Supporting Information:

# Hybridization of graphene nanosheets and carbon-coated hollow $Fe_3O_4$ nanoparticles as a high-performance anode material for lithium-ion batteries

**Yongtao Zuo[a], Gang Wang[a*], Jun Peng[a], Gang Li[a], Yanqing Ma[a,*], Feng Yu[a], Bin Dai[a], Xuhong Guo[a,b], Ching-Ping Wong[c,d]**

[a] *School of Chemistry and Chemical Engineering, Key Laboratory of Materials-Oriented Chemical Engineering of Xinjiang Uygur Autonomous Region, Shihezi University, Shihezi, P.R. China. Email:gwangshzu@163.com, mayanqing@shzu.edu.cn.*
[b] *State Key Laboratory of Chemical Engineering, East China University of Science and Technology, Shanghai 200237, P. R. China*
[c] *Department of Electronic Engineering, The Chinese University of Hong Kong, New Territories, HongKong*
[d] *School of Materials Science and Engineering, Georgia Institute of Technology, Atlanta,GA30332, United States*

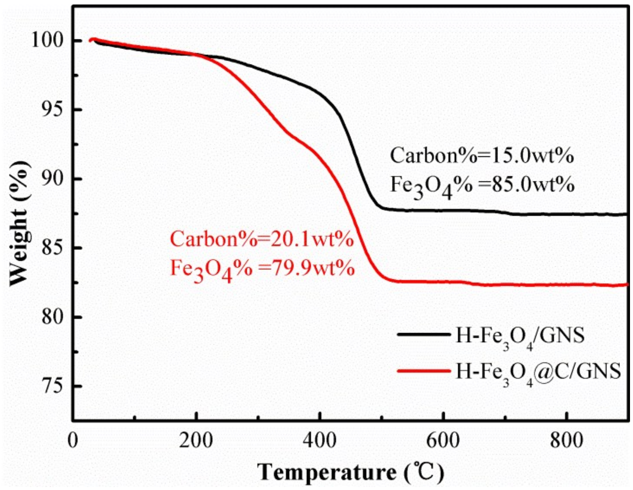


Figure S1. TGA profiles of H-$Fe_3O_4$/GNS nanosheets and H-$Fe_3O_4$@C/GNS nanosheets.

In general, a weight increase should be observed between 150°C and 400°C because of the oxidation of $Fe_3O_4$ to $Fe_2O_3$ as reported in the literatures (Yoon et al, J. Mater. Chem. 2011, 21, 17325-17330; Zhu et al, J. Phys. Chem. C 2011, 115, 9814-9820; Wang et al, RSC Adv. 2014, 4, 322-330; Dong et al, Phys. Chem. Chem. Phys. 2013, 15, 7174-7181). However, there is no sign of oxidation of $Fe_3O_4$ in the TGA profile in Figure S1. It may be attributed to the evaporation of the adsorbed solvents (the boil points of ethylene glycol and diethylene alcohol are 197.3°C and 245°C, respectively). This phenomenon is also observed in many $Fe_3O_4$/graphene composites (Li et al, J. Phys. Chem. C 2011, 115, 21567-21573; Chen et al, Adv. Mater. 2011, 23, 5679-5683; Liu et al, RSC Adv. 2014, 4, 17653-17659; Zhuo et al, J. Mater. Chem. A 2013, 1, 3954-3960; Li et al, Adv. Energy Mater. DOI: 10.1002/aenm.201500171; Fan et al, Chem. Commun. 2015, 51, 1597-1600; Guo et al, J. Mater. Chem. A DOI: 10.1039/C5TA05431A).

