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Recent advances in the synthesis and modification of carbon-based 2D materials for application in energy conversion and storage

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Graphene, reduced graphene oxide (rGO) and derived materials have emerged as promising solutions for applications in renewable energy storage/conversion devices. No alternatives are known to simultaneously exhibit large specific surface area, high electrical conductivity, good chemical stability, high mechanical strength and flexibility. This review article is a collection of some of the most relevant research efforts published in the last few years focusing on the synthesis and modification of graphene/rGO as well as doped and hybrid bi-dimensional carbon materials. For research on graphene growth, the choice of precursor and physical state (gas, solid or liquid) has been proved to be as important as the environment and synthesis approach. On the other hand, research focused on graphene oxide reduction has relied on the development of simple and efficient techniques for rGO conversion and device structuring. Modifications applied to graphene (during synthesis) or rGO (during reduction) have included doping, decoration with nanoparticles and the formation of composite microstructures. Fabrication of electrodes based on graphene/rGO for application in energy storage and conversion has been reported, including relevant performance data from real devices (supercapacitors, lithium ion batteries, fuel cells or solar cells). This review concludes with a brief discussion of some of the possible directions for promising research in the area of graphene/rGO fabrication for energy conversion and storage devices.

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1. Introduction

Since its discovery, graphene has been the most extensively studied 2D material due to its exceptional properties, such as high theoretical surface area (2600 m^2 g^{-1}) [1], excellent thermal conductivity (4840–5300 W m^{-1} K^{-1}) [2], extreme mechanical properties (tensile strength up to 130 GPa, elastic modulus of 1000 GPa) [3,4] and ultra-high electron mobilities (2×10^5 cm^2 V^{-1} s^{-1}) [5,6]. Graphene is made up of sp^2 hybridized carbon atoms bonded together in a one-atom thick plane consisting of a repeating pattern of hexagons [7,8]. Mass production of graphene remains an elusive goal, due to expensive and difficult processing. In this context, graphene oxide (GO), with oxygen-containing groups attached to the graphene sheets (GSs) as shown in Fig. 1, has attracted the attention of researchers [9–11]. GO is usually produced by the chemical oxidation of graphite with subsequent dispersion and exfoliation [10]. The presence of oxygen-containing functional groups in the basal plane (epoxy and hydroxyl) and the sheet edges (carbonyl and carboxyl) allows GO to interact with a wide range of metal oxides, so that functional hybrids and composites with unusual properties can be utilized for energy storage [12-16]. These functional groups render GO strongly hydrophilic, generating good dispersibility in many liquids and providing potential advantages for easier device processing [17–20]. The oxygen-containing groups on GO surfaces strongly affect its electronic, mechanical, and electrochemical properties [21]. By promoting the total or partial reduction of GO, a material akin to graphene and exhibiting similar property values can be generated; this reduction product is named reduced graphene oxide (rGO). Alternatively, the functional groups in GO may work as sites for chemical modification or functionalization, further extending the scope for achieving a wide range of chemical and physical properties.

There are some well-known approaches for the reduction of GO utilizing thermal energy (thermal annealing, microwave and photoreduction) and chemical energy (chemical reduction, electrochemical reduction, solvothermal reduction and photocatalytic reduction). These are frequently used for removal of oxygen-containing functional groups from GO surfaces [22]. Photoreduction and photocatalysis provide excellent alternatives for replacing harsh chemical and high temperature treatments for GO reduction [23–29]. Schematic representations of graphite, graphene and GO containing epoxide, hydroxyl, carbonyl and carboxyl groups are shown in Fig. 1a–c [30].

Research on environment-friendly energy conversion and storage devices and their practical application has attracted increasing attention in the past decades [31,32]. Lithium ion batteries (LIBs), supercapacitors (SCs), fuel cells and solar cells are some of the most attractive devices which are currently under intense development [33–37]. Fig. 2 shows the synthesis of graphene/rGO applying various approaches and its application for different kind of energy storage/conversion. Graphene, rGO and related hybrids with unique targeted properties have an important role to play as enabling materials in this vigorous research effort.

In this work we tried to provide a comprehensive review of the latest developments in the synthesis of graphene and the process of GO reduction. As the knowledge about the factors involved in the formation of graphene evolved along the years, the synthetic strategies started to incorporate a combination of operations, becoming

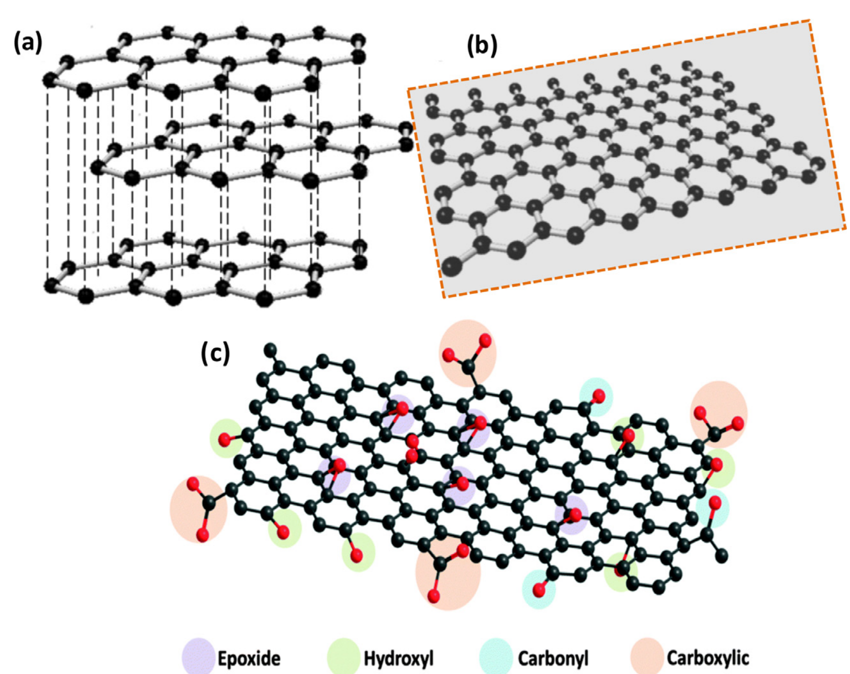

Fig. 1. (a) Graphite, which can be viewed as a stack of graphene layers and (b) graphene, a single layer honeycomb lattice of carbon atoms [30]. (c) GO containing functional groups such as epoxide, hydroxyl, carbonyl and carboxyl [9]. Reprinted (adapted) with permission from Refs. [9, 30]. Copyright INTECH Publisher (2013) and The Royal Society of Chemistry (2016).