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REVIEW

Energetic graphene oxide: Challenges and opportunities

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Summary Motivated by both its graphene-oriented applications and its own remarkable properties, interest in graphene oxide (GO) has widely spread across many disciplines. In parallel to the rapid progress of research, industrial-scale production of GO has emerged. GO is highly energetic, thermally unstable and can readily undergo exothermic disproportionation reactions to produce chemically modified graphene under mild heating conditions. This Review highlights the challenges and opportunities associated with GO's thermal instability such as the potential fire risk during large scale production and methods of mitigation, energy efficient way to reduce GO, photothermal patterning and sintering of graphene/polymer composites, and new syntheses using GO as an in situ power source to make nanoparticle decorated graphene composites for energy storage and catalysts.

Introduction: graphene oxide: new interest in an old material

Graphite oxide sheets, now called graphene oxide (GO) is the product of chemical exfoliation of graphite that has been known for more than a century [1—3]. It is typically synthesised by reacting graphite powders (Fig. 1a) with strong oxidizing agents such as $KMnO_4$ in concentrated sulfuric acid [4,5]. The oxidation of graphite breaks up the extended two-dimensional (2D) π-conjugation of the stacked graphene sheets into nanoscale graphitic sp^2 domains surrounded by disordered, highly oxidised sp^3 domains as well as defects of carbon vacancies. The resulting GO sheets are derivatized by carboxylic acid at the edges, and phenol, hydroxyl and epoxide groups mainly at the basal plane (Fig. 1b) [6,7]. Therefore, the sheets can readily exfoliate to form a stable, light brown coloured, single layer suspension in water [8,9]. This severe functionalization of the conjugated network renders GO sheets insulating. However, conductivity may be partially restored conveniently by thermal [10] or

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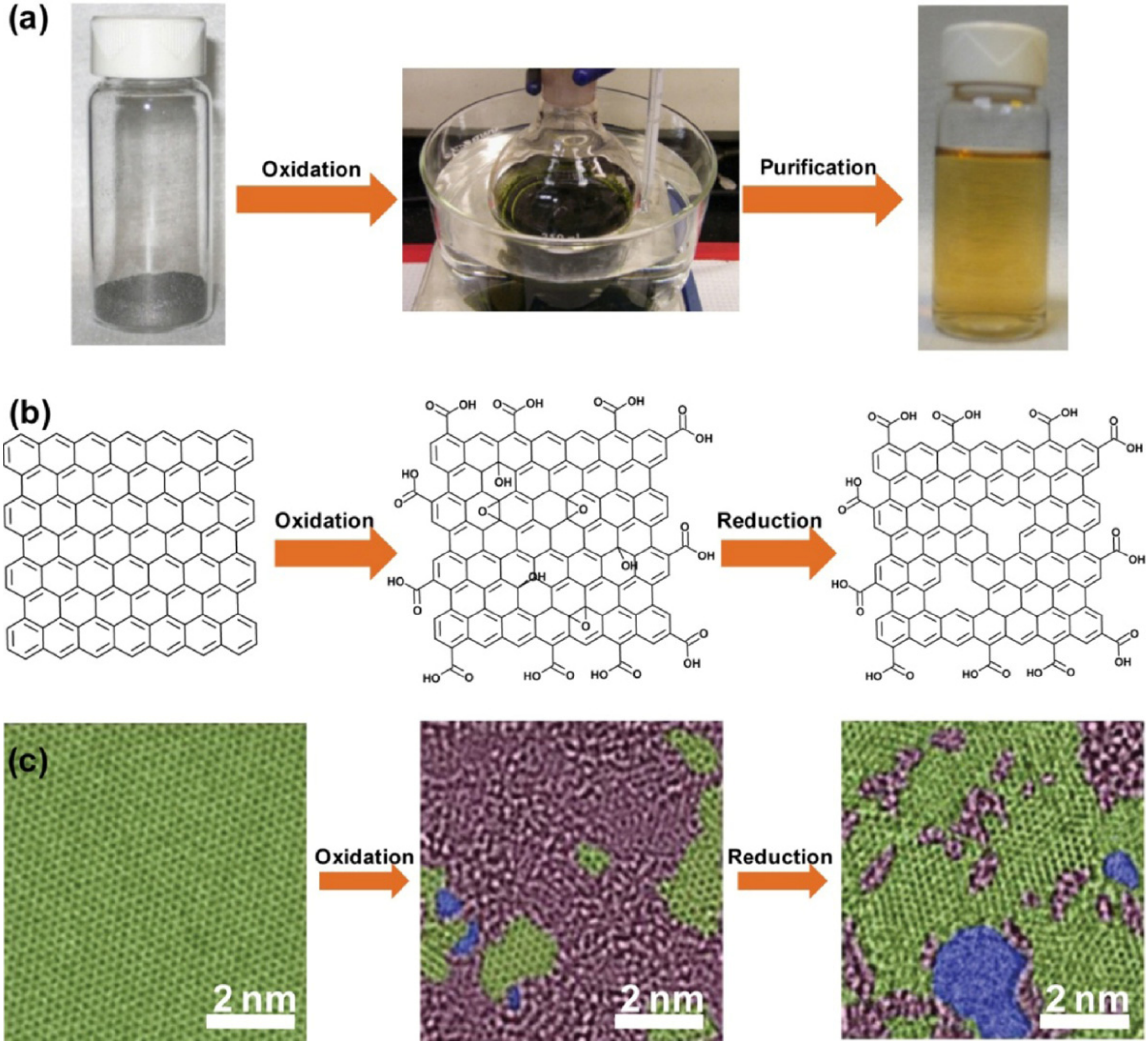