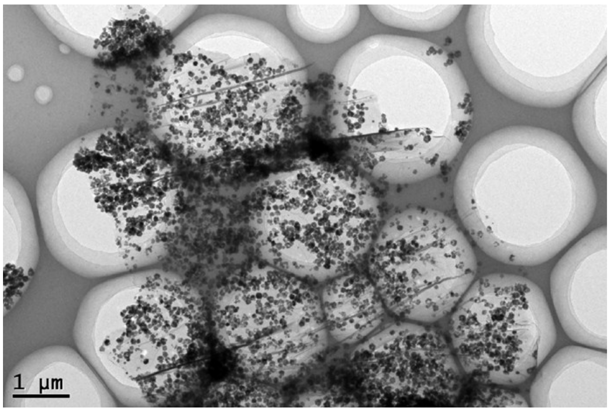


Figure S2. Low magnification TEM images of the as-prepared H-$Fe_3O_4$@C/GNS nanosheets.

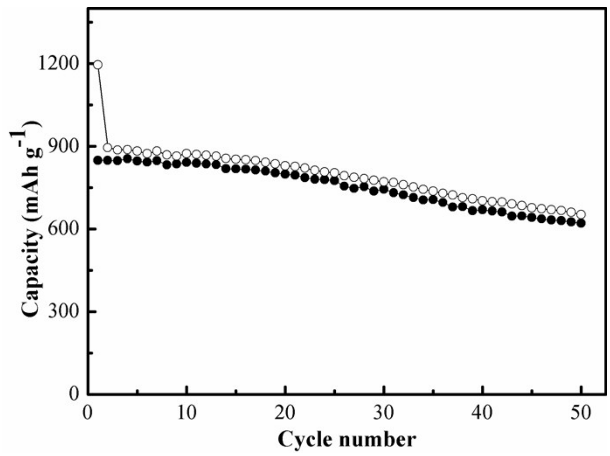


Figure S3  Cycling performance of carbon-encapsulating H-$Fe_3O_4$ (H-$Fe_3O_4$@C).

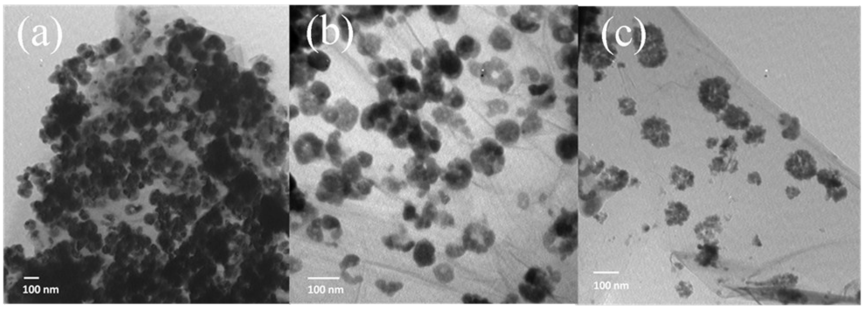


Figure S4 TEM images of $H\text{-}Fe_3O_4@C/GNS$ hyrids with different adding amount of GO during the solvothermal process. (a) 50mg; (b) 100mg; (c) 150mg.

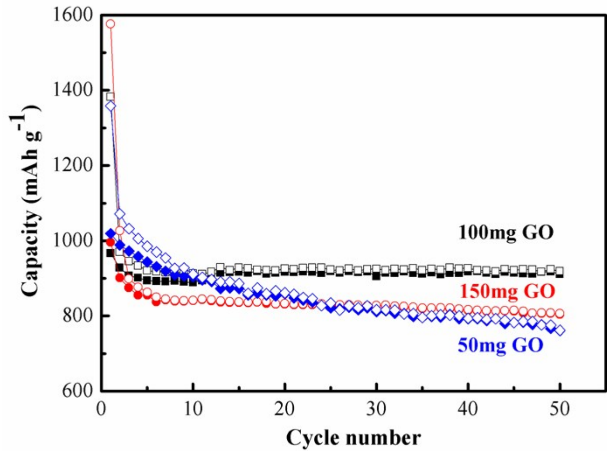


Figure S5 Cycling performance of H-$Fe_3O_4$@C/GNS hyrids with different adding amount of GO during the solvothermal process. (a) 50mg; (b) 100mg; (c) 150mg.

