

## Self-assembled SnO<sub>2</sub>/graphene composites with alternating oxide and amine layers for high Li-storage and excellent stability

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For the development of next-generation Li ion batteries (LIBs) with high energy densities, SnO<sub>2</sub>/graphene nanocomposite has been extensively studied as a promising alternative to graphite anodes<sup>1</sup>. Most traditional synthesis approaches for similar kind of composites rely on chemical mixing or by a random distribution of the constituent phases<sup>2,3</sup>. Several groups have recently come up with the layer by layer deposition technique to circumvent the problem. However these methods lack nanoscale spatial precision, time consuming and are difficult to extend to large scale synthesis. One simple, but powerful technique to achieve nanoscale control is to employ self assembly of metal ion anchored graphene with oppositely charged graphene sheets in solution in an orderly fashion to produce alternatively stacked metal oxide/graphene nanostructures.

We report a new facile route to fabricate densely stacked alternating layers of graphene and SnO<sub>2</sub> nanoparticles composite prepared from alternating stacks of Sn<sup>2+</sup>-anchored Graphene Oxide (GO) and amine-functionalized graphene (GN) as a next generation LIB anode. The synthesis route is driven by the oppositely charged Sn<sup>2+</sup>-anchored GO and GN layers which spontaneously form pre-aligned alternating stacks (SG/GN) in aqueous solution upon carefully tuning the pH. This allows the (SG/GN) composite to be easily collected and thermally reduced (Fig.1).

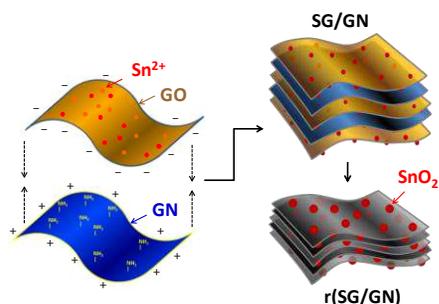


Fig.1. Schematic diagram describing the concept of this work.

The thermal reduction of (SG/GN) results in a well-organized and densely packed layered SnO<sub>2</sub>/graphene nanostructure r(SG/GN) which displays an unprecedented degree of high and stable reversible capability with excellent capacity retention (872 mAh·g<sup>-1</sup> after 200 cycles at a rate of 100 mA·g<sup>-1</sup>) in contrast to rSG (Fig.2) which was synthesized under same synthetic conditions and contained an identical amount of SnO<sub>2</sub>.

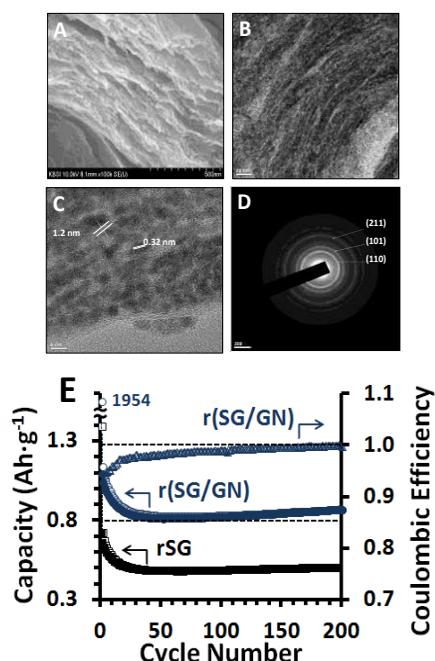


Fig.2. (A) FESEM image of r(SG/GN), (B) Bright field HRTEM image of the ultramicrotomed cross-section of r(SG/GN), showing the layered structure of a SnO<sub>2</sub>-graphene nanocomposite. (C) High-magnification HRTEM image of r(SG/GN). (D) SAED pattern of r(SG/GN), and (E) Capacity retention and coulombic efficiency of rSG and r(SG/GN) during 200 cycles at 100 mA·g<sup>-1</sup>.

The densely stacked structure in r(SG/GN) suppressed volume change for Li<sup>+</sup> during prolonged charge-discharge cycles (Fig.3) and maintained a smooth and regular surface in contrast to rSG (rough and pulverized).

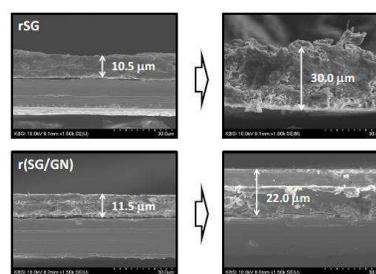


Fig.3. Cross-sectional FESEM images of rSG and r(SG/GN), comparing volume expansion at a fully lithiated state after 100 cycles.

The facile in-situ solution-based self-assembly procedure presented in this work can provide an effective platform that would be applicable to other diverse graphene layered nanocomposites.

### References

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