Sustainable Battery Recycling via Direct Regeneration of Lithium Iron Phosphate Cathodes

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Lithium iron phosphate ($LiFePO_4$, LFP) is a widely used cathode material in lithium-ion batteries due to its stability and safety. However, repeated charge and discharge cycling cause the lithium loss (delithiation) and degrade the battery performance, eventually producing battery wastes. The conventional battery recycling methods include pyrometallurgy, which requires high-temperature smelting and consumes a large amount of energy, and hydrometallurgy, which relies on strong acid/alkali reagents and involves complex steps to obtain high-purity precursors. Despite these methods could relieve the recycling pressure in a short time, neither pyro- nor hydro-metallurgical routes can realize a closed-loop battery circular economy in an environmentally sustainable manner. In the present study, we focus on regenerating delithiated $LiFePO_4$ (D-LFP) using a direct regeneration approach via solid-state reaction, yielding regenerated $LiFePO_4$ (R-LFP). Direct regeneration is a method to replenish the components and repair its structure to its original state, without changing the morphology. This method offers significant advantages, making it an attractive option for sustainable recycling. As shown in Fig. 1, the XRD characterization demonstrated that the crystal structure of D-LFP cathode materials was repaired. Additionally, the ICP measurements exhibited a Fe:Li ratio of 1:1.2 on R-LFP, verifying the successful restoration of Li from D-LFP (Fe:Li ratio of 1:0.02). The battery cycling tests in Fig. 2 presented that R-LFP achieved the specific discharge capacity of 110 mAh/g^{-1} at 0.2 C, 102 mAh/g^{-1} at 1 C, 80 mAh/g^{-1} at 5 C and 60 mAh/g^{-1} at 10C, respectively, indicating that approximately 70% of the pristine LFP capacity was recovered. These results highlight the potential of solid-state reaction for efficient and sustainable regeneration of spent LFP, offering a viable solution for recycling and extending the life of lithium-ion batteries.

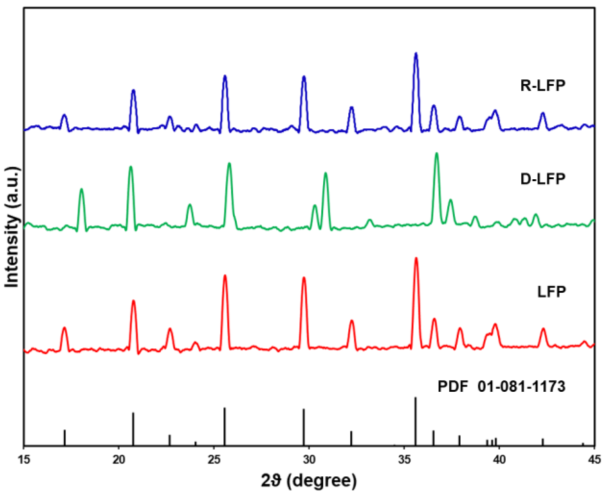

Fig.1 XRD patterns of the LFP, D-LFP and R-LFP

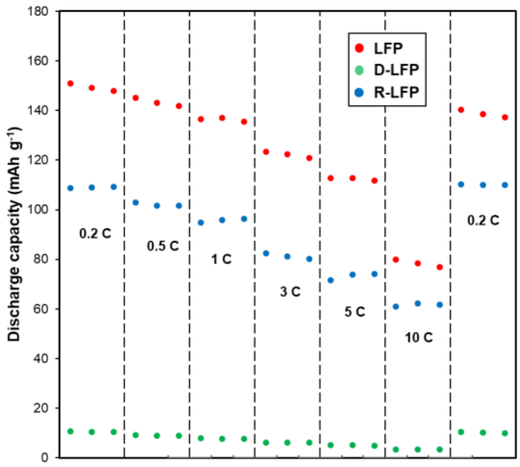

Fig.2 Rate capabilities of the LFP, D-LFP and R-LFP