# Research on the Preparation and Removal of Magnetic Graphene Materials in Organic Contamination

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## Abstract

**Graphene is a two-dimensional nano sheet composed of carbon atoms hybridized by sp2. It has excellent physical and chemical properties, super large specific surface area and easy surface modification characteristics. It has a huge application prospect in the environmental field. It is a super strong adsorption material, which can achieve efficient enrichment of organic pollutants and heavy metals in water. However, graphene nano sheets are prone to exposure of interaction stacking influence sites. Although graphene oxide has good dispersion, it is not easy to recover, and it is very easy to migrate and transform in the environment, posing a potential threat to the ecological environment. Due to the limitations of the environmental application of graphene, graphene and magnetic nanoparticles are combined and assembled into magnetic graphene, in order to not only preserve the original outstanding physical and chemical properties of graphene, but also solve the problems of easy stacking, difficult separation and difficult recovery.**

## Keywords

**Magnetic Material; Graphene; Organic Contamination; $Fe_3O_4$ Nanoparticles.**

## 1. Introduction

Graphene is a two-dimensional nano material with single-layer honeycomb lattice structure composed of carbon atoms in sp2 hybrid way. Since it was discovered in 2004, it has attracted extensive attention and research in the scientific community. Graphene has many excellent physical and chemical properties, such as great theoretical specific surface area, strong mechanical properties, excellent conductivity and thermal conductivity. It has a very broad potential application prospect in many fields such as photoelectric, energy storage, mechanical manufacturing, biomedicine and environment.

Graphene materials have excellent adsorption performance for environmental pollutants. However, due to π-π interaction and hydrophobicity between graphene layers, stacking is very easy to occur, which will suppress the exposure of adsorption sites[1]; although the dispersion of GO is good, it is not easy to separate and recycle GO, and it is easy to migrate and spread in the environment, leading to potential environmental risks[2]. In the field of environmental remediation, $Fe_3O_4$ nanoparticles are often used as adsorbent to treat heavy metal pollutants in the environment due to their easy solid-liquid separation and low cost[3]. However, because it is easy to agglomerate, its performance will be affected, and the separate $Fe_3O_4$ nanoparticles have limited adsorption capacity for pollutants, so they need to be modified to improve their dispersion and adsorption performance. Combining $Fe_3O_4$ nanoparticles with graphene material with excellent adsorption performance, the composite material has the advantages of high adsorption performance and easy separation. As a new adsorbent, magnetic graphene is widely used in the environmental field.

## 2. Preparation of Magnetic Graphene

There are many preparations of magnetic graphene composites, including hydrothermal/ solvothermal method, chemical coprecipitation method, cross-linking method (covalent bond bonding method), high-temperature decomposition method, microwave radiation heating method[4]. In addition to preparing single magnetic graphene materials, they can also be functionalized by adding ionic surfactants, polymers, carbon nanotubes, ionic liquids/eutectic solvents, metal organic frameworks, borate affinity materials to improve their dispersion and adsorption properties[5]. In addition, 3D magnetic graphene composites can also be prepared, which can not only have various excellent properties of 2D magnetic graphene, but also have a porous structure[6]. This research mainly introduces the preparation of magnetic graphene by hydrothermal/solvothermal method, chemical coprecipitation method, electrostatic self-assembly and cross-linking method.

### 2.1. Water/Solvothermal Method

The agglomeration between magnetic graphene particles prepared by water/solvothermal method is significantly reduced, the crystal size of the product is small, and the crystallinity is high. Although the water/solvothermal process is usually carried out under high pressure and high temperature, this method is an environmentally friendly and economically feasible method. The water/solvent heat rule of graphene based iron oxide magnetic composites is to add graphene materials to the solution at the same time and transfer them to the high-pressure reactor.

### 2.2. Coprecipitation Method

Chemical coprecipitation is the simplest and most efficient method for the synthesis of magnetic graphene. Iron salts are deposited on the surface of water-soluble graphene oxide to achieve $Fe_3O_4$ loading on graphene materials. The disadvantage is that the precipitation is uneven, which affects the material performance. Deng et al. added $Fe^{2+}$ and $Fe^{3+}$ in a certain proportion to GO solution at the same time, which was heated and stirred in N2 atmosphere, and then adjusted the pH to about 10 by using ammonia, finally at 90℃ for 2h of reaction to obtain magnetic graphene oxide (M-GO).

