Supporting Information

Bottom-up Approach Design, Band Structure and Lithium Storage Properties of Atomically Thin γ-FeOOH Nanosheets

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Table S1. Structure parameters for γ-FeOOH (JCPDS card, No. 44−1415. a = 12.52 Å, b = 3.87 Å, c = 3.07 Å) showing fractional coordinates (x, y, z) and occupancies (g).

Atom	x	y	z	g
Fe	0.678	0.25	0	1
O	0.282	0.25	0	1
OH	0.075	0.25	0	1

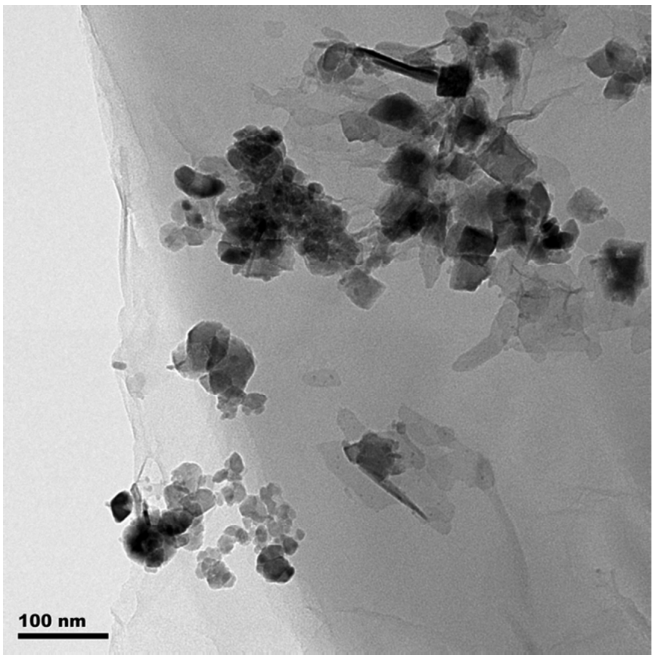

Figure S1. TEM image of the product synthesized at a high $FeCl_2$ concentration of 0.6 g L^{-1}.

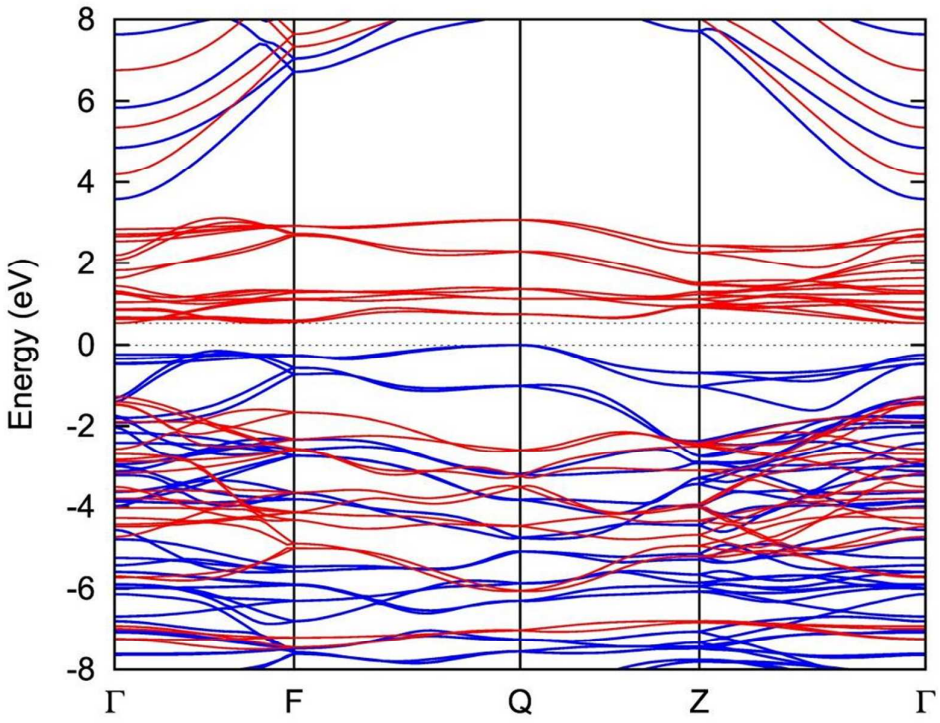

Figure S2. Band structure of γ-FeOOH 3D bulk. The Brillouin paths are chosen the same as that of 2D γ-FeOOH nanosheet for a direct comparison.