Table S1 Cycling performance of $Fe_3O_4$/carbon hybrids and other Fe-based materials as anodes for LIBs

| **Materials** | **Cycling Performance** | **Rate Performance** | **Ref.** |
|---|---|---|---|
| C-$Fe_3O_4$ nanospheres | 712 mAh $g^{-1}$ at 200 mA $g^{-1}$ (60 cycles) | / | 7 |
| magnetite-C nanocomposite | 776.7 mAh $g^{-1}$ at 100 mA $g^{-1}$ (30 cycles) | / | 15 |
| $Fe_3O_4$/CF composite | 784 mAh $g^{-1}$ at 100 mA $g^{-1}$ (50 cycles) | 570 mA h $g^{-1}$ at 0.5C<br>~140 mA h $g^{-1}$ at 1~2C | 16 |
| $Fe_3O_4$/MSU-F-C nanocomposite | ~1118 mAh $g^{-1}$ at 100 mA $g^{-1}$ (150 cycles) | 750 mA h $g^{-1}$ at 1C<br>~500 mA h $g^{-1}$ at 5C | 17 |
| Carbon-coated $Fe_3O_4$ nanospindles | 530 mAh $g^{-1}$ at 0.5C (80 cycles) | 190 mA h $g^{-1}$ at 5C | 18 |
| carbon–$Fe_3O_4$ nanocomposite | ~1000 mAh $g^{-1}$ at 100 mA $g^{-1}$ (40 cycles) | 418 mA h $g^{-1}$ at 2500 mA $g^{-1}$ | 19 |
| graphene paper@$Fe_3O_4$ nanorod array@graphene | 1211 mAh $g^{-1}$ at 0.2A $g^{-1}$ (350 cycles)<br>759 mAh $g^{-1}$ at 2A $g^{-1}$ (1450 cycles) | 728 mA h $g^{-1}$ at 5 A $g^{-1}$ | 20 |
| $Fe_3O_4$@C | 808.2 mAh $g^{-1}$ at 924 mA $g^{-1}$ (100 cycles) | / | 21 |
| GNS/$Fe_3O_4$ composite | 950 mAh $g^{-1}$ at 35 mA $g^{-1}$ (85 cycles)<br>~600 mAh $g^{-1}$ at 700 mA $g^{-1}$ (100 cycles) | 520 mA h $g^{-1}$ at 1750 mA $g^{-1}$ | 22 |
| RGO/$Fe_xO_y$ nanocomposites | 868.4 mAh $g^{-1}$ at 500 mA $g^{-1}$ (300 cycles) | 585.8 mAh $g^{-1}$ at 2000 mA $g^{-1}$ | 23 |
| $Fe_3O_4$–rGO composite | 1039 mAh $g^{-1}$ at 100 mA $g^{-1}$ (170 cycles) | 396 mA h $g^{-1}$ at 1000 mA $g^{-1}$<br>193 mA h $g^{-1}$ at 2000 mA $g^{-1}$ | 24 |
| hollow $Fe_3O_4$/graphene | 940 mAh $g^{-1}$ at 200 mA $g^{-1}$ (50 cycles)<br>660 mAh $g^{-1}$ at 500 mA $g^{-1}$ (50 cycles) | 550 mA h $g^{-1}$ at 1000 mA $g^{-1}$<br>420 mA h $g^{-1}$ at 2000 mA $g^{-1}$ | 25 |
| $Fe_3O_4$/graphene hybrids | 951 mAh $g^{-1}$ at 50 mA $g^{-1}$ (50 cycles)<br>796 mAh $g^{-1}$ at 500 mA $g^{-1}$ (200 cycles)<br>531 mAh $g^{-1}$ at 1A $g^{-1}$ (300 cycles)<br>335 mAh $g^{-1}$ at 2A $g^{-1}$ (300 cycles)<br>213 mAh $g^{-1}$ at 5A $g^{-1}$ (300 cycles) | / | 26 |
| $Fe_3O_4$ NCs–GAs | 577 mAh $g^{-1}$ at 5.2A $g^{-1}$ (300 cycles) | 392 mA h $g^{-1}$ at 12 C<br>118 mA h $g^{-1}$ at 35C | 27 |
| G-$Fe_3O_4$ -GNRs | 708 mAh $g^{-1}$ at 0.4A $g^{-1}$ (300 cycles) | ~600 mA h $g^{-1}$ at 1 A $g^{-1}$ | 28 |
| $Fe_3O_4$ nanocrystals@graphene composite | 538.7 mAh $g^{-1}$ at 0.2C (50 cycles) | ~200 mA h $g^{-1}$ at 2C | 29 |
| GF@$Fe_3O_4$ | 785 mAh $g^{-1}$ at 1C (500 cycles)<br>~400 mAh $g^{-1}$ at 6C (500 cycles)<br>300 mAh $g^{-1}$ at 10C (500 cycles) | 190 mA h $g^{-1}$ at 60C | 30 |
| $Fe_3O_4$@GS/GF | 1059 mAh $g^{-1}$ at 93 mA $g^{-1}$ (150 cycles) | 363 mAh $g^{-1}$ at 4800 mA $g^{-1}$ | 31 |
| $Fe_3O_4$@GN composite | 826 mAh $g^{-1}$ at 1A $g^{-1}$ (100 cycles) | 460 mAh $g^{-1}$ at 5A $g^{-1}$ | 32 |
| $Fe_3O_4$–Carbon–rGO | 842.7 mAh $g^{-1}$ at 0.2C (100 cycles) | ~600 mAh $g^{-1}$ at 1C<br>~400 mAh $g^{-1}$ at 2C | 34 |
| $Fe_3O_4$@GNSs | 1010 mAh $g^{-1}$ at 100 mA $g^{-1}$ (50 cycles) | 300 mAh $g^{-1}$ at 5C | 35 |
| GN-$Fe_3O_4$ | 1102 mAh $g^{-1}$ at 100 mA $g^{-1}$ (10 cycles) | 474 mAh $g^{-1}$ at 1600 mA $g^{-1}$ | 36 |
| Graphene-Encapsulated Hollow Porous $Fe_3O_4$ | ~900 mAh $g^{-1}$ at 100 mA $g^{-1}$ (50 cycles) | ~580 mAh $g^{-1}$ at 800 mA $g^{-1}$ | 37 |
| G@$Fe_3O_4$@C | 920 mAh $g^{-1}$ at 200 mA $g^{-1}$ (100 cycles) | 550 mAh $g^{-1}$ at 5000 mA $g^{-1}$ | 39 |
| $Fe_3O_4$–graphene nanocomposites | 1200 mAh $g^{-1}$ at 2C (100 cycles)<br>460 mAh $g^{-1}$ at 4C (400 cycles)<br>180 mAh $g^{-1}$ at 10C (800 cycles) | / | 40 |
| N-G–$Fe_3O_4$ composite | 800 mAh $g^{-1}$ at 100 mA $g^{-1}$ (100 cycles) | 763 mAh $g^{-1}$ at 800 mA $g^{-1}$<br>733 mAh $g^{-1}$ at 1600 mA $g^{-1}$ | 41 |
| H-$Fe_3O_4$-MS/GNS composite | 1171.6 mAh $g^{-1}$ at 200 mA $g^{-1}$ (70 cycles) | 935 mAh $g^{-1}$ at 1000 mA $g^{-1}$ | 42 |

