

Supplementary Materials: Facile Preparation of Fe₃O₄ Nanoparticles/Reduced Graphene Oxide Composite as an Efficient Anode Material for Lithium-Ion Batteries

Muhammad Usman Hameed ^{1,*}, Muhammad Yasir Akram ^{2,*}, Ghulam Ali ³, Muhammad Hafeez ⁴, Faizah Altaf ^{1,*}, Ashfaq Ahmed ¹, Shabnam Shahida ⁵ and Patrizia Bocchetta ⁶

- ¹ Department of Chemistry, Women University of Azad Jammu & Kashmir Bagh, Azad Jammu and Kashmir 12500, Pakistan; ashfaqahmad1546@gmail.com
 - ² Department of Chemistry and Humanities, Khawaja Fareed University of Engineering and Information Technology, Rahim Yar Khan 64200, Pakistan
 - ³ USPCAS-E, National University of Sciences and Technology (NUST), Islamabad 44000, Pakistan; ghulamali143@gmail.com
 - ⁴ Department of Chemistry, University of Azad Jammu and Kashmir, Muzaffarabad code, Pakistan; smhafeezkhan@yahoo.com
 - ⁵ Department of Chemistry, University of Poonch, Rawalakot Azad Kashmir code, Pakistan; shabnamshahida01@gmail.com
 - ⁶ Department of Innovative Engineering, University of Salento, Edificio La Stecca, via per Monteroni, 73100 Lecce, Italy; patrizia.bocchetta@unisalento.it
- * Correspondence: usmanhamid506@gmail.com (M.U.H); yakram44@gmail.com (M.Y.A.); faizhaltaf@gmail.com (F.A.)

Citation: Hameed, M.U.; Akram, M.Y.; Ali, G.; Hafeez, M.; Altaf, F.; Ahmed, A.; Shahida, S.; Bocchetta, P. Facile Preparation of Fe₃O₄ Nanoparticles/Reduced Graphene Oxide Composite as an Efficient Anode Material for Lithium-Ion Batteries. *Coatings* **2021**, *11*, 836. <https://doi.org/10.3390/coatings11070836>

Academic Editor: Danut-Ionel Vaureanu

Received: 8 June 2021

Accepted: 8 July 2021

Published: 11 July 2021

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institu-



Copyright: © 2021 by the authors. Submitted for possible open access publication under the terms and conditions of the Creative Commons Attribution (CC BY) license (<http://creativecommons.org/licenses/by/4.0/>).

Table S1. Capacity of carbon based Fe₃O₄ anode materials for LIBS.

| Samples | Capacity (mAh/g) | References |
|---|--|------------|
| Fe ₃ O ₄ /RGO | 701.8 (200 mA/g, 50 cycles) | This work |
| Fe ₃ O ₄ /graphene | 605.0 (92.5 mA/g, 50 cycles) | [1] |
| Fe ₃ O ₄ @graphene | 538.7 (200 mA/g, 50 cycles) | [2] |
| Fe ₃ O ₄ /C microbelts | 710 (100 mA/g, 50 cycles) | [3] |
| Graphene-Fe ₃ O ₄ @Carbon | 710 (100 mA/g, 50 cycles) | [4] |
| Nanocomposites | | |
| Porous carbon-encapsulated Fe ₃ O ₄ | 450 (200 mA/g, 100 cycles) | [5] |
| Fe ₃ O ₄ @C Microcapsules | 600 (92.8 mA/g, 50 cycles) | [6] |
| Fe ₃ O ₄ /Fe/carbon | 600 (50 mA/g, 40 cycles) | [7] |
| Fe ₃ O ₄ -graphene | 410 (75 cycles) at 1 A·g ⁻¹ | [8] |
| Fe ₃ O ₄ NPs-layered graphene | 700 (200 mA/g, 20 cycles) | [9] |
| CNT-Fe ₃ O ₄ @graphene | 408 (100 mA/g, 35 cycles) | [10] |
| Graphene nanosheets- | | |
| Fe ₃ O ₄ NPs | 857 (100 mA/g, 10 cycles) | [11] |
| Fe ₂ O ₃ /rGO | 478 (100 mA/g, 50 cycles) | [12] |
| 78.8 wt.% Fe ₃ O ₄ /rGO | 568 (0.05 A/g, 100 cycles) | [13] |
| Fe ₃ O ₄ /rGO | 446 (5, 50 Cycles) | [14] |
| Fe ₃ O ₄ /rGO | 300 (1, 100 Cycles) | [15] |

