



Review

Graphene-based materials: Fabrication and application for adsorption in analytical chemistry[☆]Xin Wang^a, Bo Liu^a, Qipeng Lu^c, Qishu Qu^{b,*}^a Department of Chemistry, School of Science, Beijing JiaoTong University, Beijing 100044, China^b School of Materials and Chemical Engineering, Anhui Jianzhu University, Hefei 230601, China^c Institute of Optoelectronic Technology, Beijing JiaoTong University, Beijing 100044, China

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ABSTRACT

Graphene, a single layer of carbon atoms densely packed into a honeycomb crystal lattice with unique electronic, chemical, and mechanical properties, is the 2D allotrope of carbon. Owing to the remarkable properties, graphene and graphene-based materials are likely to find potential applications as a sorbent in analytical chemistry. The current review focuses predominantly on the recent development of graphene-based materials and demonstrates their enhanced performance in adsorption of organic compounds, metal ions, and solid phase extraction as well as in separation science since mostly 2012.

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1. Introduction

Trace analysis is always a subject of great interest especially in the field of analytical chemistry [1,2]. However, because of the strong matrix interferences, to determine the low concentrations of analytes directly from samples is very difficult. Therefore, it is necessary to increase the concentration of analytes and decrease the

background interference by enriching trace analytes before using preliminary separation techniques [3–6]. However, for the preconcentration technique, the high recovery and enrichment factor only can be achieved by using a suitable solid sorbent. Nowadays, with the advancements in nanomaterials synthesis, several complementary strategies have been developed for new analytical procedures including carbon-based materials, inorganic particles, and organic-inorganic composites [7–10]. Among different types of materials, carbon nanomaterials are the research focus due to its chemical stability, durability, corrosion resistance and large surface area [11,12].

Carbon nanomaterials, which could be used as sorbent materials for preconcentration, comprise a wide range of allotropic forms

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of carbon, including nanodiamond, fullerenes, carbon nanotubes, graphite nanofibers, and graphene [7]. These nano-sized carbon materials could yield high preconcentration factor and good selectivity result from the large surface area. Since graphene was first described in the scientific literature in 2004 by Novoselov et al., the graphene-based materials have been studied with a view to developing several applications [13]. In the view of structure, graphene possesses one or a few layers thickness of sp²-hybridized carbon atoms arranged in a honeycomb pattern [14], which cause graphene with various superior electrical, electrochemical, optical and mechanical properties [15–18]. Therefore, versatile methods have been developed for fabrication, growth or synthesis of graphene and its derivatives [19–27]. RGO and GO have found potential applications like sensors [28–44], energy storage [45–51], solar cells [52], electrochemical devices, ultracapacitors [53–57], and so forth. In particular, very large specific surface area (theoretical value 2630 m²/g) [58] and its electron-rich double-sided polycyclic aromatic scaffold makes it a promising analytical candidate as an wonderful adsorbent [59–65]. Recently, a series of new sorbents based on graphene (RGO) and graphene oxide (GO) represent a promising analytical technology with many advantages, including large specific surface area, high adsorption capacity and strong affinity.

The most popular technique used for synthesizing graphene is chemical reduction of graphene oxide as shown in Fig. 1. GO was obtained by oxidation graphite with various oxidants in acidic media. The Hummer method of synthesizing GO was usually used and considered as one of the most efficient methods [66,67]. Subsequently GO can be chemically reduced to RGO by various inorganic and organic reducing agents (Fig. 1) [68]. Due to their huge surface area, spectacular physical and chemical properties, RGO and GO have attracted great interest in analytical chemistry.

The present review attempts to summarize current progress in the preparation of graphene-based materials for the adsorption of various kinds of compounds, including organic compounds and metal ions. Also, the applications of graphene-based materials in solid-phase extraction and chromatography separation were introduced briefly.

2. Adsorption of organic compounds

Graphene-based materials have been applied to adsorb various organic pollution via non-covalent interaction including Van der Waals type interactions, electrostatic interaction, hydrogen bonds, π - π stacking, dispersion forces, dative bonds and the hydrophobic effect [69]. Table 1 lists recent reports on the adsorption of organic compounds by graphene-based materials.

The advantage of the RGO is selective adsorption. Various adsorption mechanisms play predominate roles in different kinds of organic chemical systems. Because of the large localized π -electron systems, the RGO could adsorb the aromatic rings from several organic compounds through strong π - π interaction. Different from the RGO, for GO, the rich functional groups play a key role in forming hydrogen bonding or electrostatic interaction with organic compounds containing oxygen- and nitrogen-functional groups.

