

## Supplementary Information

### **Novel layered $\text{Li}_3\text{V}_2(\text{PO}_4)_3/\text{rGO}\&\text{C}$ sheets as high-rate and long-life lithium ion battery cathode**

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## Experimental

### **Materials Synthesis**

#### **Synthesis of Graphene oxide nanosheets (GO-NSs)**

Graphene oxide (GO) was prepared using the famous Hummers method as previously reported.<sup>1</sup> Briefly, the graphite (1.0 g) was mixed with concentrated sulfuric acid (23 mL) with stirring for 24 hours at room temperature. Then the flask was placed in a water bath at 40°C and  $\text{NaNO}_3$  (0.1 g) was added to the suspension and allowed to dissolve for 5 minutes. This step was followed by slow addition  $\text{KMnO}_4$  (1.5 g), keeping the temperature below 45°C. The solution was stirred for 30 minutes. Next, deionized (DI) water were added to the beaker, followed by another DI water (3 mL) after 5 minutes. In another 5 minutes, another DI water (40 mL) were added. After 15 minutes later, the reaction was taken out from water bath and DI water (140 mL) and 30%  $\text{H}_2\text{O}_2$  (10 mL) were added to end the reaction. The suspension was then repeatedly centrifuged and washed twice with 5% HCl solution and then repeatedly with water. The collected precipitate was dispersed in DI water (100 mL) and sonicated for 60 minutes. After 5000 rpm centrifuged for 5 minutes, a brown homogeneous GO-NSs supernatant was obtained.

#### **Synthesis of layered LVP/rGO&C sheets**

A mole ratio of 1.5:2:3 for lithium carbonate ( $\text{Li}_2\text{CO}_3$ ), vanadium (III) acetylacetonate ( $\text{V}(\text{C}_5\text{H}_7\text{O}_2)_3$ ) and ammonium dihydrogen phosphate ( $\text{NH}_4\text{H}_2\text{PO}_4$ ) were dissolved in 40

mL deionized (DI) water under stirring overnight to obtain homogenous solution. And then the solution was transferred into a 50 mL Teflon-lined stainless steel autoclave and kept at 180 °C for 6 hours. Subsequently, a jelly-like  $\text{Li}_3\text{V}_2(\text{PO}_4)_3$  (LVP) gels were obtained. Then, 3 mL GO-NSs solution ( $3 \text{ mg mL}^{-1}$ ) was added into the LVP gels under vigorous stirring until get a stable dispersion. After freeze-drying process, the LVP gels/GO sheets were obtained. The dried product was annealed at 750 °C for 8 hours with a heating rate of  $2 \text{ }^\circ\text{C min}^{-1}$  in  $\text{Ar}/\text{H}_2$  (95:5 in vol.) and finally the layered LVP/rGO&C sheets were obtain.

### ***Synthesis of LVP/C sheets***

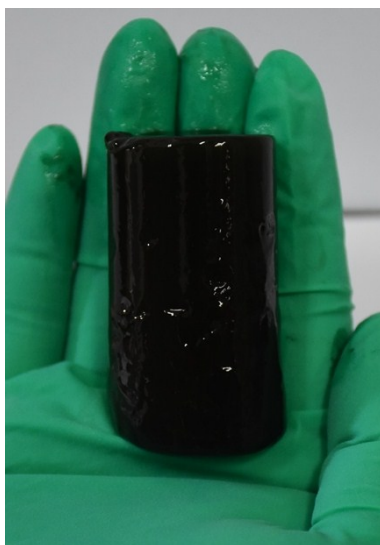
As control experiments, the LVP/C composites were prepared through the same process without adding GO-NSs.