It can be seen from Table S1 that the current work showed excellent electrochemical performance, which was primarily related to the uniform dispersion of as-prepared nano-Fe₃O₄ on the surface of graphene. The ultra-high conductivity of graphene can provide an ultra-fast electron transmission network for electrons, and graphene folds can also provide more lithium storage sites [16,17]. Compared with the specific capacities reported in the Table S1, the results of this study are higher than other Fe₃O₄/graphene composite.

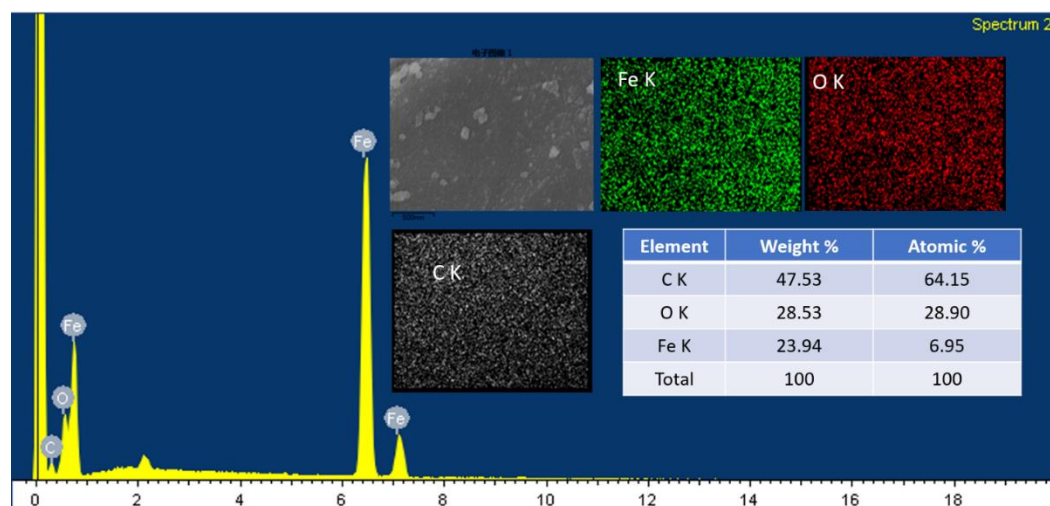


Figure S1. EDX and corresponding elemental mapping of C, O, and Fe.