| | | | |
|---|---|---|---|
| | 940.4 mAh $g^{-1}$ at 500 mA $g^{-1}$ (70 cycles) | 745 mAh $g^{-1}$ at 3000 mA $g^{-1}$ | |
| graphene/$Fe_3O_4$ aerogel | ~990 mAh $g^{-1}$ at 200 mA $g^{-1}$ (30 cycles) | 730 mAh $g^{-1}$ at 1600 mA $g^{-1}$ | 43 |
| $Fe_3O_4$/GNSs | ~605 mAh $g^{-1}$ at 92.5 mA $g^{-1}$ (50 cycles) | / | 44 |
| nitrogen-doped graphene/$Fe_3O_4$ | 1130 mAh $g^{-1}$ at 100 mA $g^{-1}$ (200 cycles) | 648 mAh $g^{-1}$ at 1600 mA $g^{-1}$ | 45 |
| $Fe_3O_4$–graphene composite | 868 mAh $g^{-1}$ at 200 mA $g^{-1}$ (100 cycles)<br>539 mAh $g^{-1}$ at 1000 mA $g^{-1}$ (200 cycles) | 282 mAh $g^{-1}$ at 2000 mA $g^{-1}$ | 46 |
| $Fe_3O_4$@C@PGC nanosheets | 998 mAh $g^{-1}$ at 1A $g^{-1}$ (100 cycles)<br>792 mAh $g^{-1}$ at 5C (350 cycles)<br>556 mAh $g^{-1}$ at 10C (350 cycles) | 364 mAh $g^{-1}$ at 15C<br>311 mAh $g^{-1}$ at 20C | 47 |
| Mesoporous $ZnFe_2O_4$ Microrods | ~434 mAh $g^{-1}$ at 1000 mA $g^{-1}$ (51 cycles)<br>542 mAh $g^{-1}$ at 1000 mA $g^{-1}$ (488 cycles) | ~326 mAh $g^{-1}$ at 1500 mA $g^{-1}$ | 54 |
| ZnO/$ZnFe_2O_4$@C mesoporous nanospheres | ~893 mAh $g^{-1}$ at 500 mA $g^{-1}$ (60 cycles)<br>~718 mAh $g^{-1}$ at 1000 mA $g^{-1}$ (500 cycles) | ~770 mAh $g^{-1}$ at 1500 mA $g^{-1}$ | 55 |
| ZnO/$ZnFe_2O_4$ submicrocubes | 837 mAh $g^{-1}$ at 1000 mA $g^{-1}$ (200 cycles) | ~667 mAh $g^{-1}$ at 2000 mA $g^{-1}$ | 56 |
| H-$Fe_3O_4$@C/GNS nanosheets | 887.8 mAh $g^{-1}$ at 0.1C(100 cycles)<br>744.9 mAh $g^{-1}$ at 1C (200 cycles)<br>284.9 mAh $g^{-1}$ at 10C (200 cycles) | ~560 mAh $g^{-1}$ at 3C<br>~448 mAh $g^{-1}$ at 5C<br>~300 mAh $g^{-1}$ at 10C | This work |