### ***Material Characterization***

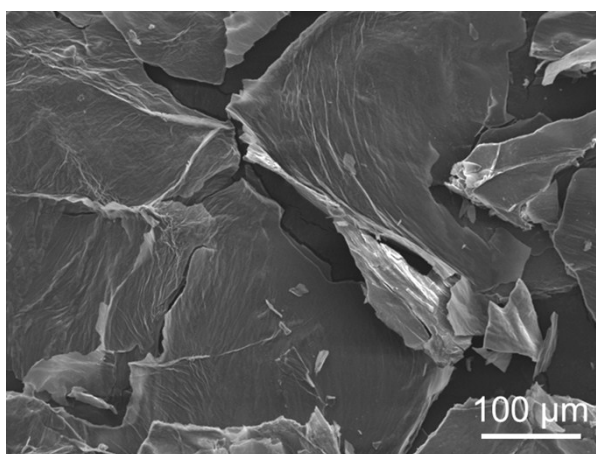
Zeta potential (Zetasizer 3000HS) was measured to characterize the effective surface charge of the samples. XRD (Burker D8 Advanced X-ray diffractometer with  $\text{Cu-K}\alpha$  radiation) was employed to characterize the structural properties of the samples. CHN elemental analyzer was involved in determining the carbon contents in the as-prepared samples. Raman spectra were obtained using a Renishaw INVIA micro-Raman spectroscopy system. SEM images were collected with a JEOL-7100F microscopy. TEM, HRTEM, STEM-HAADF images and STEM-DEX mapping were recorded by using Titan G2 60-300 with image corrector. Brunauer-Emmet-Teller surface area was measured using Tristar II 3020 instrument by adsorption of nitrogen at 77 K.

*Electrochemical Characterization:* The electrochemical properties were evaluated by assembly of 2016 coin cells in a glove box filled with pure argon gas. The working electrodes were prepared by mixing the 70% active material, 20% acetylene black and 10% Poly(vinylidene Fluoride) (PVDF) with an appropriate amount of N-methyl-2-pyrrolidone (NMP) as a solvent binder. The slurry was cast onto Al foil by using a doctor blade and dried in a vacuum oven at 100 °C for 8 h. The mass loading is  $1.0\text{-}2.0 \text{ mg cm}^{-2}$ . In the coin cell, lithium was used as the anode, and 1 M  $\text{LiPF}_6$  dissolved in ethylene carbonate/dimethyl carbonate with a volume ratio of 1:1 was used as

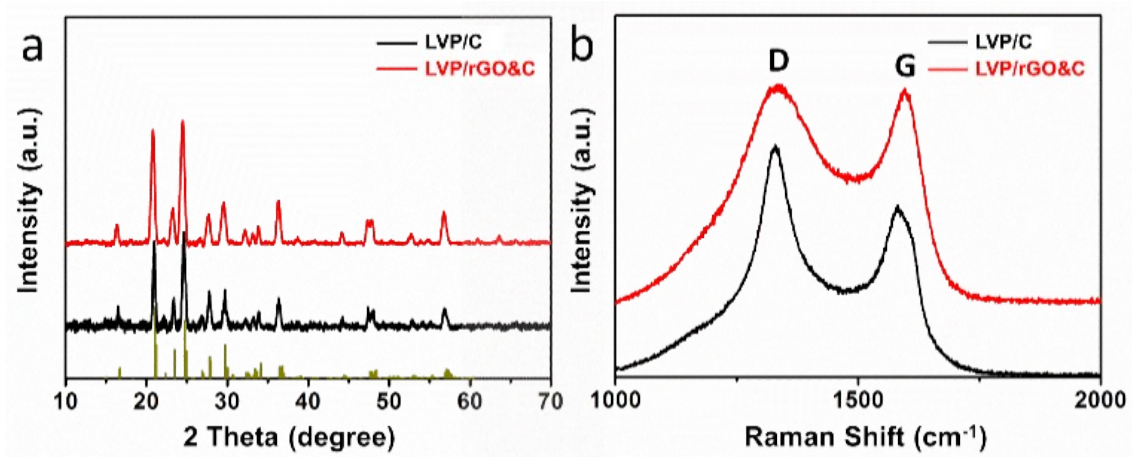
electrolyte. The galvanostatic charge–discharge cycling tests were carried out at 3.0–4.3 V by a battery test system (LAND CT2001A). For the rate test, the voltage window is set at 3–4.3 V for the rates lower than 20 C, and a voltage window of 3–4.8 V for 50, 80, and 100 C, while the charge rate and discharge rate are the same. Electrochemical impedance spectroscopies (EIS) were tested with an Autolab Potentiostat Galvanostat (Autolab PGSTAT 302N). All the measurements were carried out at room temperature.



