

Electronic Supplementary Information

A Rationally Assembled Graphene Nanoribbon/Graphene Framework for High Volumetric Energy and Power Density Li-Ion Batteries

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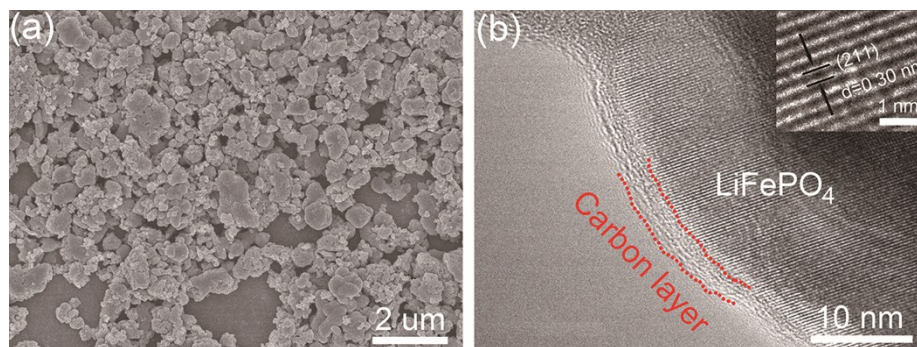


Fig. S1 (a) SEM and (b) TEM images of commercial LiFePO_4 used in this work.

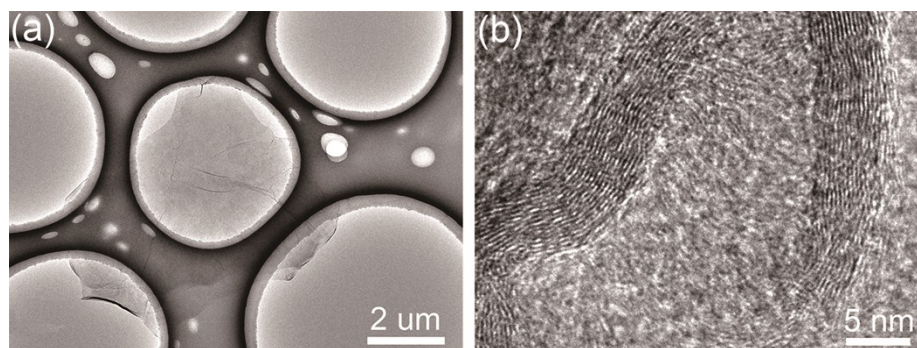


Fig. S2 (a) TEM image of a single piece of GO; (b) HRTEM image of the edges of reduced graphene oxide (rGO) showing 15~20 layers of graphene.

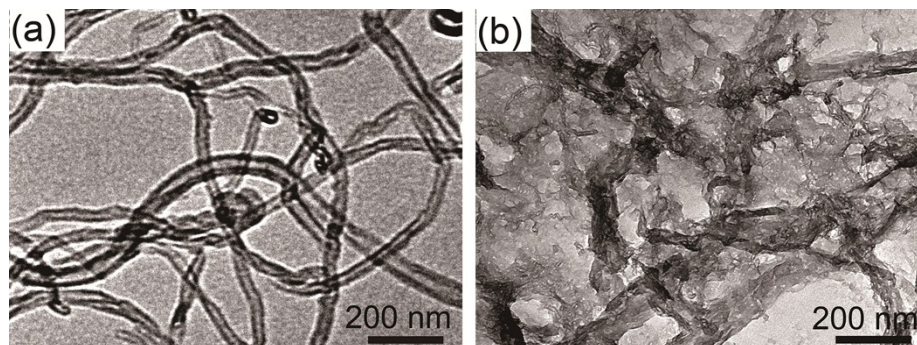


Fig. S3 TEM images of (a) CNT precursor (diameter ~50 nm) as well as resultant (b) GONR (width ~150 nm) obtained by a chemical oxidation method.¹

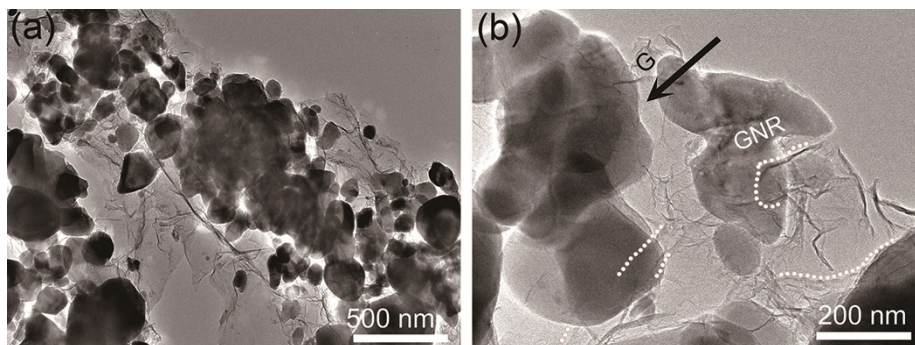


Fig. S4 TEM images of LFP/GNR/G sample. White dot lines in (b) outline some GNRs which bridge LFP particles and G sheets.