Figure 1 Synthesis, structural model and microstructures of GO sheets. (a) GO is typically synthesised by reacting graphite powder with strongly oxidizing agents such as $KMnO_4$ in concentrated H_2SO_4, followed by purification and exfoliation in water to yield a colloidal dispersion of single layers. (b) Structural model of graphene (left), GO (middle) and its reduction product r-GO. GO is insulating due to broken conjugation in the basal plane. After reduction, r-GO becomes conductive but is still a very defective structure compared to graphene. (c) Colour coded high resolution TEM images showing the atomic structures of graphene (left), GO (middle) and r-GO (right). The green, purple and blue areas depict ordered, graphitic sp^2 domains, disordered highly oxidised sp^3 domains, and holes on the sheet, respectively.
[(c) is adapted from [33]. Copyright 2010 Wiley-VCH].

chemical treatment [8], producing chemically modified graphene sheets. Therefore, interest in this century-old material has resurged after 2004's discovery on graphene [11]. The oxidization—exfoliation—reduction cycle effectively makes the insoluble graphite powders processable in water, enabling many ways of using the conducting graphene or reduced GO (r-GO) products [12,13]. Although the resulting graphene product or r-GO is more defective and thus less conductive than pristine graphene [14], it is sufficiently conductive for many applications such as electrodes for various energy devices [15—19]. Therefore, the ease of synthesizing GO and its solution processability have made it a very attractive precursor for large scale production of graphene in applications including transparent conductors [16,20—22], chemical sensors [23,24], biosensors [23], polymer composites [25,26], battery and ultracapacitors [15] as described in a number of review articles [5,14—16,21,23,25,27—30].

Apart from making graphene, GO itself has many intriguing properties. Like graphene, GO sheets are characterised by two abruptly different length scales. The measured thickness is of typical molecular dimensions (ca. 1 nm) [8], but the lateral dimensions are those of common colloidal particles, ranging from nanometers [31] up to hundreds of micrometers [32]. Therefore, GO sheets can be viewed as either molecules or particles, depending on which length scale is of greater interest. On the other hand, GO can be characterised as an unconventional *soft* material such as a 2D polymer, anisotropic colloid, soft membrane, liquid crystal, or even amphiphile. A few examples are given below, where GO is treated as surfactant, diblock copolymer, and liquid crystal. An earlier structural model [6] and