Supporting Information

Evaluation of Graphene-Wrapped $LiFePO_4$ as Novel Cathode Materials for Li-Ion Batteries

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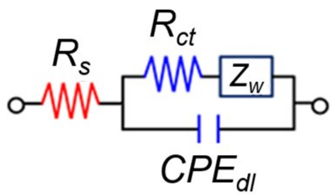
Table S1. Carbon content and grain size of the bare $LiFePO_4$, graphene-wrapped commercial $LiFePO_4$, and graphene-wrapped $LiFePO_4$.

Sample	C [wt. %]	Grain Size [nm]
Bare $LiFePO_4$	-	~400 (from SEM)
***c*-$LiFePO_4$/G**	2.1	~400 (from SEM)
$LiFePO_4$/G	1.7	83 ± 10 (from XRD) 67 ± 25 (from SEM)

Table S2. Impedance parameters extracted using a modified Randles' equivalent circuit for the $LiFePO_4$/G and *c*-$LiFePO_4$/G. In Randles' circuit, R_s (series resistance) represents the high-frequency resistance of the electrolyte. CPE_{dl} (constant phase element) reflects imperfect capacitance of the electrochemical double layer between the electrolyte and electrode. R_{ct} (charge-transfer resistance) is associated with the ion injection and the electron transfer from the electrolyte into the electrode. Z_w is the Warburg impedance.



Parameters	R_{ct} [Ω g]	Warburg factor *σ*	D_{EIS} [× 10^{-12} cm^2 s^{-1}]
$LiFePO_4$/G (C0)	0.0181 ± 0.0001	0.04652 ± 0.00133	143
$LiFePO_4$/G (C1)	0.0164 ± 0.0005	0.00687 ± 0.00014	3.1
$LiFePO_4$/G (C2)	0.0184 ± 0.0003	0.00621 ± 0.00058	2.5
$LiFePO_4$/G (C3)	0.0191 ± 0.0003	0.00625 ± 0.00014	2.6
$LiFePO_4$/G (C4)	0.0195 ± 0.0003	0.00629 ± 0.00035	2.6
$LiFePO_4$/G (C5)	0.0198 ± 0.0003	0.00638 ± 0.00052	2.7
$LiFePO_4$/G (C6)	0.0199 ± 0.0003	0.00665 ± 0.00072	2.9
$LiFePO_4$/G (C7)	0.0201 ± 0.0003	0.00731 ± 0.00079	3.5
$LiFePO_4$/G (C8)	0.0197 ± 0.0003	0.00892 ± 0.00093	5.2
$LiFePO_4$/G (C9)	0.0197 ± 0.0002	0.02432 ± 0.00023	38.5
$LiFePO_4$/G (C10)	0.0222 ± 0.0002	0.01744 ± 0.00141	20.1
$LiFePO_4$/G (D1)	0.0222 ± 0.0002	0.01744 ± 0.00141	20.1
$LiFePO_4$/G (D2)	0.0181 ± 0.0001	0.00348 ± 0.00003	0.8
$LiFePO_4$/G (D3)	0.0164 ± 0.0005	0.00388 ± 0.00004	0.9
$LiFePO_4$/G (D4)	0.0184 ± 0.0003	0.00433 ± 0.00004	1.2
$LiFePO_4$/G (D5)	0.0191 ± 0.0003	0.00487 ± 0.00003	1.6
$LiFePO_4$/G (D6)	0.0195 ± 0.0003	0.00554 ± 0.00002	2.1
$LiFePO_4$/G (D7)	0.0198 ± 0.0003	0.00637 ± 0.00002	2.7
$LiFePO_4$/G (D8)	0.0199 ± 0.0003	0.00753 ± 0.00003	3.7
$LiFePO_4$/G (D9)	0.0201 ± 0.0003	0.00931 ± 0.00006	5.7
$LiFePO_4$/G (D10)	0.0197 ± 0.0003	0.01248 ± 0.00009	10.3
$LiFePO_4$/G (D11)	0.0197 ± 0.0002	0.01965 ± 0.00003	25.4

