Supporting Information

$LiFePO_4$/C Twin Microspheres as Cathode Materials with Enhanced Electrochemical Performance

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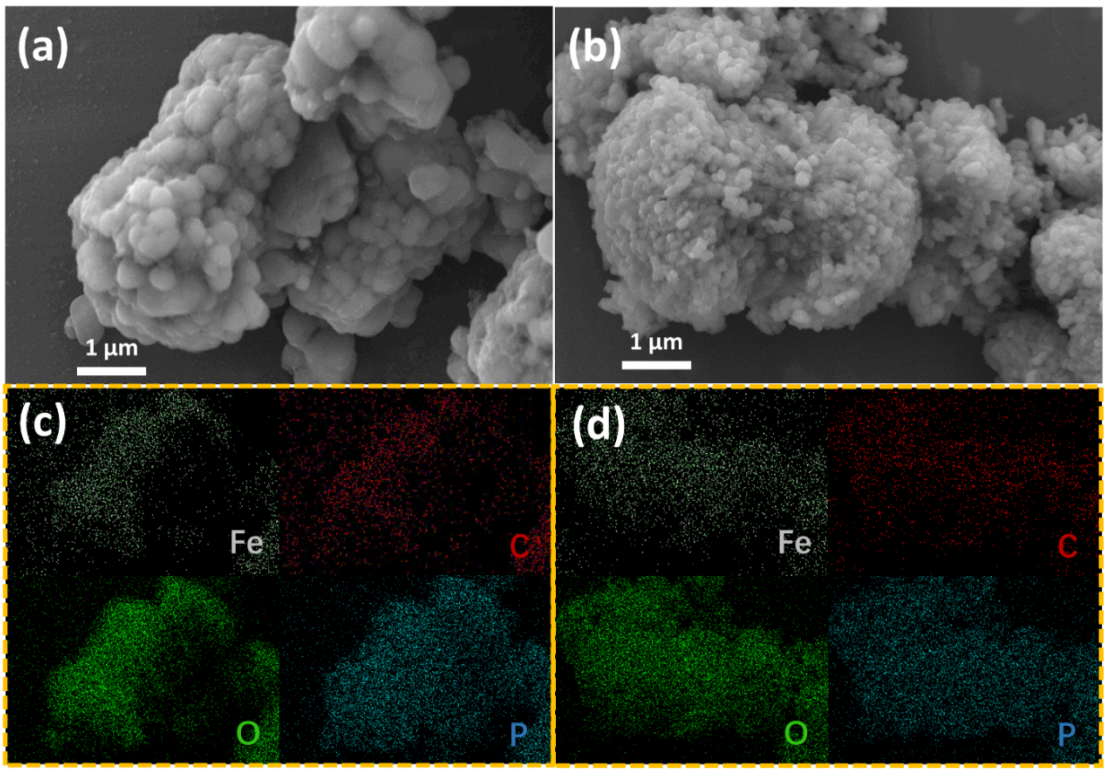

Fig. S1. SEM images (a, b) and corresponding EDS mapping (c, d) of LFP/C-40, LFP/C-80;

SEM images in Fig. S2 showed that most of the $LiFePO_4$/C-60 samples appeared as twin microsphere structure with slightly different size. In addition, a small number of twin microspheres were broken during ultrasound process during sample preparation.

