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2D and 3D graphene materials: Preparation and bioelectrochemical applications

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ABSTRACT

The attractive properties of graphene materials have stimulated intense research and development in the field of bioelectrochemistry. In particular, the construction of 2D and 3D graphene architectures provides new possibilities for developing flexible and porous carbon scaffolds, which not only inherit some of the key properties of individual graphene sheets, but also develop additional functions that are of considerable interest for bioelectrochemical applications. In this review article, we will first summarize the recently developed approaches to preparing graphene sheets, and then focus on the methods to assemble them into macroscopic 2D and 3D structures. Furthermore, we will highlight the potential applications of these materials in electrochemical biosensors and biological fuel cells.

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1. Introduction

The successful isolation of high quality graphene with the size of hundreds of microns from graphite in 2004 (Novoselov et al., 2004) aroused tremendous efforts towards investigating the unique properties, controlling the growth, introducing diverse functionalities, and eventually exploring the potential applications of graphene materials (Novoselov et al., 2012; Qian et al., 2011a; Y.W. Zhu et al., 2010a). In particular, graphene derivatives have been actively studied in the field of electrochemistry because of their unique physical and chemical properties in comparison with other carbon materials, such as large specific surface area (2630 $m^2 g^{-1}$) (Stoller et al., 2008), superior electrical conductivity (200 $S m^{-1}$) (Stankovich et al., 2007; Yoo et al., 2011), excellent thermal stability with oxidation resistance temperature up to 601 °C (Wu et al., 2009b), high thermal conductivity between 3080 and 5150 $W m^{-1} K^{-1}$ (Ghosh et al., 2008), remarkable mechanical strength with Young's modulus of around 1.0 TPa (Lee et al., 2008), and outstanding optical transmittance of 97.7% (Nair et al., 2008). Moreover, it also shows fascinating electrochemical properties, including wide electrochemical potential windows, low charge-transfer resistance, and excellent electrochemical activity (Chen et al., 2014; Qian et al., 2011b; Shao et al., 2010b). The aforementioned physical and electrochemical properties enable the widespread use of graphene in the field of bioelectrochemistry, and possibly offer new solutions to address the current energy and healthcare issues that are of great importance for the sustainable development of the society.

Chemically derived graphene, including graphene oxide (GO) and reduced graphene oxide (rGO), are promising building blocks to construct 2D and 3D materials (Bi et al., 2012; Cheng and Li, 2013). In addition to the inherent properties of individual graphene sheets, versatile functionalities can be built in the hierarchical graphene materials for improved performance (Chen et al., 2013; Ren and Cheng, 2013). In comparison with the extensive research on individual graphene sheets, less attention has been focused on 2D and 3D graphene materials. One of the goals of this review is to increase the visibility of the new class of hierarchical materials and call attention for further research from academia and industry. From a practical point of view, the use of these materials into practical applications relies on the development of efficient preparation procedures that allow for tailoring their structures and functions for improved performance. We will first summarize the approaches for preparing chemically derived graphene, with particular focuses on the recently developed environmentally-friendly methods, and then summarize the strategies to assemble individual graphene sheets into 2D and 3D structures. Their bioelectrochemical applications will be discussed afterwards with an emphasis on the specific topics of electrochemical biosensors and biological fuel cells.

2. Preparation of graphene sheets

Although the scotch-tape method can directly produce high-quality graphene, it is lack of the feasibility of functionalization and not suitable for large-scale production. A diverse range of approaches, therefore, have been developed to produce graphene, including epitaxial growth (Sutter et al., 2008), chemical vapor deposition (CVD) (Mattevi et al., 2011), liquid-phase exfoliation (Hernandez et al., 2008), chemical exfoliation (Dreyer et al., 2010), and organic synthesis (Wu et al., 2007). Among these methods, chemical exfoliation has attracted widespread interest because it does not rely on special equipments and allows for scaled-up preparation of GO carrying a high density of functional groups, which open the possibility to introduce additional functionalities (Park and Ruoff, 2009). This route is referred as oxidation–exfoliation–reduction process, which generally involves chemical oxidation of graphite into graphite oxide, followed by exfoliation of graphite oxide into single-layered GO, and reduction of GO to rGO with low oxygen to carbon atomic ratio (Fig. 1) (Bai et al., 2011).

2.1. Preparation of GO

The precursor of GO is graphite oxide which contains oxygen functionalities and retains stacked structure similar to that of graphite. The history for preparing graphite oxide can be traced back to 1859, when $KClO_3$ and fuming HNO_3 were used to oxidize graphite powder, leading to a material with carbon to oxygen ratio around 2:1 (Brodie, 1859). The utilization of $KClO_3$ and HNO_3 must be handled with special caution because of the generation of highly toxic ClO_2 gas and the possible production of explosion (Compton and Nguyen, 2010). Hummers and Offeman (1958) developed an approach to synthesize graphite oxide with similar level of oxidation but employing different oxidation reagents ($NaNO_3$, $KMnO_4$, and concentrated H_2SO_4) in 1958. Through Hummers' method, the reaction can be completed within relatively shorter reaction time while avoiding the generation of hazardous ClO_2 gas. A recent study revealed that the amount of basal plane in graphite oxide can be improved by eliminating $NaNO_3$, increasing the amount of $KMnO_4$, and performing the reaction in a 9:1 mixture of H_2SO_4 and H_3PO_4 (Marcano et al., 2010). The exfoliation of graphite oxide into single-layered GO can be achieved

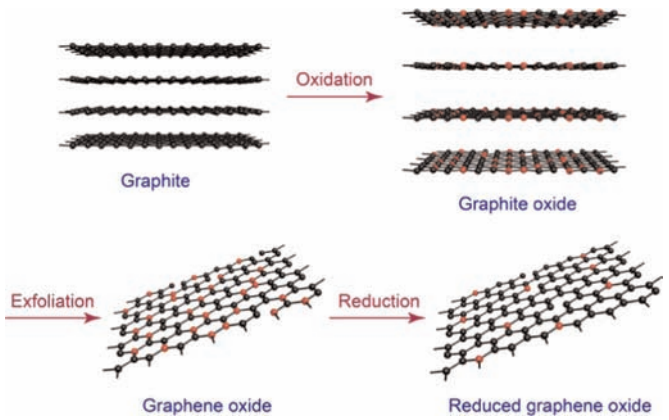

Fig. 1. Schematic illustration of the chemical oxidation, exfoliation, and reduction methods to prepare reduced graphene oxide from graphite.

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