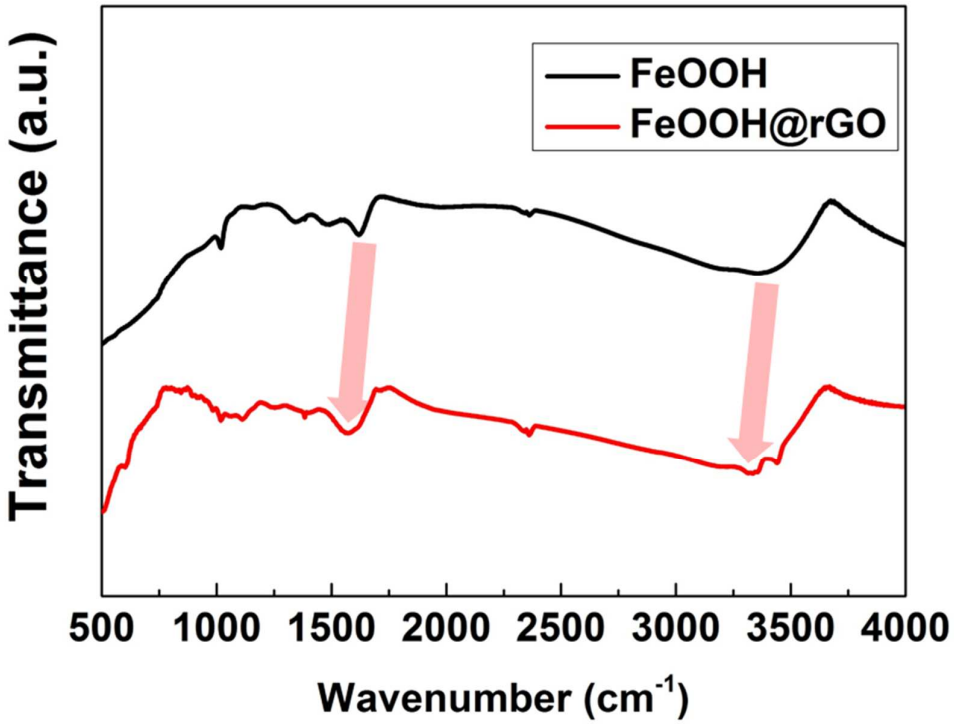

Fig. S3. FTIR spectra of FeOOH and FeOOH@rGO composite

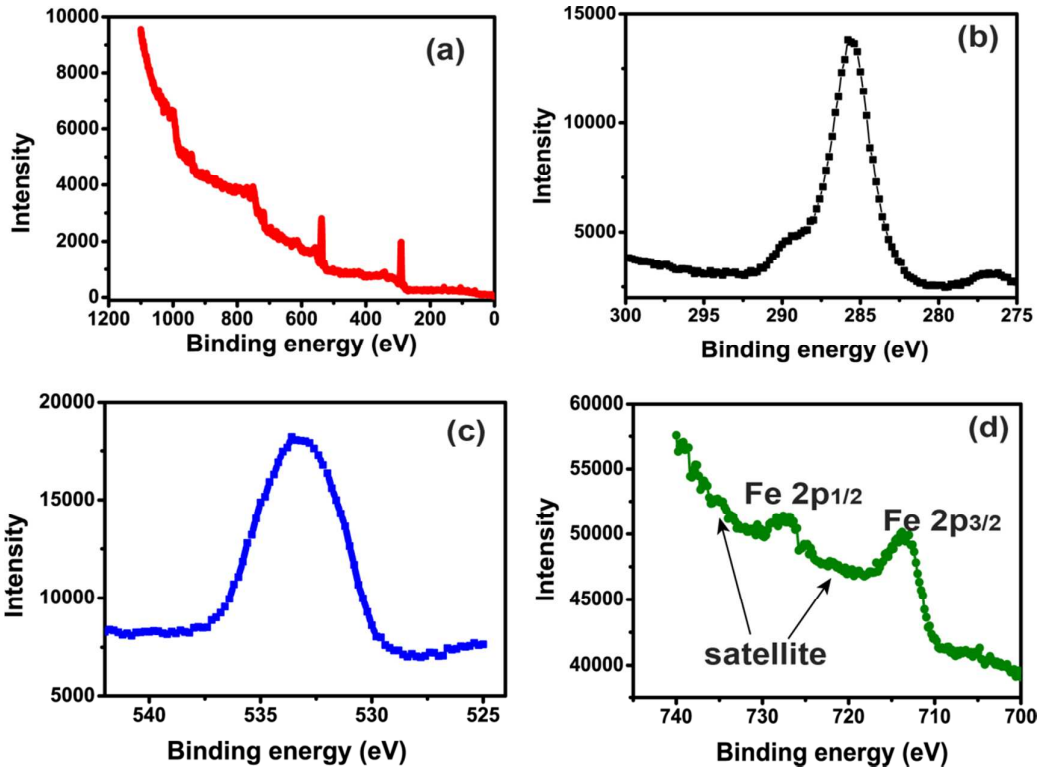

Figure S4. XPS spectra of (a) γ-FeOOH@rGO nanocomposites, (b–d) high resolution XPS spectra for C, O and Fe elements, respectively. Two peaks at 713.8 and 727.6 eV can be assigned to Fe $2p_{3/2}$ and Fe $2p_{1/2}$ main peaks, respectively. The appearance of satellite bands near the Fe 2p main peaks is generally regarded as an indicator of the valence state of Fe^{3+}. [1] In addition, The C1s spectrum of FeOOH@rGO nanocomposites can be resolved into two peaks centered at 284.5 and 287.8 eV, assigning to C-C and C=O bonds, respectively. The intensity of C-C bond is much higher than that of C=O bond, demonstrating the successful reduction of GO into rGO in our process.

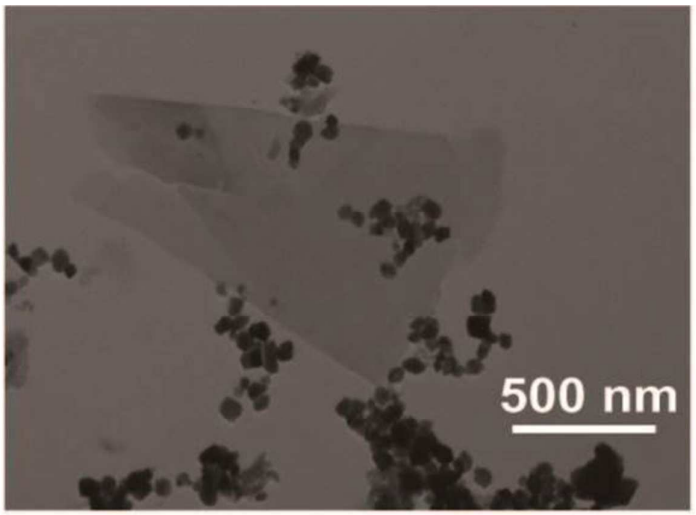

Figure S5. The TEM image of the sample prepared by mixing γ-FeOOH nanosheets and GO at a hydrothermal temperature of 170 ºC. Higher hydrothermal temperature of 170 °C is disadvantageous for the homogenous dispersion of γ-FeOOH on rGO.

Table S2. Comparison of the cycling stability of the as-obtained γ-FeOOH@rGO and previously reported FeOOH-based materials (>50 cycles). This fast activation process of γ-FeOOH@rGO nanocomposites is superior to other reported FeOOH systems, which always suffer from longstanding activation process.