Parameters	***R_{ct}*** **[Ω g]**	**Warburg factor *σ***	***D_{EIS}*** **[× 10^{-10} cm^2 s^{-1}]**
***c*-$LiFePO_4$/G (C0)**	0.068 ± 0.017	0.289 ± 0.005	55.3
***c*-$LiFePO_4$/G (C1)**	0.069 ± 0.001	0.226 ± 0.003	33.9
***c*-$LiFePO_4$/G (C2)**	0.124 ± 0.045	0.055 ± 0.001	2.0
***c*-$LiFePO_4$/G (C3)**	0.179 ± 0.009	0.056 ± 0.002	2.0
***c*-$LiFePO_4$/G (C4)**	0.199 ± 0.016	0.008 ± 0.001	4.7
***c*-$LiFePO_4$/G (C5)**	0.349 ± 0.026	0.182 ± 0.007	21.8
***c*-$LiFePO_4$/G (C6)**	0.483 ± 0.003	0.211 ± 0.003	29.1
***c*-$LiFePO_4$/G (D1)**	0.483 ± 0.003	0.211 ± 0.003	29.1
***c*-$LiFePO_4$/G (D2)**	0.471 ± 0.016	0.107 ± 0.008	7.6
***c*-$LiFePO_4$/G (D3)**	0.464 ± 0.027	0.116 ± 0.007	8.9
***c*-$LiFePO_4$/G (D4)**	0.453 ± 0.029	0.129 ± 0.005	10.9
***c*-$LiFePO_4$/G (D5)**	0.383 ± 0.012	0.137 ± 0.001	12.3
***c*-$LiFePO_4$/G (D6)**	0.382 ± 0.011	0.136 ± 0.001	12.1
***c*-$LiFePO_4$/G (D7)**	0.334 ± 0.009	0.152 ± 0.005	15.2

The Li-ion diffusion coefficient (D_{EIS}) from *in situ* electrochemical impedance spectra (EIS) is presented as:

$$D_{EIS} = \left(\frac{R^2 T^2}{2 A^2 n^4 F^4 {C_{Li}}^2 \sigma^2} \right), \quad (1)$$

where D_{EIS} is the diffusion coefficient (cm^2 s^{-1}) of Li in the low-frequency (≤ 10 Hz) region, A is the surface area of the electrode (cm^2 g^{-1}), n is the number of electrons, F is the Faraday constant (96485 C mol^{-1}), C_{Li} is the concentration of Li^+ (mol cm^{-3}), and σ is the Warburg factor obtained from the slope of the lines between the real axis Z' and the reciprocal square root of the lower angular frequencies $\omega^{-1/2}$ ($Z' = R_s + R_{ct} + \sigma\, \omega^{-1/2}$).

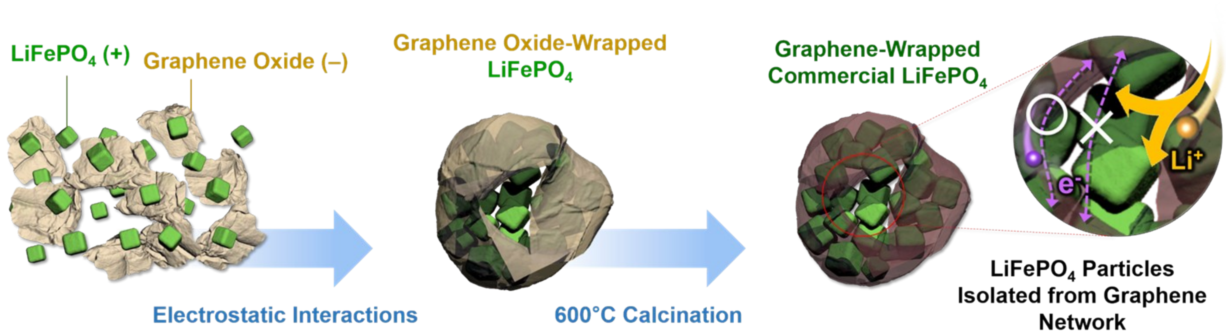


Figure S1. Schematic illustration showing the synthetic process for the graphene-wrapped commercial $LiFePO_4$.