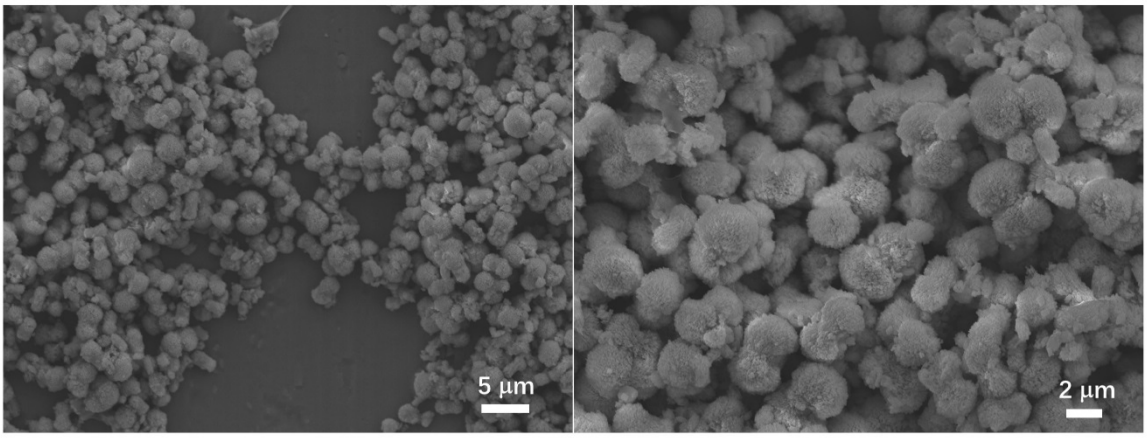

Fig. S2. SEM images of $LiFePO_4$/C-60.

The $LiFePO_4$/C-60 twin microspheres present a hierarchical structure assembled by nanoparticles. The hierarchical structure facilitates the rapid and effective transport of matter and charge due to its unique geometric structure[1]. Twin microspheres structures have some advantages. Firstly, the transport of lithium ions in lithium iron phosphate occurs on one-dimensional channels. When there are two or more point defects in the diffusion channel, Li^+ cannot enter the material through any side of the tunnel opening, resulting in low utilization of active materials[2]. Nano-sized primary particles promote the transport of Li^+ ions by reducing the diffusion distance, improve the utilization of active lithium ions, and smooth the diffusion path to ensure high capacity and rate

capability[2]. Secondly, in practical applications, the low ion diffusion rate is a key factor in the performance of $LiFePO_4$[3]. The porous structure made of uniformly distributed primary particles allows the electrolyte to penetrate finely into the internal active material. The hierarchical structure contributes to the effective contact between $LiFePO_4$ particles and electrolyte, thereby reducing polarization[4]. Thirdly, electrodes made of nanoscale materials are prone to collapse because they cannot be stacked as densely on collectors as micron materials. The micron-sized and robust secondary porous microstructure framework can effectively buffer the aggregation of particles. The micron size and stable secondary porous microstructure can effectively alleviate the aggregation of particles[5]. Fourthly, the large surface area and porous structure of nanomaterials usually result in lower tap density, which leads to lower bulk density of the electrode loaded with nanomaterials [2]. Fourthly, the large surface area and porous structure of nanomaterials usually result in lower tap density, which leads to lower bulk density of the electrode loaded with nanomaterials [6]. In summary, the twin microspheres hierarchical structure integrates the advantages of microsphere particles, porosity, nanocrystallization and carbon coating, ensuring excellent electrochemical performance and good processing performance. Therefore, this twin microsphere structure is a key factor in lithium $LiFePO_4$/C samples.

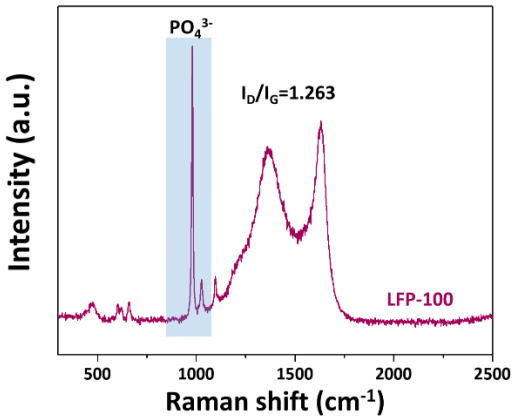

Fig. S3. Raman spectrum of LFP-100;

