenization and (b) R-LFP



Fig. S4. (a) Charge-discharge curves and (b) Long-term cycling stability of R-LFP-

without homogenization.



Fig. S5. Process flow diagrams of (a) LFP materials preparation (b) LFP recycling



Fig. S6. The cost comparison between R-LFP and solid state synthesis LFP

XPS elemental composition
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Mass ratio /%	Fe	0	Р	С
S-LFP	4.23	32.39	5.34	58.04
Homo-LFP	5.11	38.86	7.34	48.69
R-LFP	1.31	14.61	1.12	82.96

### Table S2

### ICP-MS results of spent lithium iron phosphate

Elemental types	Maga function (mtl)	Amount of substance per		
	Mass fraction (wt%)	kg (mol/kg)		
Fe	34.53%	6.183935		
Li	4.41%	6.356335		
Р	15.58%	6.029794		

### Table S3

### The raw materials cost breakdown of R-LFP and solid state synthesis LFP

Raw materials cost of production of 1 ton R-LFP			Raw materials cost of production of 1 ton of LFP via solid state				
raw materials	unit price ( *10 <sup>4</sup> yuan)	quantity (ton)	cost ( *10 <sup>4</sup> yuan)	raw materials	unit price ( *10 <sup>4</sup> yuan)	quantity (ton)	cost ( *10 <sup>4</sup> yuan)
S-LFP	6.71	1	6.71	iron phosphate (FP)	2.45	0.956	2.3422
Li <sub>2</sub> CO <sub>3</sub>	50.5	0.01	0.505	Li <sub>2</sub> CO <sub>3</sub>	50.5	0.234	11.817
glucose	0.27	0.07	0.0189	glucose	0.27	0.07	0.0189
SUM			7.2339	SUM			14.1781

# Economic and Environment Analysis

We have carefully considered the cost of this regeneration method. The current market price of LFP raw material is ¥157,500 per ton, the price of S-LFP black powder is ¥66,100 per ton, the price of lithium carbonate is ¥505,000 per ton, the price of iron phosphate (FP) is ¥24,500 per ton, and the price of food grade glucose is ¥2,700 per ton. To obtain 1 ton of recycled lithium iron phosphate, we need approximately 1t of S-LFP black powder, 0.01t of lithium carbonate, and 0.07t of glucose. After calculation, the cost of regenerating 1t of lithium iron phosphate material is ¥72,339 per ton, which is approximately 51.02% of the marketed LFP. The regeneration process is also close to the current lithium iron phosphate manufacturing process, enabling direct use of existing equipment for production and reducing upfront investment costs, with the potential for industrial production.