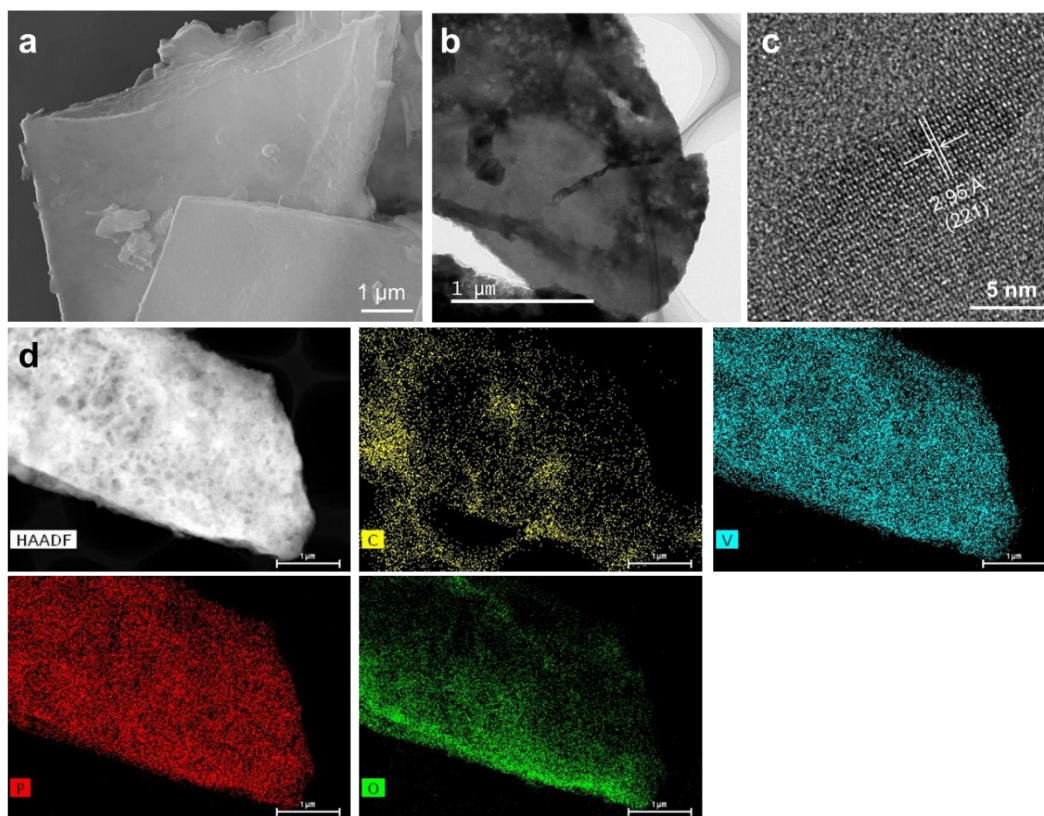
**Fig. S1** The photo of LVP jell-like gels, obtained after hydrothermal reaction.