Anode	activation cycle number	valley capacity (mAh g^{-1})	stable capacity (mAh g^{-1})	fluctuation ratio	Ref
Mn-doped α-FeOOH nanorods	~200	~500	883	~43%	2
β-FeOOH nanorods	~300	285	~500	~45%	3
10 wt.% graphite doped β-FeOOH nanorods	~300	~300	~600	~50%	3
amorphous FeOOH particle/rGO	~100	~600	767	~21%	4
ultrathin γ-FeOOH nanosheets@rGO	**~10**	**790**	**860**	**8%**	**This work**

REFERENCES

(1) Ma, R.; Liang, J.; Takada, K.; Sasaki, T. Topochemical Synthesis of Co-Fe Layered Double Hydroxides at Varied Fe/Co Ratios: Unique Intercalation of Triiodide and Its Profound Effect. *J. Am. Chem. Soc.* **2011**, *133*, 613-620.

(2) Zhai, Y. J.; Ma, X. J.; Mao, H. Z.; Shao, W. W.; Xu, L. Q.; He, Y. Y. Mn-Doped Alpha-FeOOH Nanorods and Alpha-Fe_2O_3 Mesoporous Nanorods: Facile Synthesis and Applications as High Performance Anodes for LIBs. *Adv. Electron. Mater*. **2015**, *1*, 1400057.

(3) Yu, L. H.; Xi, S. B., Wei, C.; Zhang, W. Y.; Du, Y. H.; Yan, Q. Y.; Xu, Z. J. Superior Lithium Storage Properties of Beta-FeOOH. *Adv. Energy Mater*. **2015**, *5*, 1401517.

(4) Sun, Y.; Hu, X.; Luo, W.; Xu, H.; Hu, C.; Huang, Y. Synthesis of Amorphous FeOOH/Reduced Graphene Oxide Composite by Infrared Irradiation and Its Superior Lithium Storage Performance. *ACS Appl. Mater. Interfaces* **2013**, *5*, 10145-10150.