Xu et al. used RGO for the decontamination of bisphenol A (BPA) and the maximum adsorption capacity of RGO for BPA was 182 mg/g at 302.5 k [70]. The mechanism was schematically represented in Fig. 2. In this case, the π - π interactions between the benzene rings of BPA and RGO as well as the hydrogen bonds between the oxygen containing groups contained in BPA and RGO might be responsible for the adsorption of BPA on RGO.

Pei et al. studied the adsorption of 1,2,4-trichlorobenzene (TCB), 2,4,6-trichlorophenol (TCP), 2-naphthol, and naphthalene (NAPH)

based on RGO and GO [71]. The structure of TCP and NAPH are similar with TCB and 2-naphthol. And the differences between them are TCP and 2-naphthol containing the polar hydroxyl group. From the experimental results, RGO had similar adsorption capacity for four aromatics at pH 5.0 in despite of their different chemical properties, which indicated that four aromatics were adsorbed on RGO mainly via π - π interaction than hydrophobic interaction. In the case of GO, the order of adsorption affinity for four aromatics was NAPH < TCB < TCP < 2-naphthol. The higher adsorption of TCP and 2-naphthol on GO could be attributed to H-bonding between hydroxyl groups of TCP or 2-naphthol and the oxygen-containing functional groups on GO.

Maliyekka et al. studied the adsorption of chlorpyrifos (CP), endosulfan (ES), and malathion (ML) onto GO and RGO from water [72]. They determined the energies of adsorptive interactions between graphene and a pesticide (binary complex) as well as the interaction among graphene, pesticide and water complexes by the first-principles pseudopotential-based density functional theory (DFT) analysis. The results showed that both graphene and pesticide have an attractive interaction with water molecules and formed the Graphene–Water–Pesticide (G–W–P) complexes through electrostatic interaction. The adsorption of pesticide on graphene was feasible with the existence of water molecule, and the adsorption of pesticide on dry graphene was expected to be rather weak or unlikely. By using this water-assisted adsorption, the adsorption capacity of graphene observed up to 1200 mg/g which was much higher than any other materials.

Wu et al. investigated the removal of hazardous chemicals from wastewater by using RGO as adsorbent [73]. It was found that the adsorption capacity and adsorption speed increased with the increasing of molecular size and number of the benzene rings possessed by these organic chemicals. The maximum adsorption capacity for p-toluenesulfonic acid, 1-naphthalenesulfonic acid, and methyl blue could reach to 1.43 g/g, 1.46 g/g, and 1.52 g/g, respectively.

Although graphene-based materials have strong ability to adsorb the different kinds of compounds, it suffers the problem of difficult to be separated from the mother solution. Therefore, the introduction of magnetic properties into RGO and GO systems is an excellent strategy to solve the above problem. By combining the high adsorption capacity from graphene and the separation convenience from magnetic materials, the adsorbents can be easily separated and recycled by a simple magnetic process. With the advancements in nanomaterial's synthesis, various kinds of magnetic nanoparticles (Ni, CoFe₂O₄, Fe₃O₄) combined with functional GO and RGO were used successfully as adsorbents for removing dyes and organic contaminant.

Geng et al. synthesized RGO/Fe₃O₄ nanocomposites with tunable RGO/Fe₃O₄ ratio and achieved an exceptionally high yield [74]. The RGO/Fe₃O₄ hybrid was used for the decontamination of a serious dyes, such as rhodamine 6G (R6G), acid blue 92 (AB92), orange (II) (OII), malachite green (MG), and new cocaine (NC). Because of the large surface area of RGO and magnetic property of Fe₃O₄, the hybrid possessed quite a good adsorption capacity for these dyes. Moreover, the hybrid could be easily and rapidly extracted from water by magnetic attraction. Compared with the single dye adsorption, the hybrid could work well even exposed to a multi dye cocktail without suppressing the adsorption capacity for each of the dyes. This hybrid adsorbent could be easily and efficiently regenerated by simply annealing for reuse with hardly any compromise of the adsorption capacity.

Zhang et al. decorated RGO with the superparamagnetic Fe₃O₄ and used the RGO/Fe₃O₄ composites as adsorbent for BPA [75]. The strong π - π interaction is the main drive force for the adsorption. Due to the superparamagnetism, RGO/Fe₃O₄ could be easily

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