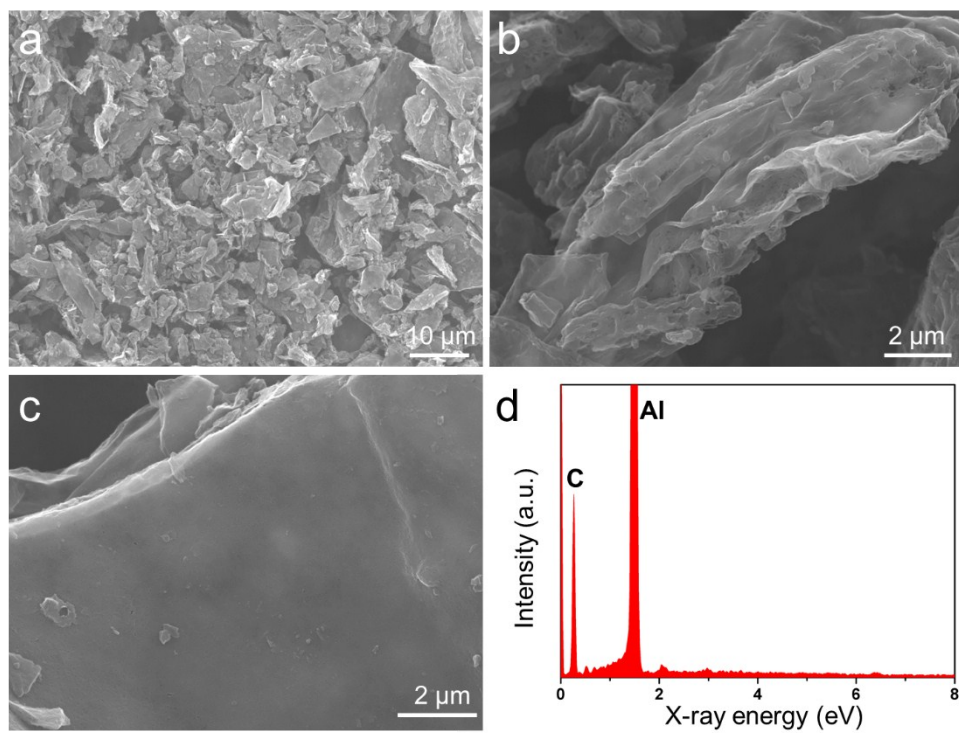
**Fig. S2** SEM image of LVP gels/GO sheets obtained after freeze-drying.



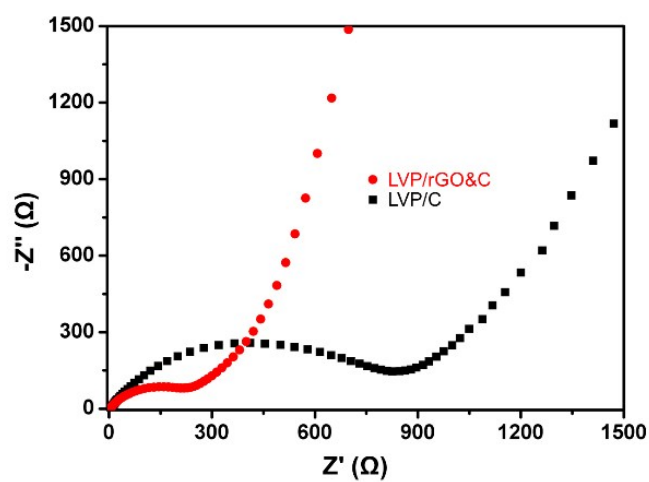
**Fig. S3** XRD patterns (a) and Raman spectra (b) of layered LVP/rGO&C and LVP/C sheets, respectively.



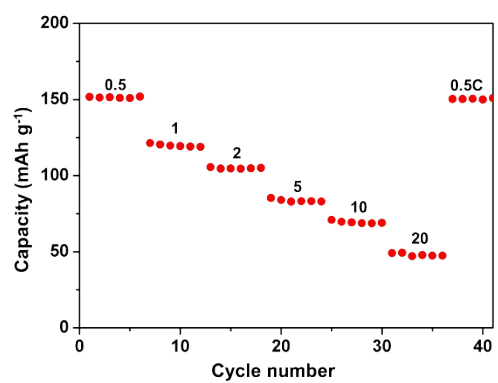
**Fig. S4** SEM image (a), TEM images (b) HRTEM image (c) and STEM-EDS mappings (d) of LVP/C sheets.



**Fig. S5** (a-c) SEM images of the residual carbon after removing the LVP in LVP/C sheets. (d) EDX spectrum of the residual carbon.



**Fig. S6** Nyquist plots of the LVP/rGO&C and LVP/C sheets.



**Fig. S7** Rate performance of commercialized LiFePO<sub>4</sub>/C powders.

## References

1. Y. Zhao, J. Feng, X. Liu, F. Wang, L. Wang, C. Shi, L. Huang, X. Feng, X. Chen, L. Xu, M. Yan, Q. Zhang, X. Bai, H. Wu and L. Mai, *Nature Commun*, 2014, **5**, 4565.