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# Graphene-based materials: Synthesis and gas sorption, storage and separation



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## ABSTRACT

Graphene-based materials have generated tremendous interest in a wide range of research activities. A wide variety of graphene related materials have been synthesised for potential applications in electronics, energy storage, catalysis, and gas sorption, storage, separation and sensing. Recently, gas sorption, storage and separation in porous nanocarbons and metal–organic frameworks have received increasing attention. In particular, the tuneable porosity, surface area and functionality of the lightweight and stable graphene-based materials open up great scope for those applications. Such structural features can be achieved by the design and control of the synthesis routes. Here, we highlight recent progresses and challenges in the syntheses of graphene-based materials with hierarchical pore structures, tuneable high surface area, chemical doping and surface functionalization for gas (H<sub>2</sub>, CH<sub>4</sub>, CO<sub>2</sub>, N<sub>2</sub>, NH<sub>3</sub>, NO<sub>2</sub>, H<sub>2</sub>S, SO<sub>2</sub>, etc.) sorption, storage and separation.

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## 1. Introduction

Graphene is a two-dimensional (2D) sp<sup>2</sup> bonded carbon sheet, arranged in a hexagonal honeycomb lattice [1–4]. From a fundamental point of view, graphene is nothing but a single layer of graphite, which is an infinite three-dimensional (3D) material made up of stacked layers of graphene. The layers in graphite interact weakly through van der Waals (vdW) forces. In terms of properties graphene is unique; it is a soft membrane and at the same time possesses a high Young's modulus, and good thermal and electrical conductivities [4–6]. In addition, a single-layer graphene is a zero band gap material and highly transparent, exhibits optical transmittance of 97.7%. With its high theoretical specific surface area of ~2600 m<sup>2</sup>/g graphene provides a rich platform for surface chemistry [7–12]. The combined extraordinary physical and chemical properties of graphene, in turn, has ignited extensive research in nanoelectronics, supercapacitors, fuel-cells, batteries, photovoltaics, catalysis, gas sorption, separation and storage, and sensing [13–28]. A roadmap of graphene materials is described in a recent review [4]. It is important to note that most of the graphene properties are sensitive to structural defects and the number of layers [4,29–31]. Thus in order to exploit most of the proposed applications, the synthesis routes and conditions is important in tuning the structure and properties of graphene. There are a number of reviews on graphene related materials for possible applications in relation to optical, electronic, photocatalytic and electrochemical properties [1–50]. However, there is a lack of consideration on their important molecular interactions, e.g. for molecular adsorption and storage. This article attempts to address such issues.

Several large scale processing methods have been involved for different graphene based materials through either graphitic top-down or molecular carbon precursor bottom-up approaches, as summarised in Fig. 1. The flexibility in modification and functionalization of the graphene surface has opened up many possibilities for the development of tailored functional materials. For example, surface modification has been applied to tune the band-gap of single-layer graphene for microelectronic devices. Similarly, the intrinsic non-porous 2D graphene is tuned to highly porous 3D architectures for electrochemical devices (batteries, fuel-cells and supercapacitors) and gas sorption, storage, separation and sensing.

On a relatively large scale, graphene is mostly obtained from graphite precursors through oxidation–exfoliation–reduction, i.e. in the form of graphene oxide (GO) as schematically shown in Fig. 1a [4,33,49–52]. The GO structure contains abundant oxygen-rich functional groups; hydroxide and epoxide groups on the basal plane and carbonyl and carboxyl groups on edges of the graphene

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