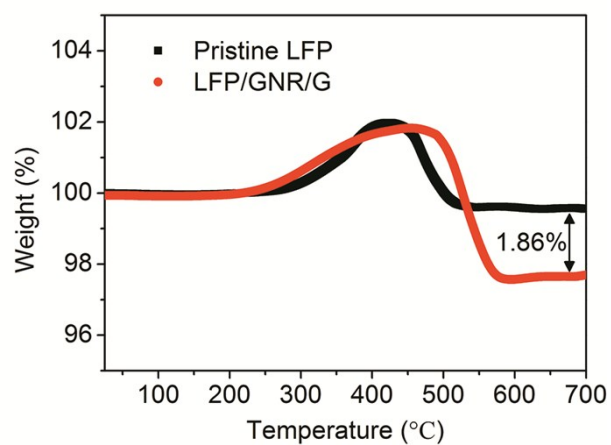


Fig. S5 Thermogravimetric curves of pristine LFP and LFP/GNR/G. The GNR/G content in LFP/GNR/R was 1.86%, which confirmed the result of 2% in Table S2.

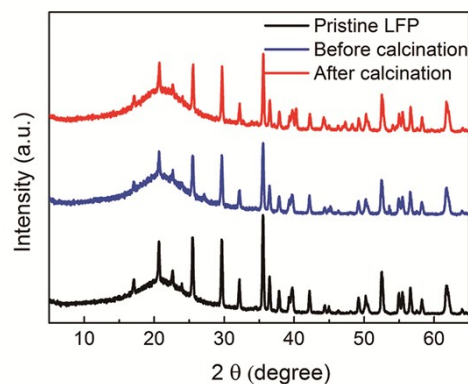


Fig. S6 XRD patterns of commercial LFP, as well as LFP/GNR/G sample before and after a calcination at 600°C in Ar/H_2 (90/10, V/V) for 5 h.

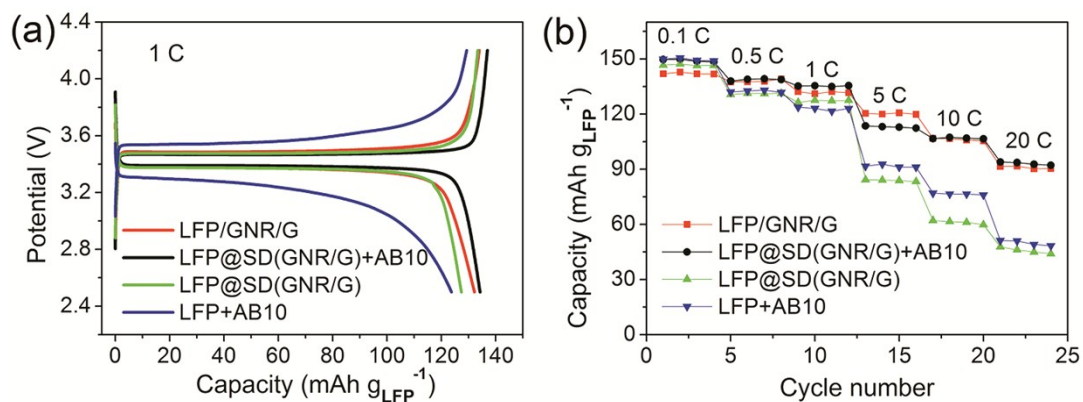


Fig. S7 Comparison of (a) charge/discharge curves at the current rate of 1 C, and (b) rate capabilities of LFP+AB10, LFP@SD(GNR/G), LFP@SD(GNR/G)+AB10 and LFP/GNR/G based on the commercial LFP weight only.

