Influence of graphene flakes morphology on the lithium ion storage capability

Haiyan Sun[a], Alberto Varzi[b], Duc Anh Dinh[a], Rinaldo Raccichini[b], Antonio Esau Del Rio Castillo[a], Roberto Cingolani[a], Vittorio Pellegrini[a], Bruno Scrosati[a], Stefano Passerini[b], and Francesco Bonaccorso[a]

[a]Graphene Labs, Istituto Italiano di Tecnologia IIT, Via Morego 30, Genoa, 16163, Italy
[b]Helmholtz Institute Ulm (HIU), Karlsruhe Institute of Technology, Helmholtzstrasse 11, 89081, Ulm, Germany
haiyan.sun@iit.it

Abstract

Graphene, a 2D crystal with large specific surface[1] area combined with its high electrical conductivity,[2] high mechanical strength,[3] ease of functionalization[4] and potential for mass production,[5] is an ideal material for energy applications.[6,7] In particular, graphene-based lithium ion batteries (LIBs) are considered as one of the most promising strategies[6] to boost the requirement of efficient energy-storage devices needed for the rapid development of portable devices. In this context, strong efforts have been dedicated on improving the performance of graphene-based LIBs.[6,7,8,9] However, thus far the focus was on both oxidized and functionalized flakes that normally suffer from large irreversible capacity and rapid capacity fade,[7] while, on the contrary, only a few works have exploited the full potential of pristine graphene flakes.[8-11] Moreover, the mechanism of lithiation/de-lithiation processes in graphene-based anode in LIB, as well as the effects of flakes size and edges on Li hosting capability is still unclear.[7,8,9]

In this work, we unravel the lithiation/de-lithiation processes of graphene-based anodes in LIBs linking the electrochemical performances of the anodes to the morphology (lateral size and thickness) of the graphene flakes. We first exfoliated graphite flakes by liquid phase exfoliation[5,12] and then sorted graphitic flakes by lateral size and thickness exploiting the sedimentation-based separation via ultracentrifugation[8,9], see Fig.a. Finally, the sorted graphitic flakes (4 samples) were deposited onto copper foils and used as anode in LIBs. The reduction of both lateral size and thickness of the graphene flakes is associated with an increase of the initial capacity from 591 to 1267mAh/g, see Fig.b, however, at the expense of columbic efficiency, possibly due to the trapping of Li^+ at the edges on the small graphene flakes.[6,8] Additionally, the shrinking of the lateral size and thickness of the flakes is associated with more specific capacity delivered at high potentials, which is highly unwanted in a battery, leading to a reduction from 41% to 25% of the energy efficiency. Our work provides guidelines for the exploitation of graphene flakes as anode material in LIBs.

References

[1] Stoller, M.D. et al., *Nano Lett.*, **8** (2008) 3498.

[2] Geim, A.K. and K.S. Novoselov, *Nature Mater.*, **6** (2007)183-191.

[3] Lee, C. et al.,Science, 385 (2008) 321.

[4] Ferrari, A.C., et al. *Nanoscale*, **7** (2015) 4598-4810.

[5] Bonaccorso, F. et al. Mater. Today **15**, (2012) 564.

[6] Bonaccorso, F. et al., Science, **2617** (2015) 1246501.

[7] Raccichini, R. et al,. *Nature Mater.*, **14** (2015) 271-279.

[8] Hassoun, J. et al., *Nano Lett.*, **14** (2014) 4901-4906.

[9] Sun, H. et al., *J. Mater. Chem.* A, (2016) 10.1039/C5TA08553E.

[10] Pappas, G.S., S. Ferrari, and C.Y. Wan, *Current Organic Chemistry*, **19** (2015) 1838-1849.

[11] Raccichini, R. et al. J. Power Sources, **281** (2015) 318-325.

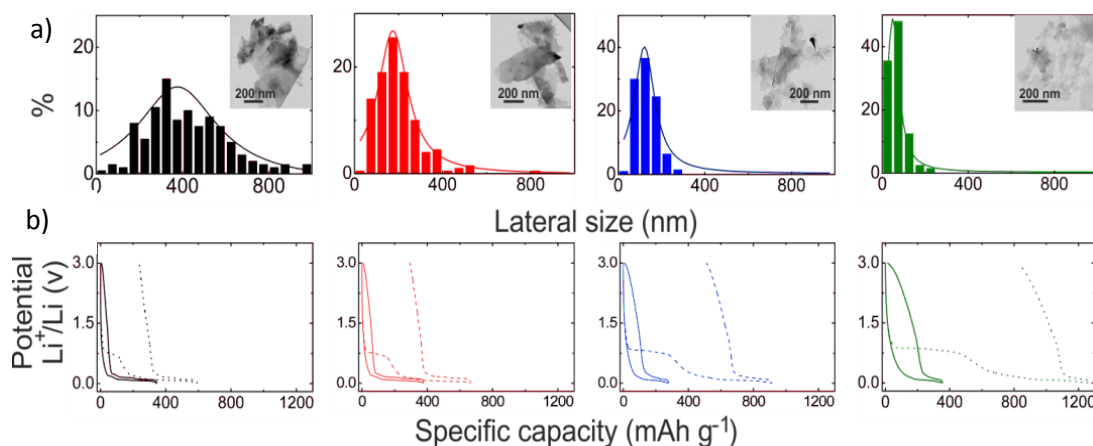[12] Hernandez, Y. et al., Nature Nanotech., **3** (2008) 563-568.

Figure: (a) Statistical lateral size distribution of the graphene flakes. Insets: TEM images; (b) Electrochemical properties of graphene anode with selected lateral size and thickness.