High-Surface-Area Nitrogen-doped Reduced Graphene Oxide for Electric Double Layer Capacitors

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Abstract

Graphene has garnered tremendous research attention in the fields of sensors, energy storage, and energy conversion devices owing to the unique physical and chemical properties associated with its single-atomic-layered sp2 carbon network.[1] In particular, considerable research in the field of energy storage devices has focused on graphene as an electrode material for EDLCs because of its beneficial characteristics: superior high surface area and electrical conductivity.[2, 3] However, most of graphene-based electrode materials reported in the literature showed relatively low specific capacitance compared to the theoretical value (ca. 550 F/g) of a single layer graphene supported by an intrinsic EDL capacitance of 21 μF/cm and a specific surface area of 2650 m^2/g.[2, 4] As an electrode material for EDLCs, reduced graphene oxide (denoted as RGO) prepared by exfoliation and reduction of graphene oxide is extensively investigated due to its advantages of bulk-scale productivity and versatility in chemical functionalization.[5] However, oxidation treatment of graphite involved in the preparation of graphite oxide (denoted as GO) induces a variety of defects and oxygen functional groups, which break up the π-conjugated electronic structure of graphene and thereby degrade the electrical conductivity. The thermal/chemical reduction processes employed to reduce GO to RGO cannot completely restore this π-conjugated structure. Furthermore, the RGO sheet tends to agglomerate due to strong π-π interaction between them during the reduction process, and thereby loses the specific surface area. Therefore, key issues in preparing RGO as an electrode material for EDLCs are an efficient exfoliation for high specific surface area and an extended recovery of the π-conjugated structure of RGO for high electrical conductivity.

The heteroatom doping is another consideration for improving the electrochemical properties by manipulating local electronic structure of the RGO and hence increase in the EDL capacitance and electronic conductivity.[6] Since the quantum capacitance of RGO is thought to be in series with its EDL capacitance, the specific capacitance of RGO, which is the equivalent capacitance for the two capacitances in series, is expected to increase with an increase of the quantum capacitance. A nanoscale EDL is between a carbonaceous active material and an electrolytic solution and contribute to the specific capacitance of RGO, therefore, care should be taken not to decrease the specific surface area of RGO during heteroatom doping.[7]

In this study, a nitrogen-doped RGO (denoted as N-RGO) with high specific surface area, electrical conductivity and low oxygen contents was synthesized using a time-efficient and scalable process composed of microwave irradiation and heat-treatment under NH_3 gas. The near-edge X-ray absorption fine structure (NEXAFS) spectroscopy was employed to investigate the sequential recovery of the π-conjugated structure with removal of oxygen functional groups as well as the chemical bonding environments of incorporated nitrogen atoms in N-RGO. More detailed on the synthetic procedure, morphology, electrochemical and structural properties of encapsulated selenium in graphene micro-ball hybrid materials will be discussed at the meeting.

References

[1] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, A. A. Firsov. Science, **306** (2004) 666
[2] M. D. Stoller, S. Park, Y. Zhu, J. An, R. S. Ruoff. Nano Letters., **8** (2008) 3498
[3] X. Huang, Z. Zeng, Z. Fan, J. Liu, H. Zhang. Advanced Materials, **24** (2012) 5979
[4] J. Xia, F. Chen, J. Li, N. Tao. Nature Nanotechnology, **4** (2009) 505
[5] M. Choucair, P. Thordarson, J. A. Stride. Nature Nanotechnology **4** (2009) 30
[6] L. L. Zhang, X. Zhao, H. Ji, M. D. Stoller, L. Lai, S. Murali, S. McDonnell, B. Cleveger, R. M. Wallace, R. S. Ruoff. Energy & Environmental Science, **5** (2012) 9618.
[7] Y. Qiu, X. Zhang, S. Yang. Physical Chemistry Chemical Physics, **13** (2011) 12554