Room temperature Plasma-Assisted Simultaneous Reduction and Nitrogen Doping of Graphene Oxide Nanosheets for Energy Applications

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Abstract

Recent activity in the research community has seen much interest in graphene and related materials. This material, with its unique physical, chemical, and electronic properties has seen many studies conducted with potential applications in sensing, electronics and energy storage and conversion. Of significant importance to the realisation of these applications and devices is the cheap production of large scale quantities of graphene material. The chemical doping of graphene with heteroatoms has been shown to enhance the performance of graphene in many of these applications. In particular, nitrogen-doping of graphene has been widely demonstrated to improve its performance in a range of applications such as energy conversion and storage and sensing.

This study presents an environmentally benign and scalable route for the production of gram scale quantities of nitrogen-doped graphene using a downstream microwave plasma source. Simultaneous reduction and doping of graphene oxide is achieved and the process negates the need for high temperatures and toxic solvents associated with existing methods. Material characterisation, including XPS, FTIR and Raman spectroscopy, demonstrates significant reduction in oxygen content of the parent material with a nitrogen doping level up to 5.8 at. %. This gas-phase, room temperature process is completely dry and, thus, minimizes re-aggregation of graphene flakes which is typically associated with liquid phase reduction methods. Nitrogen-doped graphene has been proposed as an alternative to expensive platinum/carbon materials used as electrode materials for oxygen reduction in hydrogen fuel cells[1]. Here, preliminary results are presented which demonstrate the potential of our nitrogen-doped graphene in this application. Results indicate that the oxygen reduction mechanism proceeds via a four electron pathway; the most energetically favourable route towards oxygen reduction.

References

[1] L. Qu, Y. Liu, J.-B. Baek, and L. Dai, *ACS Nano*, **4** (2010), 1321–1326
[2] N. McEvoy, H. Nolan, A. K. Nanjundan, T. Hallam, and G. S. Duesberg, *Carbon,*(2012), *In press,* DOI: 10.1016/j.carbon.2012.11.040

Figures

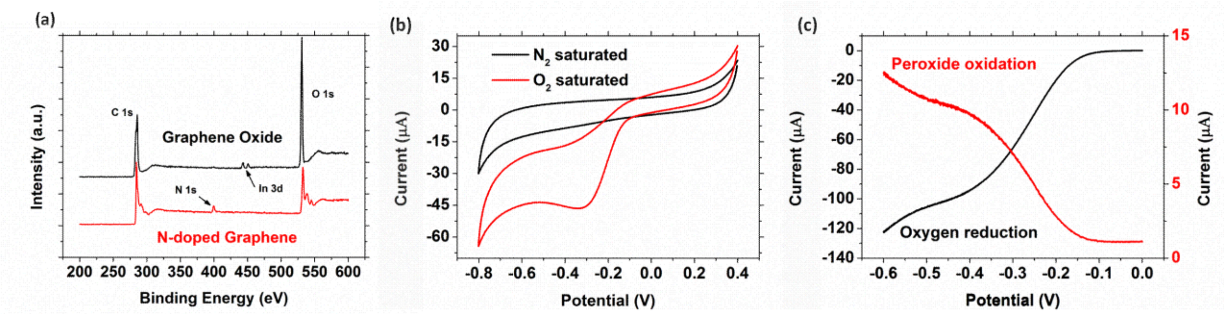

Figure 1: (a) XPS data showing the significant reduction in oxygen content after exposing graphene oxide to the plasma treatment; as evidenced by the smaller oxygen 1s peak for N-doped graphene. Also of note is the introduction of the characteristic nitrogen 1s peak in the doped material. The indium peaks are due to sample mounting procedures and may be ignored. (b) Cyclic voltammograms for N-doped graphene employed as the working electrode in a three electrode set-up in both N_2- saturated and O_2-saturated 1M NaOH. (c) Linear sweep voltammograms of the same material measured using a rotating ring disc electrode system. Currents associated with the reduction of oxygen (black) and oxidation of hydrogen (red) are shown; indicating a four electron oxygen reduction mechanism.