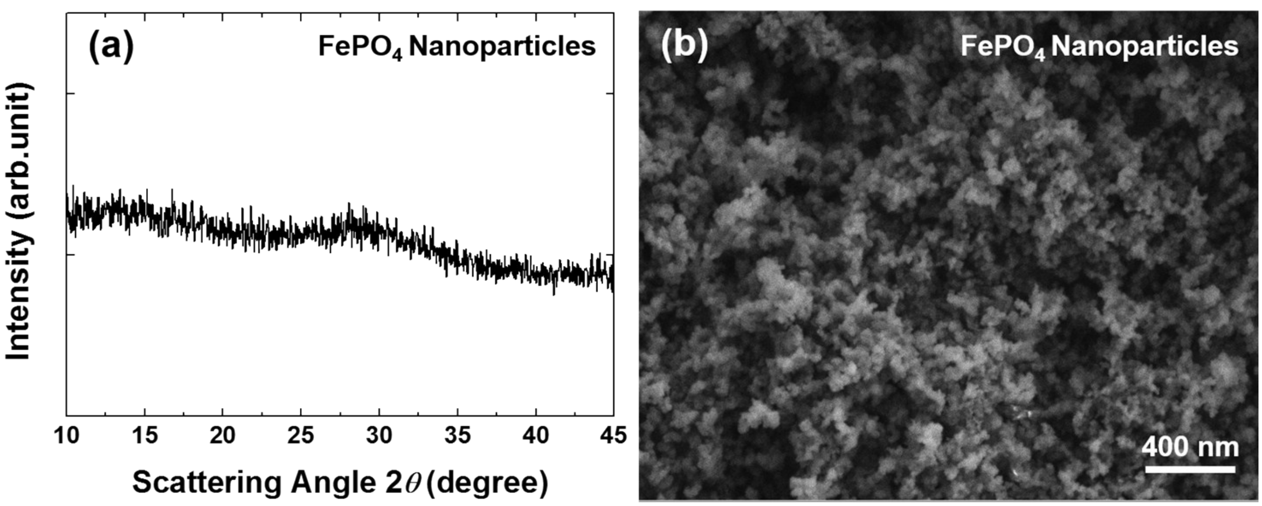


Figure S2. (a) XRD pattern and (b) SEM image of the amorphous $FePO_4$ nanoparticles.

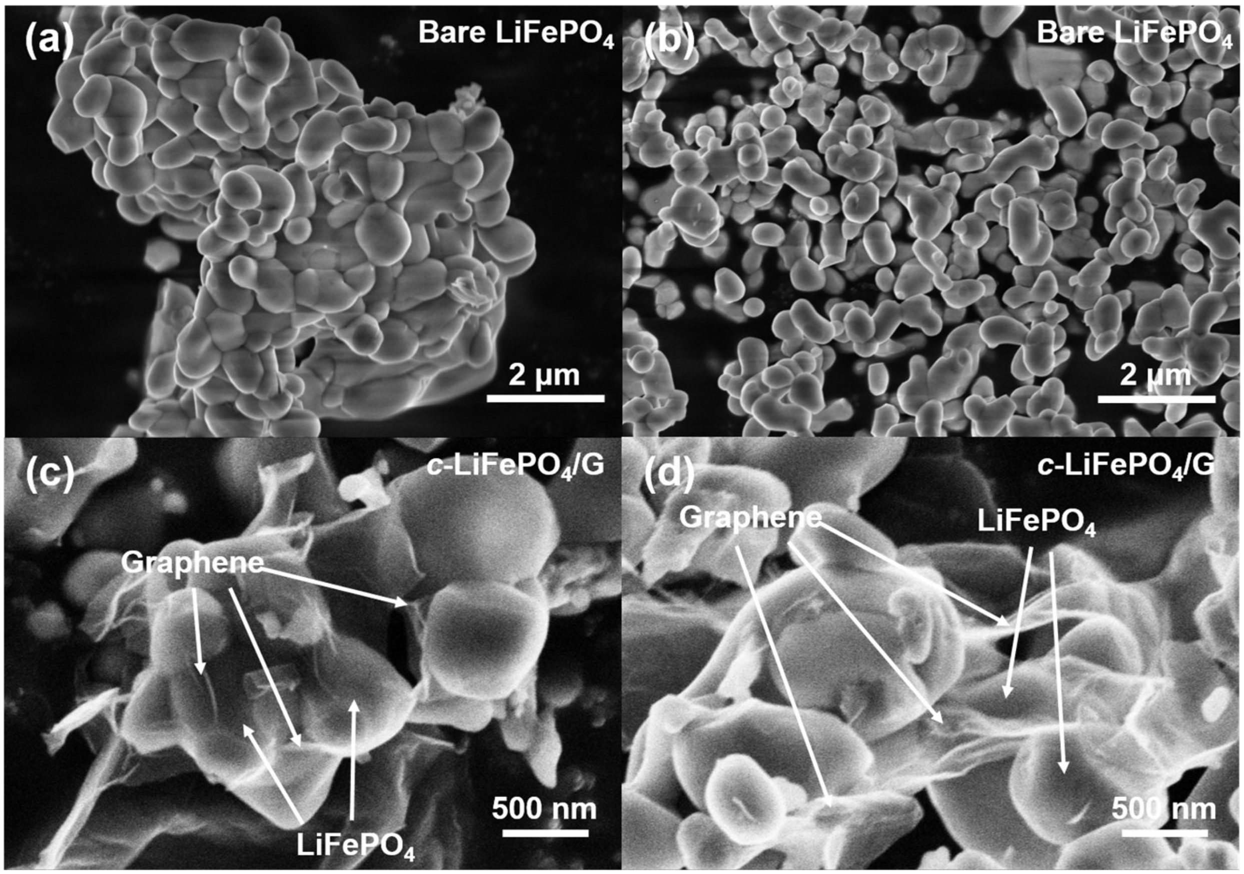


Figure S3. SEM images of the (a-b) bare $LiFePO_4$ and (c-d) graphene-wrapped commercial $LiFePO_4$.

