
CHEMICAL DERIVED GRAPHENE ASSEMBLIES AND HYBRIDS FOR ADVANCED ENERGY STORAGE

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As a one-atom-thick two-dimensional crystal with unique electronic, mechanical, optical, and thermal properties, graphene has attracted great attention from scientific community. Graphite, a natural mineral compound formed after aeons of harsh geological process underground, is fundamentally composed of millions of graphene sheets stacking together. It is believed to be a low cost, easy accessible resource for scale-up graphene production, because functional graphene nanoplates with single to several layers of graphene could be easily exfoliated from graphite oxide through mild ultrasonication or thermal expansion.

The chemically derived graphene sheets are often decorated by abundant intrinsic active sites, such as functional groups (mainly -OH, -COOH, -C=O, -C-O-C-), lattice defect (atom vacancy, distortion, dangling bonds) on the lateral surface and edge. On one hand, the introduction of functional groups increased the wettability and solvent compatibility of graphene, which offers an opportunity for macroscopic ordered assembly of graphene in solution based approach. On the other hand, the active sites could be further modified by surface reaction with heteroatoms such as nitrogen, boron, and phosphorus, so as to tune the acid/base property and electronic structure of graphene. These specific advantages provided tunable surface chemistry of graphene for advanced energy conversion and storage.

The present research has been carried out following generally two mainlines including graphene assemblies from graphene oxide and graphene hybrids from reduced graphene oxide (as shown in Figure 1). The highlights of our work are as follows,

a) Graphite oxide was an element for graphene based advanced functional materials. Flexible, semi-transparent, and free-standing graphite oxide membranes (Figure 2a) could be fabricated at the liquid/air interface through a facile and efficient self-assemble method. Such macroscopic membranes are constructed from individual graphene oxide sheets by layer-by-layer stacking and show excellent mechanical and optical performance.

b) To efficiently produce chemical-exfoliation-based production of graphene starting from graphite oxide, the preheated high-temperature environment is believed to be very critical. By introducing a high vacuum to the exfoliation process, high quality graphene sheet (Figure 2b) is available at a mild temperature, the exfoliated graphenes demonstrate an excellent energy storage performance, and the electrochemical capacitance is much higher than that of the high-temperature exfoliated ones. This vacuum exfoliation approach presents us with

a possibility for a mass production of graphenes at low cost and great potentials in energy storage applications of graphene-based materials.

c) Graphene-based architectures have attracted much attention as a viable support material to improve the efficiency of various catalytic reactions, energy conversion and storage. The abundant functional groups, lattice defect provided tunable surface chemistry of graphene sheets. However, graphene sheets are always tending to form irreversible agglomerates through the van der Waals interaction. To demonstrate the extraordinary performance of the graphene, novel type hybrid composites by combining nanomaterials with distinct structures and dimensions with unexpected properties and unique applications was a good solution. We proposed two strategies to fully demonstrate its potential for energy storage. The first route is introducing the secondary phase to separate graphene sheets. For instance, in present research, tin, which has a theoretical capacity of 992 mAhg^{-1} , has been proposed as one of the most promising anode materials for the coming decades and selected as the separator. A SnO_2 /graphene hybrid was explored for energy storage application. On the other hand, the graphene sheet was mediated by N, P, B doping. As an electron donor and acid-base adjuster, nitrogen modification has been proved to be a valid way to tune the chemical and electrical environment of carbon surface. The N modified nanocarbon materials often exhibits an exceptional property towards anchoring and nucleation of active species, such as metal ions, clusters and coordinated complex in solution based process. We expected that doped graphene was a novel platform for advanced graphene based materials for energy storage.

Through those efforts, graphene based assemblies and hybrids are expected to a novel platform for advanced energy conversion and storage for sustainable society.

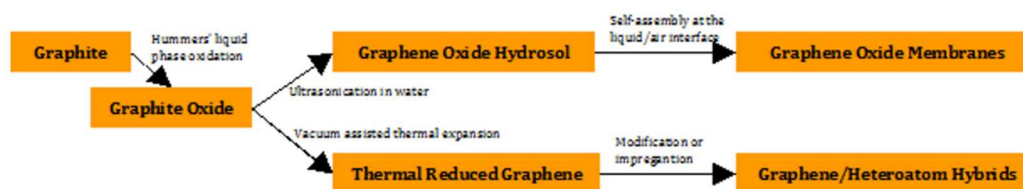


Figure 1: Route map of graphene based assemblies and hybrids for advanced energy conversion and storage

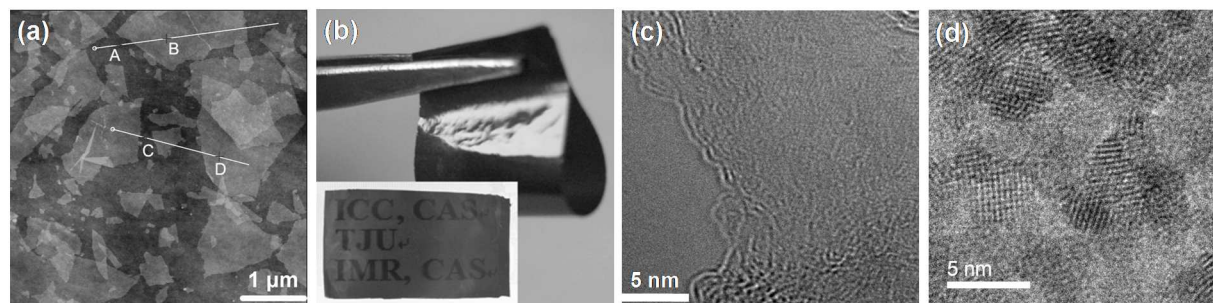


Figure 2: (a) AFM image of graphene oxide; (b) Self-assembled free-standing graphite oxide membrane; (c) HRTEM image of thermal reduced graphene oxide; (d) HRTEM image of SnO_2 @RGO hybrids

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