

# Controllable synthesis of sandwich-like graphene-supported structures for energy storage and conversion

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**Abstract:** This paper reviews recent progress on the controllable synthesis of sandwich-like graphene-supported structures for energy storage and conversion by harnessing the two-dimensional structure of graphene. These structures could be divided into three major categories: graphene-supported nanoparticles (0D), graphene-supported nanorods, nanowires or nanoribbons (1D), and graphene-supported nanoplates (2D). In these structures, the intrinsic incompatibility between graphene and the functional materials was circumvented by modifying or functionalizing the graphene or graphene oxide. A graphene intermediate provides a fast electron-transfer pathway for energy storage and conversion. It also gives a way to design and fabricate sandwich-like graphene and even graphene-analogue-supported functional materials with well-defined structures for broad applications such as catalysts, sensors, energy storage and conversion.

**Key Words:** Graphene; Sandwich-like structure; Controllably synthesis; Lithium ion batteries; Fuel cells

## 1 Introduction

Graphene is a monolayer of carbon atoms, and possesses an ultrahigh specific surface area, superior electrical conductivity, excellent thermal and chemical stability, and superior mechanical strength, which is anticipated for efficient energy storage and conversions, sensors, as well as other applications since its emerging at 2004<sup>[1]</sup>. Moreover, associated with many merits, graphene has been employed as matrix for abundant functional materials with a high catalytic activity or superior energy storage capability, such as platinum<sup>[2]</sup>, SnO<sub>2</sub><sup>[3]</sup>, Co<sub>3</sub>O<sub>4</sub><sup>[4,5]</sup>, and Mn<sub>3</sub>O<sub>4</sub><sup>[6]</sup>, MoS<sub>2</sub><sup>[7]</sup> to further enhance the electrochemical performances either for energy conversion or storage.

To harness the merits of graphene, an efficient one is to grow functional materials on both sides of graphene to generate sandwich-like graphene-supported architectures<sup>[8,9]</sup>. For the well-defined architectures, electron could be transferred easily through graphene, self-aggregation and random stacking of the functional materials and graphene nanosheets were alleviated, and volume changes of functional materials during electrochemical reaction were also accommodated. Therefore, controllable synthesis of sandwich-like graphene-supported architectures is an important strategy to improve the electrochemical performances of functional materials for energy storage and conversions.

As graphene is hydrophobic and inert, and most of the functional materials is hydrophilic, the intrinsic incompatibility of these two classes of materials resulted in the random stacking and self-aggregation, many approaches were developed to solve the problem by making use of electrostatic effect,  $\pi$ - $\pi$  interactions, hydrogen bonding and so on. In this paper, sandwich-like graphene-supported architectures for energy storage and conversions, including their controllable synthesis approaches and electrochemical performances, are systematically reviewed. These architectures are divided into three major categories based on their nanostructures: graphene-supported nanoparticles (0D), graphene-supported nanorods, nanowires or nanoribbons (1D), and graphene-supported nanoplates (2D). This would be beneficial for the development of the well-defined two-dimensional architectures with broad applications.

## 2 Controllable synthesis of graphene

Since single-layer graphene was first synthesized by Geim's group via mechanical exfoliation of graphite at 2004, numerous methods such as liquid-phase exfoliation, chemical vapor deposition (CVD), arc discharge and oxidation-reduction were widely developed to synthesize single-layer or multi-layer graphene<sup>[10-13]</sup>. At present, oxidation-reduction method is the most widely used one owing to its cost-effective and "easy to scale-up" properties. With this method, reduced graphene oxides (rGOs), also

Received date: 05 Dec 2016; Revised date: 26 Jan 2017

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DOI: 10.1016/S1872-5805(17)60101-X

called graphene nanosheets, were typically synthesized through chemical oxidation of graphite and the subsequent chemical or thermal reduction<sup>[11-15]</sup>. The high-resolution microstructure of graphene, GO and rGO sheet were presented in Fig. 1<sup>[16]</sup>. However, strong acid and strong oxidant were used during the process, which would result in the environmental pollution, furthermore, the crystalline of the graphene was destroyed and the conductivity was reduced notably.