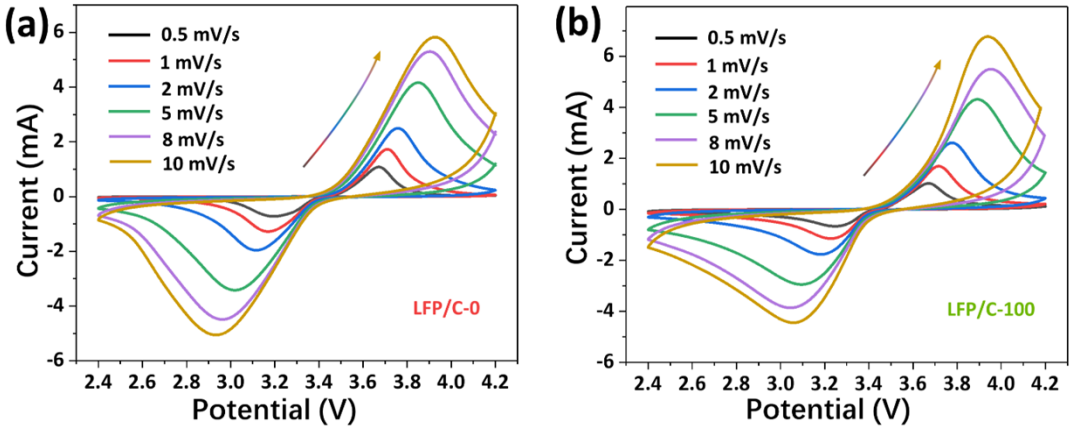

Fig. S4. CV curves of LFP/C-0 (a), and LFP/C-100 (b) with different scanning rate between 0.5mV/s and 10mV/s;

High loading increases the ratio of active materials to inactive packaging materials, but it may cause a decrease in discharge capacity[2]. In order to explore the influence of loading amount, the medium (4 mg cm^{-2}) and high (8 mg cm^{-2}) loading were assembled into button batteries. The discharge capacity was tested at different discharge rates at room temperature, as shown in Fig. S5. The discharge capacity of LFP-60 with medium load and high load reached 146.6 $mA{\cdot}h{\cdot}g^{-1}$ and 135.1 $mA{\cdot}h{\cdot}g^{-1}$

at 0.2 C discharge rate, while the discharge capacity of LFP-0 is 68.0 mA h g^{-1}, 66.3 mA h g^{-1}, and the discharge capacity of LFP-100 is 96.9 mA·h·g^{-1}, 92.4 mA·h·g^{-1}, respectively. The high-loading discharge performance of LFP/C-60 is significantly better than the other two groups of samples. The three-dimensional conductive network constructed in the twin microspheres structure may be the reasons for its excellent performance.

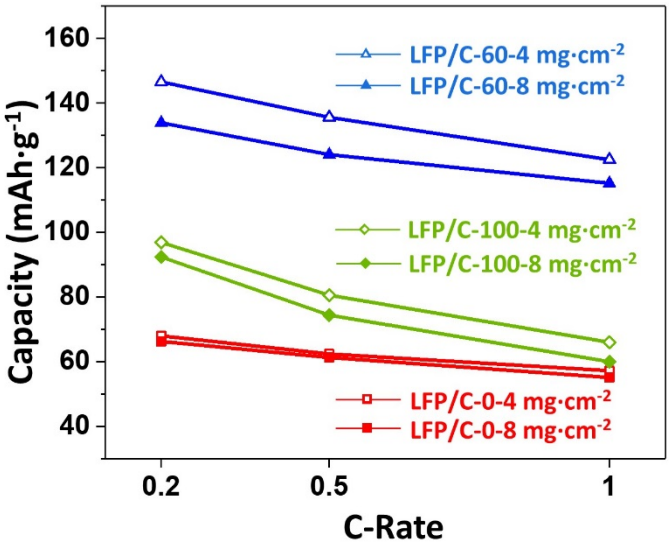

Fig. S5. Discharge capacity of LFP/C-0, LFP/C-60, LFP/C-100 at different C-rates with medium and high loading.

References

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