

Two dimensional polymerization of graphene oxide: Bottom-up approach



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H I G H L I G H T S

- We demonstrate a bottom-up synthesis of structures similar to graphene oxide.
- The proposed underlying reaction mechanism is aldol condensation.
- The synthetic crystallites are characterised by physical methods.

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We demonstrate a bottom-up synthesis of structures similar to graphene oxide via a two dimensional polymerization. Experimental evidence and discussion are conveyed as well as a general framework for this two dimensional polymerization. The proposed morphologies and lattice structures of these graphene oxides are derived from aldol condensation of alternating three nucleophilic and three electrophilic centers of benzenetriol.

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1. Introduction

The synergy between unmatched electrical, optical and mechanical properties of graphene and graphene oxide (GO) has resulted in vigorous research into methods for their large-scale production [1–3].

Sheets of GO with atomic thickness have established themselves as a new carbon-based nanoscale material with an optical band gap of 1.7 eV–2.1 eV [4] and soluble in water and other solvents which allows it to be spray or spin coated [5,6]. Controlled oxidation provides tunability of its electronic and mechanical properties up to

the point of its turning into the semi-metallic graphene upon complete removal of the C–O bonds. Therefore, we assume the synthesis of GO via a two dimensional polymerization a successful attempt at bottom-up synthesis of graphene.

In general, chemical oxidation methods such as Brodie's [7], Staudenmaier's [8], Hummers' [9] or a variation of these [10], produce GO by introducing functional groups in between the layers forming graphite and they peel off. However, it represents a top-down approach.

In this communication we suggest a bottom-up approach towards GO synthesis. We start from a simple monomer and through a tailored two dimensional polymerization arrive at GO platelets.

Consider the monomer unit in Fig. 1a. This unit contains alternating functional groups A and B attached to a benzene or cyclohexane carbon ring. Imposing a polymerization rule that depends

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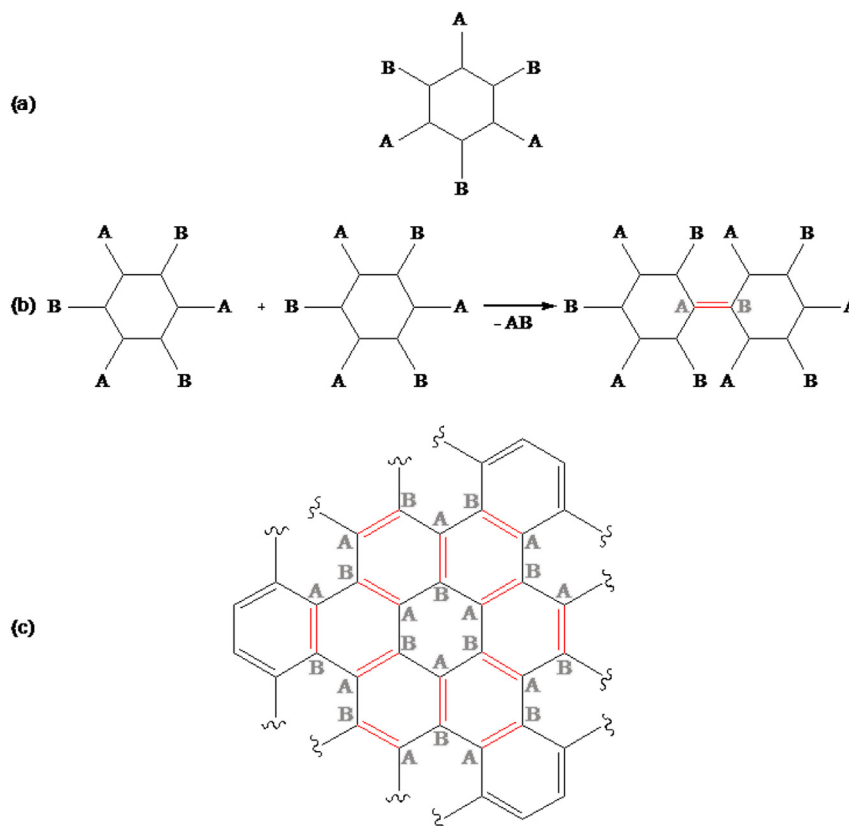


Fig. 1. (a.) Monomer unit; (b.) Polymerization rule; (c.) Upon polymerization the bonds formed between the benzene rings span a graphene plane.

on the chemical reaction between A and B we can produce the step depicted in Fig. 1b. The newly formed bond (depicted in color) appears in an arbitrary position in the ring with equal probability for all positions. The letters in the ring denote the positions functional groups had prior to the reaction. Given this rule, one is tempted to assume that the A and B functional groups above and below the bond would also react. However, they are attached with covalent bonds to an sp^2 hybridized carbon atom and these bonds are rigid. Such a reaction would require their bending and this is energy prohibitive. More probable is the reaction with an additional monomer unit. When this occurs we arrive at the lattice represented in Fig. 1c. The hexagonal symmetry is present in this newly formed structure. The bonds formed during the polymerization are depicted in color. The key to the understanding of the formation of this lattice is the observation that the intermediate (to the monomer rings) hexagons are spanned by three newly formed bonds. If one traces the central monomer hexagon and the surrounding it six monomer hexagons, one is convinced that this is the only possible configuration the original monomer units undergone two dimensional polymerization can arrange in space.

2. Results and discussion

The particulars behind the experimental test of the general framework are dependent on the choice of the functional groups A and B reacting to form the lattice pattern of graphene or GO. One possible choice for the monomer in a proof-of-concept experiment is 1,3,5-trihydroxybenzene (phloroglucinol) given in Fig. 2a and the chemical reactions behind the polymerization (more appropriate here is polycondensation) are nucleophilic additions to carbonyl groups.

The feasibility of the two dimensional polymerization using this monomer and GO in mind as the final product must be addressed in

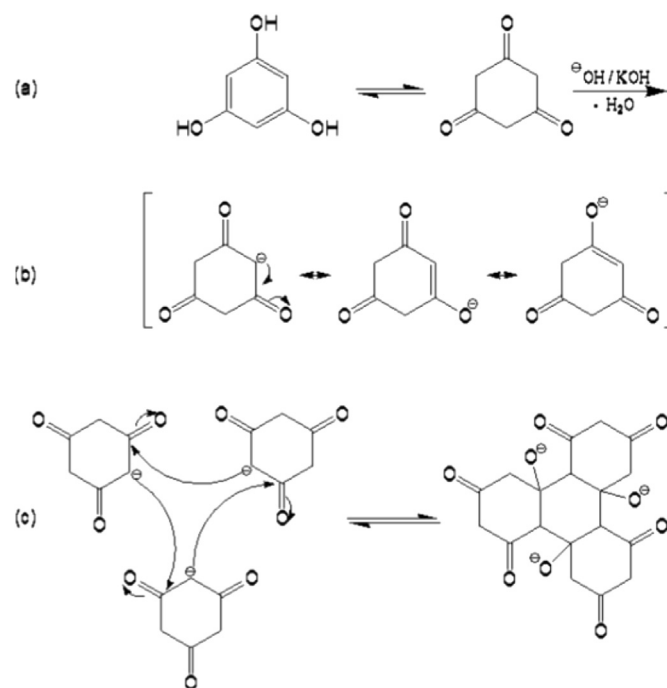


Fig. 2. Proposed synthesis of GO (a.) 1,3,5-trihydroxybenzene a phenol-like substance whose oxo- and enol-forms represent our monomer; (b.) deprotonation in typical for aldol-condensation conditions; (c.) polycondensation in a hexagonal planar lattice.

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