References

- Li, X.Y.; Huang, X.L.; Liu, D.P.; Wang, X.; Song, S.Y.; Zhou, L.; Zhang, H.J. Synthesis of 3D Hierarchical Fe₃O₄/graphene composites with high lithium storage capacity and for controlled drug delivery. *J. Phys. Chem.* **2011**, *115*, 21567–21573.
- Li, B.J.; Cao, H.Q.; Shao, J.; Qu, M.Z.; Warner, J.H. Superparamagnetic Fe₃O₄ nanocrystals@graphene composites for energy storage devices. *J. Mater. Chem.* **2011**, *21*, 5069–5075.
- Lang, L.M.; Xu, Z. In situ synthesis of porous Fe₃O₄/C microbelts and their enhanced electrochemical performance for lithium-ion batteries. *ACS Appl. Mater. Interfaces* **2013**, *5*, 1698–1703.
- Zhao, L.; Gao, M.M.; Yue, W.B.; Jiang, Y.; Wang, Y.; Ren, Y.; Hu, F.Q. Sandwich-structured graphene-Fe₃O₄@carbon nanocomposites for high-performance lithium-ion batteries. *ACS Appl. Mater. Interfaces* **2015**, *7*, 9709.
- Zhou, Y.P.; Sun, W.P.; Rui, X.H.; Zhou, Y.; Ng, W.J.; Yan, Q.Y.; Fong, E. In situ synthesis of porous Fe₃O₄/C microbelts and their enhanced electrochemical performance for lithium-ion batteries. *Nano Energy* **2016**, *21*, 71.
- Yuan, S.M.; Li, J.X.; Yang, L.T.; Su, L.W.; Liu, L.; Zhou, Z. Preparation and lithium storage performances of mesoporous Fe₃O₄@C microcapsules. *ACS Appl. Mater. Interfaces* **2011**, *3*, 705.
- Zhao, X.Y.; Xia, D.G.; Zheng, K. Fe₃O₄/Fe/carbon composite and its application as anode material for lithium-ion batteries. *ACS Appl. Mater. Interfaces* **2012**, *4*, 1350.
- Lian, P.C.; Zhu, X.F.; Xiang, H.F.; Li, Z.; Yang, W.S.; Wang, H.H. Enhanced cycling performance of Fe₃O₄-graphene nanocomposite as an anode material for lithium-ion batteries. *Electrochim. Acta* **2010**, *56*, 834.
- Liu, Y.; Zhan, Y.; Ying, Y.; Peng, X. Fe₃O₄ nanoparticle anchored layered graphene films for high performance lithium storage. *New J. Chem.* **2016**, *40*, 2649–2654.
- Zhu, W.; Kierzek, K.; Wang, S.; Li, S.; Holze, R.; Chen, X. Improved performance in lithium ion battery of CNT-Fe₃O₄@graphene induced by three-dimensional structured construction. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* **2021**, *612*, 126014.
- Gu, S.; Zhu, A.; Graphene nanosheets loaded Fe₃O₄ nanoparticles as a promising anode material for lithium ion batteries. *J. of Alloys and Compounds* **2020**, *813*, 152160.
- Liu, Z.; Fu, H.; Gao, B.; Wang, Y.; Li, K.; Sun, Y.; Yin, J.; Kan, J. In-situ synthesis of Fe₃O₄/rGO using different hydrothermal methods as anode materials for lithium-ion batteries. *Rev. Adv. Mater. Sci.* **2020**, *59*, 477–86.
- Liang, Y.; Lu, W. Gamma-irradiation synthesis of Fe₃O₄/rGO nanocomposites as lithium-ion battery anodes. *J. Mater. Sci. Mater. Electron* **2020**, *31*, 17075–17083.
- Bhuvaneswari, S.; Pratheeksha, P.M.; Anandan, S.; Rangappa, D.; Gopalan, R.; Rao, T.N. Efficient reduced graphene oxide grafted porous Fe₃O₄ composite as a high-performance anode material for Li-ion batteries. *Phys. Chem. Chem. Phys.* **2014**, *16*, 5284–5294.
- Liang, C.-L.; Liu, Y.; Bao, R.-Y.; Luo, Y.; Yang, W.; Xie, B.; Yang, M.-B. Effects of Fe₃O₄ loading on the cycling performance of Fe₃O₄/rGO composite anode material for lithium ion batteries. *J. Alloys Compd.* **2016**, *678*, 80–86.
- Wang, B.; Xu, B.; Liu, T.; Liu, P.; Guo, C.; Wang, S.; Wang, Q.; Xiong, Z.; Wang, D.; Zhao, X. Mesoporous carbon-coated LiFePO₄ nanocrystals co-modified with graphene and Mg²⁺ doping as superior cathode materials for lithium ion batteries. *Nanoscale* **2014**, *6*, 986–995.
- Wang, B.; Wang, D.; Wang, Q.; Liu, T.; Guo, C.; Zhao, X. Improvement of the electrochemical performance of carbon-coated LiFePO₄ modified with reduced graphene oxide. *J. of Materials Chemistry. A, Materials for Energy and Sustainability* **2013**, *1*, 135–144.