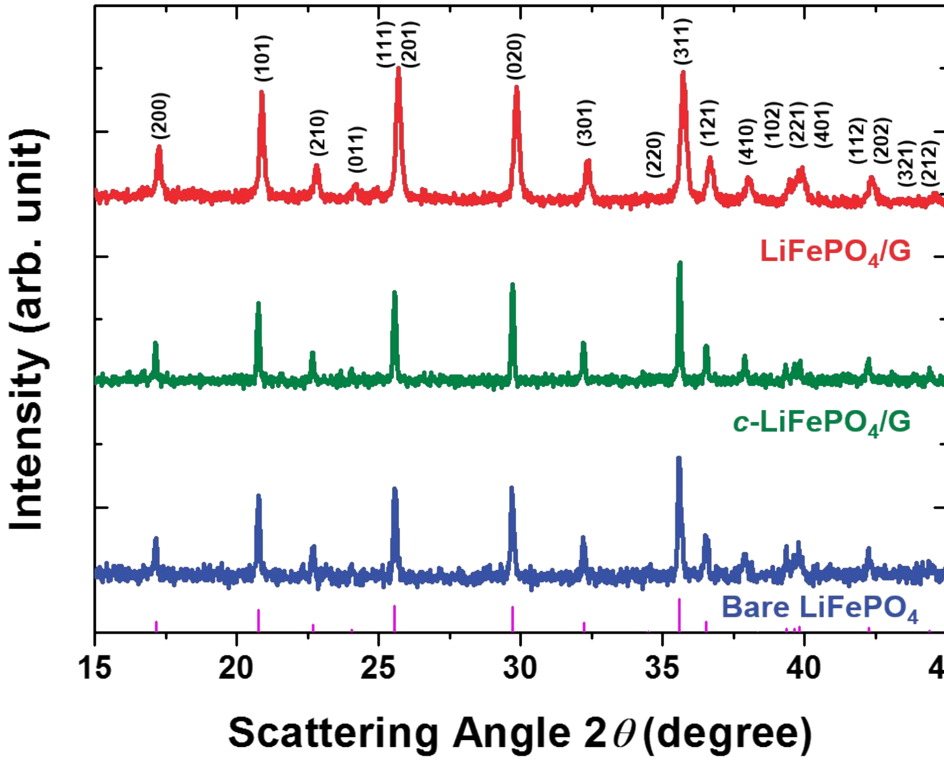


Figure S4. XRD patterns of the bare $LiFePO_4$, graphene-wrapped commercial $LiFePO_4$, and graphene-wrapped $LiFePO_4$. The ideal peak positions and intensities for $LiFePO_4$ (JCPDS #40-1499) are marked at the bottom.

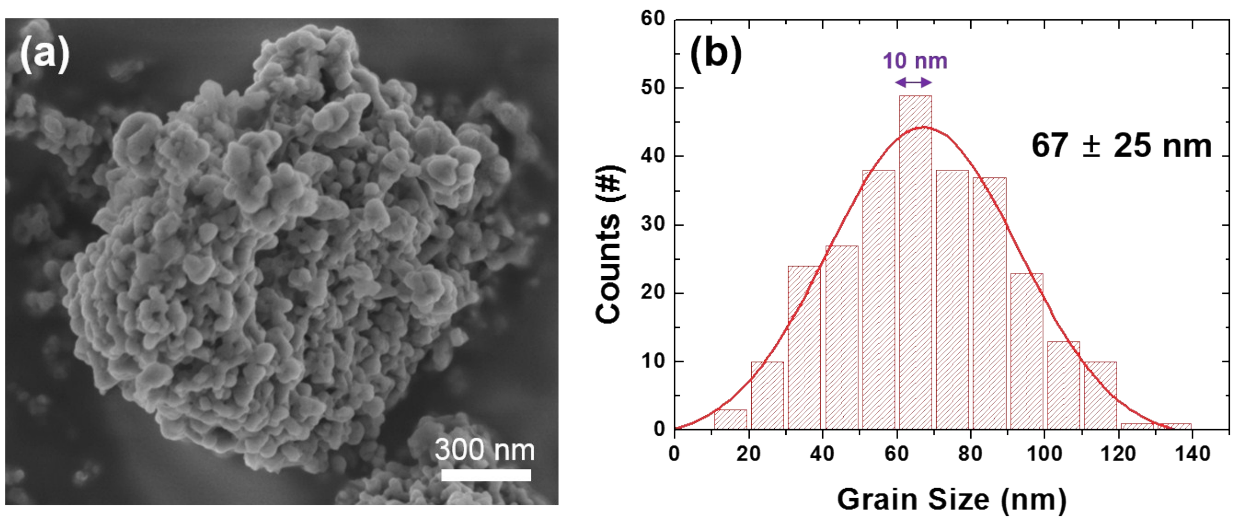


Figure S5. (a) SEM image of the graphene-eliminated $LiFePO_4$, and (b) grain-size distribution of the graphene-wrapped $LiFePO_4$.