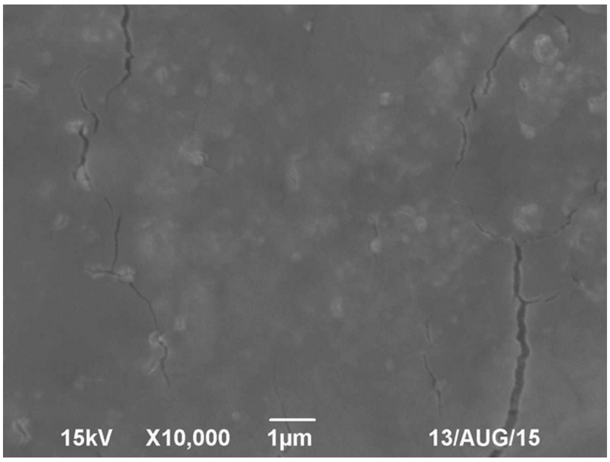


Figure S6 SEM image of the H-$Fe_3O_4$@C/GNS nanosheets electrodes after 200 discharge/charge cycles at a current rate of 1 C.

Table S2 Kinetic Parameters of the H-$Fe_3O_4$ NPs, H-$Fe_3O_4$/GNS nanosheets and H-$Fe_3O_4$@C/GNS nanosheets.

| Materials | SEI film resistance ($R_f$) | charge-transfer resistance ($R_{ct}$) |
|---|---|---|
| H-$Fe_3O_4$ NPs | 105Ω | 97.3Ω |
| H-$Fe_3O_4$/GNS | 73Ω | 28.6Ω |
| H-$Fe_3O_4$@C/GNS | 28Ω | 9.8Ω |