### 2.3. Electrostatic Self-assembly

The electrostatic self-assembly method is to use the electrostatic interaction between GO and Fe3O4 to package and self-assemble $Fe_3O_4$/GO by adjusting the pH value in a suitable range. Han et al added $Fe_3O_4$ solution to GO dispersion, and $Fe_3O_4$/GO was synthesized by self-assembly using electrostatic interaction between them. Huang[7] used 1 mol L-1 HCl to adjust the pH of $Fe_3O_4$ suspension to about 3~4, so as to ensure that the surface charge of its nanoparticles is positive and no iron ions are precipitated. Next they heated it to 80℃ in a water bath in a high-purity $N_2$ atmosphere and stirred it vigorously, and then added GO dispersion drop by drop. Due to the electrostatic interaction between GO with negative electricity on the surface and $Fe_3O_4$ with positive electricity on the surface, the GO lamella will soon wrap $Fe_3O_4$ nanoparticles for self-assembly, heated them in a water bath at 80℃ under $N_2$ atmosphere and stirred vigorously for 3 hours to obtain MGO with good dispersion, uniform magnetic particles and small size (10~20 nm).

### 2.4. Crosslinking Method

In the process of composite preparation, the reaction between different functional groups of polymer can be used to combine the two monomers. Through this organic covalent modification, more stable magnetic graphene composites can be prepared.

## 3. Adsorption and Removal of Organic Pollutants by Magnetic Graphene

Magnetic graphene can be used as a new adsorption material to adsorb and remove organic pollutants such as dyes, polycyclic aromatic hydrocarbons (PAHs) and their derivatives, antibiotics, etc. in water bodies[8-10] due to its excellent adsorption performance and easy separation characteristics. It has very excellent effects, and can be magnetic recovered and reused after the adsorption and enrichment of pollutants. The desorption of pollutants adsorbed on magnetic graphene will be affected by the nature of the pollutants themselves and the adsorbent, which may lead to desorption lag, migration and transformation in the environment, thus causing potential environmental risks.

In addition, the surface structure of magnetic graphene is usually heterogeneous, and the structure and properties of magnetic graphene prepared with different materials and methods are quite different. The study on the adsorption behavior and interaction mechanism of different types of organic pollutants on magnetic graphene and the exploration of its structure-activity relationship can provide a theoretical basis for the development of new magnetic graphene functional materials with potential for pollution remediation in the actual environment, the prediction of its environmental behavior and the assessment of environmental risks.

In the natural environment, multiple pollutants often coexist [11]. Organic and inorganic pollutants often coexist. Magnetic graphene can not only adsorb organic pollutants, but also has certain adsorption performance for inorganic pollutants. The coexisting inorganic pollutants in the environment may affect the adsorption of magnetic graphene on organic pollutants[12].

To sum up, the interactions between magnetic graphene and organic pollutants mainly include: π-π interaction; hydrophobic action; electrostatic force action; hydrogen bonding; the interaction of n-π EDA is similar to that of graphene on organic pollution[13]. For non-polar organic pollutants, the sp2 hybrid plane on magnetic graphene oxide can still interact with it through π-π interaction and hydrophobic interaction [14], and sp3 hybrid plane can still interact through charge transfer. For polar organic pollutants, interaction can be realized through electrostatic interaction, hydrogen bond and cation-π bond.

## 4. Conclusion

Magnetic graphene, as a new type of environmental functional nanomaterial, has excellent advantages such as adsorption properties, easy recovery and recycling. Its synthesis, modification and application have attracted extensive attention and research in the environmental field. Magnetic graphene has high adsorption performance for many organic pollutants, and its interface interaction sites are closely related to its structure. However, the interface interaction mechanism and structure activity relationship of magnetic graphene on different types of organic pollutants need to be further studied on the structure and property regulation of magnetic graphene.

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