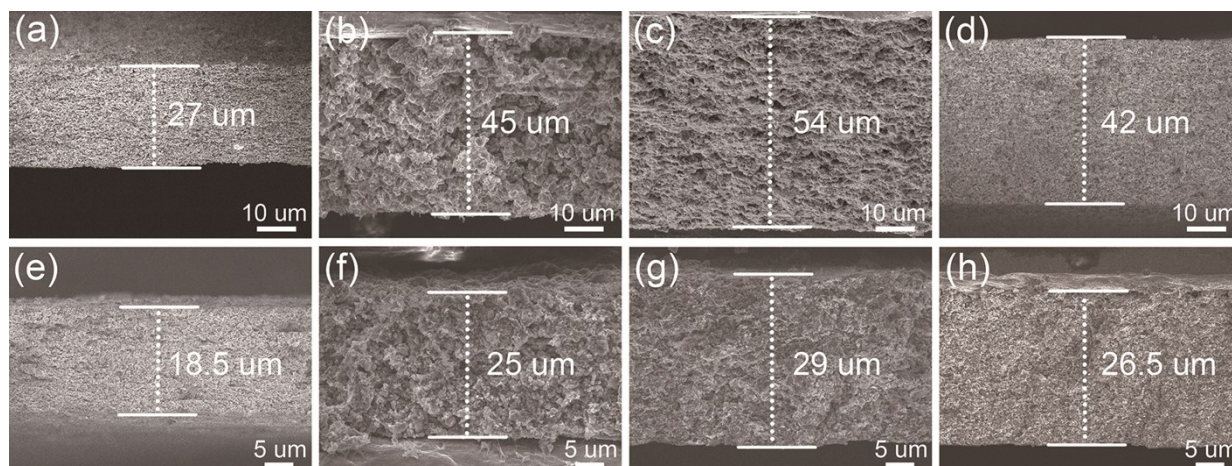


Fig. S8 Cross-sectional SEM images of four electrode samples with electrode weight of 5 mg cm^{-2} (including active material, conductive additive and binder) before and after compression under a pressure of 50 MPa: (a, e) LFP/GNR/G; (b, f) LFP@SD(GNR/G); (c, g) LFP@SD(GNR/G)+AB10; (d, h) LFP+AB10.

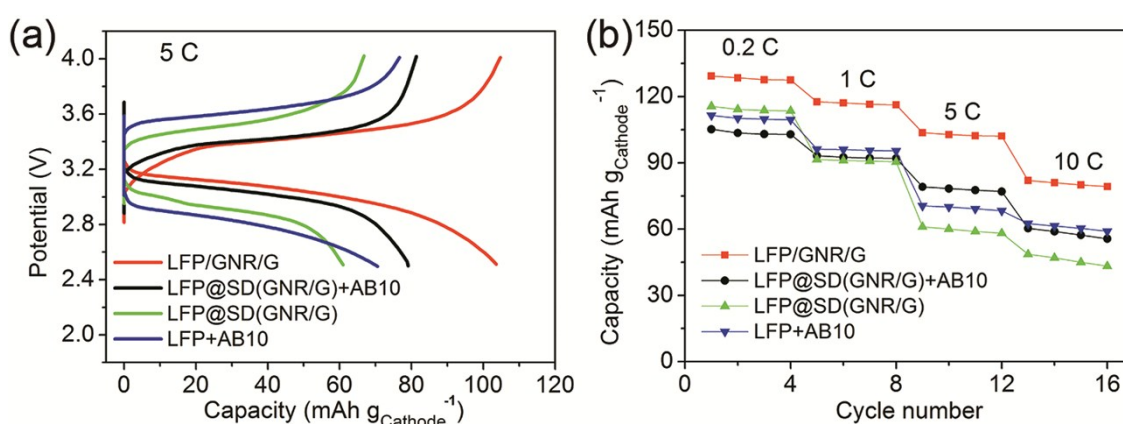


Fig. S9 Comparison of electrodes LFP+AB10, LFP@SD(GNR/G)+AB10, LFP@SD(GNR/G) and LFP/GNR/G based on the cathode weight in full cells with graphite as the anode material. (a) Charge/discharge curves at the current rate of 5 C and (b) rate capabilities.

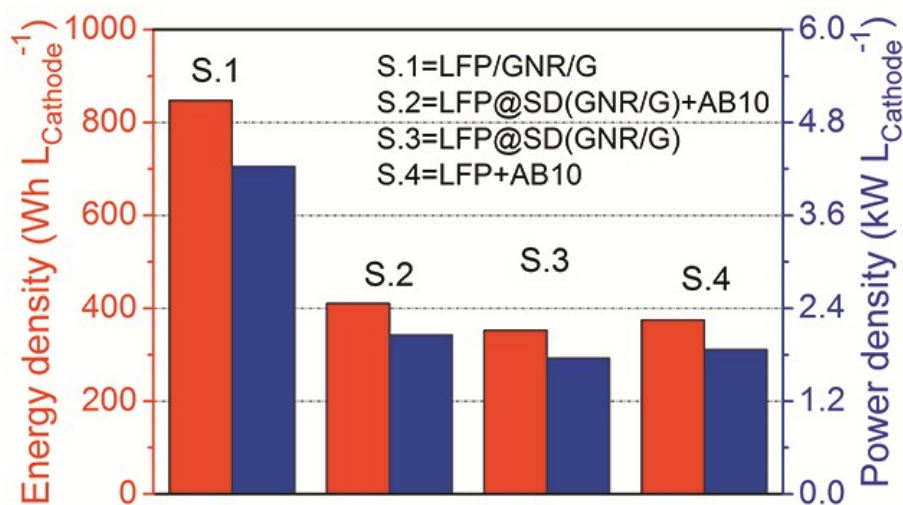


Fig. S10 Energy density and power density comparisons of LFP+AB10, LFP@SD(GNR/G)+AB10, LFP@SD(GNR/G) and LFP/GNR/G samples in full cells at 5 C rate. The discharge median-potentials of 2.81 V, 2.91 V, 3.02 V and 3.03 V were used for LFP+AB10, LFP@SD(GNR/G), LFP@SD(GNR/G)+AB10 and LFP/GNR/G samples, respectively.

