Improving the Electrochemical Performance of Graphene Nanosheets as Anode in Half and Full Lithium-Ion Cells

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Abstract

Graphene based materials have raised increasing interest for their outstanding properties and numerous applications [1]. Research in Li-ion batteries is not an exception, in particular regarding to anode materials with higher performances in replacement of the commercial graphite. Graphene nanosheets (GNS) as electrode for lithium batteries are produced by several methods, being the reduction of graphitic oxide with hydrazine one of the most studied [2]. Graphene nanosheets (GNS) were prepared from graphitic oxide in aqueous solution with N₂H₄ (1 M) as reducing agent, at 100 °C during 6 h reflux. Figure 1 shows X-ray and transmission microscopy analysis, these reveal an amorphous carbon structure with characteristic signatures at 2θ values of about 25° and 42°, and the typical, thin graphene layers with micrometric growth in two dimensions.

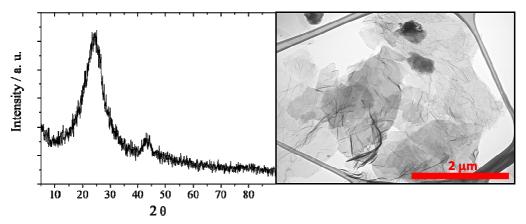


Figure 1. X Ray diffractogram and Transmission Electron Microscopy image of GNS

Figure 2 shows the electrochemical properties of GNS in half cells (vs. Li metal electrode) and full cells (vs. LiFePO₄ as cathode). In half cells the material (black line) can deliver a reversible capacity of ca. 600 mAh g⁻¹ at the end of 30th cycle. However, drawbacks such as the high irreversible capacity during the first cycles (marked with a black ellipse) are still limiting the applicability of the graphene-based materials in full lithium-ion cells. There are some strategies to overcome these drawbacks. [3, 4, 5] The strategy used here consists of a pre-lithiation of the anode through a contact treatment; the anode was placed in contact under pressure with Li foil soaked in LiPF₆ based electrolyte for 5 min. As a result of this treatment the electrolyte is partially decomposed over the surface of graphene, forming the so called *Solid Electrolyte Interface* (SEI); such SEI formation has consumed enough Li⁺ as to reduce the

irreversible capacity in the first cycles, as it is evident from the pre-lithiated curve (red line) in the half cells of Fig. 2.

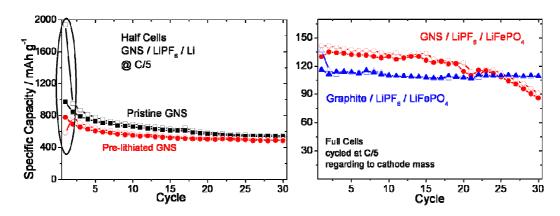


Figure 2. Half cells with and without treatment. Full cells, comparison between Graphite and GNS

Finally, with the pre-lithiated anode a full Li-ion cell has been assembled with LiFePO₄ as cathode. The so formed cell exhibits an average capacity of ca. 120 mAh g⁻¹ over 30 cycles (red line in the full cells of Fig. 2), a higher capacity than that of a similar full cell made with commercial graphite as anode. In spite of the excellent initial performance, the maintenance of capacity is still an aspect to be optimized.

References

- [1] D. A. C. Brownson, C. E. Banks, Analyst 135 (2010) 2768
- [2] O. A. Vargas C., A. Caballero, J. Morales, Nanoscale 4 (2012) 2083
- [3] F. Bonino, S. Brutti, P. Reale, B. Scrosati, L. Gherghel, J. Wu, K. Müllen, Adv. Mater. 17 (2005) 743
- [4] A. Caballero, L. Hernán, J. Morales, ChemSusChem 4 (2011) 658
- [5] J. Hassoun, K. S. Lee, Y. K. Sun, B. Scrosati, J. Am. Chem. Soc. 133 (2011) 3139