How much does size really matter? Exploring the limits of graphene as Li ion battery anode material

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Graphene has been recognised as a promising material for electrodes in Li-ion batteries (LIBs) thanks to its large surface area,[1] high electrical conductivity[2] and the potential for mass production.[3] So far, the Li storage of multi- (MLG) and single layer graphene (SLG) is still controversial.[4] Although MLG has shown some appealing features for niche applications,[5] there is no remarkable improvement in specific capacity with respect to graphite (372 $mAhg^{-1}$).[6] The Li storage of SLG is able to provide a high specific capacity of 744 $mAhg^{-1}$ thanks to the achievement of Li_2C_6,[7] but it is unstable upon cycling because only low Li occupancy levels can be achieved in SLG.[7,8] This raises a natural question. What about what lies in between? Is there a critical flakes size where both beneficial properties of graphite (e.g. low operating voltage) and SLG (high conductivity and short diffusion paths) are found? Is pristine graphene or few-layer graphene (FLG) a good active material for next-generation Li-ion batteries? Hence, our work unravel the role of flake dimensionality on the electrochemical performance of anodes based on FLG and MLG flakes prepared by liquid phase exfoliation of pristine graphite. The flakes with different lateral sizes and thicknesses, see figure1, are sorted by sedimentation-based separation,[8] finally, deposited onto Cu disks for the realization of four binder-free anodes.[9] The electrochemical data (figure1) shows that both initial specific capacity and irreversible capacity are increasing with the decrease in lateral size and thickness of flakes. For the anode based on small lateral size FLG flakes (<100 nm), we discover a detrimental effect on the average de-lithiation voltage, resulting on low voltage efficiency due to the preferential Li ions storage by adsorption rather than intercalation. Our study provides the guidelines for the practical exploitation of graphene-based electrodes.[10]

References

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Figures

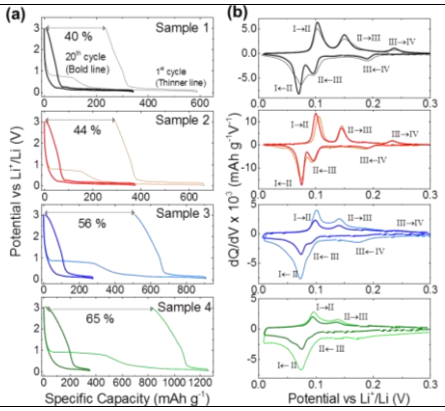

Figure 1: Electrochemical response of the four electrodes based on four batches of graphene flakes prepared at different centrifugation speeds, namely 2,000 rpm (Sample 1), 4000 rpm (Sample 2), 10,000 rpm (Sample 3), and 30,000 rpm (Sample 4). a) Voltage profiles and b) differential capacity plots for the 1st (dash) and 20th (solid) cycle (current rate: 0.1 Ag^{-1}).