Table S1 EDS elemental analysis of commercial LiFePO₄.

Element	Mass%	Atomic%
C _K	8.16	14.54
O _K	46.79	62.57
P _K	18.32	12.65
Fe _K	26.73	10.24
Total amount	100.00	100.00

Table S2 The residual contents of freeze-dried GO and GONR after 600 °C treatment in Ar/H₂ (90/10, V/V) for 5 h.

Samples	Freeze-dried GO sponge	Freeze-dried GONR sponge
Mass before calcinations [mg]	28.27	14.53
Mass after calcinations [mg]	11.35	5.98
Residual content [%]	40.1	41.2

Table S3 Specific capacity comparison of reported LFP electrodes.

Size [nm] / morphology of LFP	Resource of LFP	With Polym -eric binder or not	Conductive additives		Specific properties at 5 C rate (current collector excluded)			Ref.
			Material	Content [wt.%]	Gravimetric capacity [mAh g ⁻¹]	Volumetric capacity [mAh cm ⁻³]	Volumetric energy density [Wh L ⁻¹]	
100 – 2000 (particles)	Commercial	N	G, GNR	1.0, 1.0	118	318	1020	This work
100 – 800 (particles)	Commercial	N	G	7.5	98.5			2
140 (particles)	Commercial	Y	Carbon black	5	~145	~260	~850	3
3000 – 7000 (porous particles)	Hydrothermal	Y	Carbon black	15	~100	~150	~500	4
D=30 – 100 L=80 – 400 (rods)	Hydrothermal	Y	G, Carbon black	<0.7, 8	~105			5
4000– 6000 (porous particles)	Hydrothermal and chemical lithiation	Y	CNT, Carbon black	~1.2, 10	~95	~150	~500	6
~40 (particles)	Hydrothermal	Y	CNT, Carbon black	~2.2, ~12.8	~95			7
D=20 L=100 (rods)	Hydrothermal	Y	N-dopped carbon, G, Carbon black	~1.6, ~2, 10	~126	~180	~600	8
~50 – 200 (particles)	Precipitation and chemical lithiation	Y	N-doped G	~11.4	~125	<200	~650	9
~100 (particles)	Hydrothermal	Y	G, Carbon black	3.6, 10	~84			10

^a)Y: Yes; ^b)N: No; ^c)G: Graphene

References

- 1 C. Wang, Y.S. Li, J. Jiang, W.H. Chiang, *ACS Appl. Mater. Interfaces*, 2015, **7**, 17441-17449.
- 2 Y. Huang, H. Liu, Y.-C. Lu, Y. Hou, Q. Li, *J. Power Sources*, 2015, **284**, 236-244.
- 3 D.P. Singh, F.M. Mulder, A.M. Abdelkader, M. Wagemaker, *Adv. Energy Mater.*, 2013, **3**, 572-578.
- 4 M.-Y. Cho, H. Kim, H. Kim, Y.S. Lim, K.-B. Kim, J.-W. Lee, K. Kang, K.C. Roh, *J. Mater. Chem. A*, 2014, **2**, 5922-5927.
- 5 Y. Long, Y. Shu, X. Ma, M. Ye, *Electrochim. Acta.*, 2014, **117**, 105-112.
- 6 M. Chen, C. Du, B. Song, K. Xiong, G. Yin, P. Zuo, X. Cheng, *J. Power Sources*, 2013, **223**, 100-106.
- 7 B. Wang, T. Liu, A. Liu, G. Liu, L. Wang, T. Gao, D. Wang, X.S. Zhao, *Adv. Energy Mater.*, 2016, **6**, 1600426.
- 8 K. Zhang, J.T. Lee, P. Li, B. Kang, J.H. Kim, G.R. Yi, J.H. Park, *Nano Lett.*, 2015, **15**, 6756-6763.
- 9 J.-P. Jegal, K.-C. Kim, M.S. Kim, K.-B. Kim, *J. Mater. Chem. A*, 2014, **2**, 9594-9599.
- 10 W.-B. Luo, S.-L. Chou, Y.-C. Zhai, H.-K. Liu, *J. Mater. Chem. A*, 2014, **2**, 4927-4931.