Atomically precise graphene nanoribbons could be controllably synthesized from organic molecules through coupling reaction and the subsequent cyclodehydrogenation<sup>[17,18]</sup>. For instance, as depicted in Fig. 2(a), graphene nanoribbons were successfully synthesized from 10,10'-dibromo-9,9'-bianthryl monomers, whose microstructures were depicted in Fig. 2(b,c)<sup>[17]</sup>. Nevertheless, only nanoribbons other than large size of graphene could be

prepared, it is the shortcoming of this method for practical applications.

Another effective method is CVD method, graphene with few layers and large area could be fabricated by segregation reaction or decomposition of hydrocarbons<sup>[19-22]</sup>. However, the problems of complicated process and high cost restrict its development. In addition, graphene could be synthesized electrochemically in large scale<sup>[23-25]</sup>. For instance, as displayed in Fig. 3(a), with sulfate aqueous solution as electrolyte, graphene nanosheets were obtained through electrochemical exfoliation of graphite, the microstructure images of which were depicted in Fig. 3(b,c)<sup>[23]</sup>. Compared with aforementioned methods, it is a cost-effective, simple but efficient method to controllably synthesize graphene, which is a potential way to prepare graphene nanosheets in industrial scale.

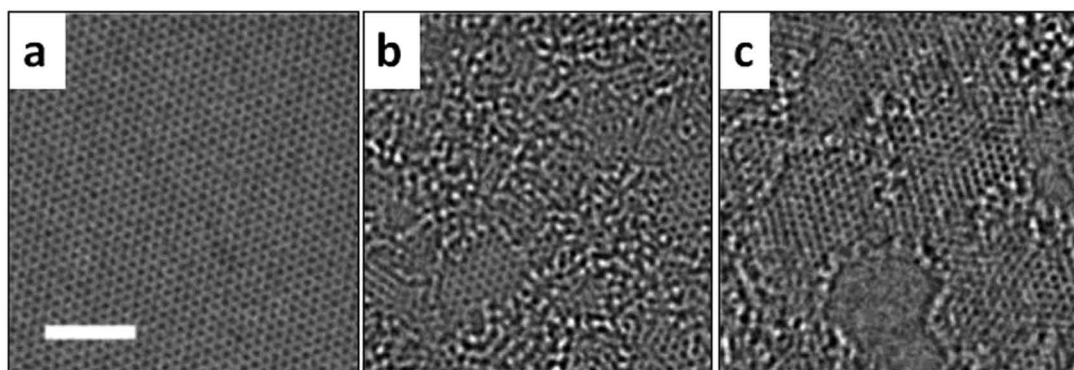


Fig. 1 (a) Aberration corrected TEM images of graphene sheet, (b) GO sheet and (c) rGO sheet, scale bar was 2 nm for all images<sup>[16]</sup>.

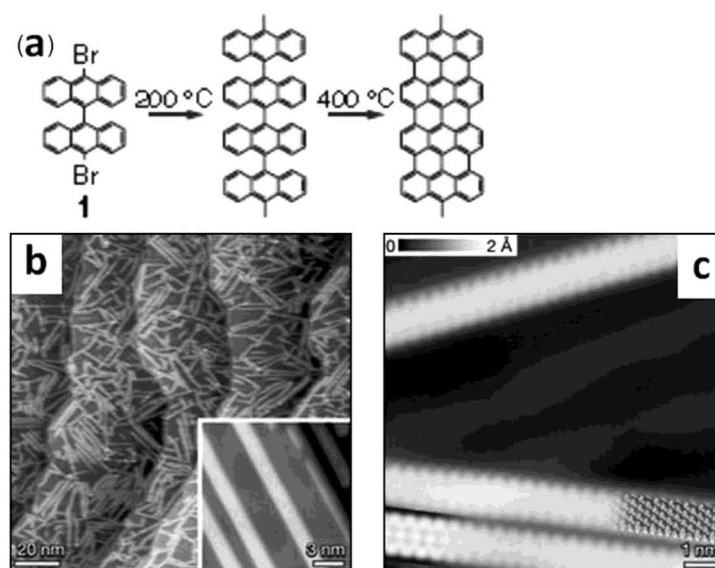


Fig. 2 (a) Reaction scheme from 10,10'-dibromo-9,9'-bianthryl monomers to graphene nanoribbons, (b) STM and (c) HR-STM of the graphene nanoribbons<sup>[17]</sup>.

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