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Structural Stability Regulation and Recycling Strategies of Graphene and its Derivatives in Water Treatment: a Review

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ABSTRACT
This article provides a systematic review of recent research on graphene-based materials in water treatment, focusing on strategies for controlling structural stability and regeneration methods. To enhance stability, surface functionalization (such as covalent modification and non-covalent coating), composite material construction (such as with polymers and metal oxides), and the design of three-dimensional cross-linked structures can significantly improve the mechanical strength and chemical resistance of the materials. In terms of regeneration strategies, methods such as chemical leaching, thermal treatment, and electrochemical regeneration have been extensively studied, but it is essential to balance the efficiency of regeneration with the preservation of the material's structural integrity. Additionally, the article analyzes the main obstacles to current industrial applications, including batch stability in large-scale production, the optimization of continuous processing equipment, and the development of anti-bacterial coatings.

KEYWORDS
Graphene; Water treatment; Structural stability; Regeneration strategy; Graphene oxide; Reduced graphene oxide

1 Introduction

With the rapid advancement of industrialization and urbanization, global water resources are facing increasingly severe pollution issues. The significant release of heavy metal ions, organic pollutants, and pathogenic microorganisms has seriously threatened the ecological environment and human health. Developing efficient and sustainable water treatment technologies has become a key focus in current environmental science research. Among various emerging materials, graphene and its derivatives (such as graphene oxide and reduced graphene oxide) have shown great potential in water pollution control due to their large specific surface area, rich functional groups, and excellent adsorption and catalytic properties. Despite significant progress in the application of graphene-based materials in water treatment, issues such as decreased structural stability and insufficient recycling capabilities still limit their practical engineering applications and large-scale promotion. Materials tend to aggregate, structurally degrade, or lose activity during multiple uses, leading to reduced treatment efficiency and even secondary pollution. Therefore, systematic research on the structural stability regulation and recycling strategies of graphene and its derivatives in water treatment is of significant theoretical and practical value. This article aims to review recent research advancements, analyze existing key challenges, and explore possible improvement directions, providing references and insights for the continuous development of this field.

2 Overview of graphene and its derivatives

2.1 Graphene

Graphene is a two-dimensional honeycomb lattice structure composed of a single layer of carbon atoms, featuring an exceptionally high mechanical strength (130 GPa) and a theoretical specific surface area (2630 m^2/g). Its unique sp^2 hybridized electronic structure endows it with outstanding electrical conductivity (carrier mobility 200,000 $cm^2/V{\cdot}s$) and thermal conductivity (5000 W/m·K). In the field of membrane separation, graphene's atomic thickness (0.34 nm) and controllable pore structure can achieve ultra-high permeability and selectivity, especially suitable for gas separation and water treatment [1].

2.2 Graphene oxide (GO)

GO, a derivative of graphene modified by oxidation, is rich in oxygen-containing functional groups such as hydroxyl, epoxy, and carboxyl groups on its surface. These groups significantly enhance GO's hydrophilicity and dispersibility, allowing it to form stable colloids in water. However, they partially disrupt the sp^2 conjugated network, leading to a decrease in conductivity. The presence of these oxygen-containing groups endows GO with strong chelating capabilities for heavy metal ions like Pb^{2+} and Cd^{2+} [2].Moreover, the permeability and selectivity of GO membranes can be adjusted by controlling the degree of oxidation.

2.3 Reduced graphene oxide (rGO)

rGO can be prepared from GO through chemical reduction, thermal reduction, or photo-reduction, partially restoring graphene's conductivity and hydrophobicity. However, residual oxygen-containing groups and structural defects can affect its stability: high-temperature reduction, while enhancing crystallinity, may lead to interlayer aggregation; mild reduction, on the other hand, retains more functional groups, enhancing adsorption of specific pollutants. In membrane applications, rGO is often cross-linked to improve mechanical strength, and its hydrophobic surface can optimize oil-water separation efficiency [3].

3 Application status of graphene materials in water treatment

3.1 Adsorption of heavy metal ions

Graphene and its derivatives, due to their high specific surface area, abundant oxygen-containing functional groups, and excellent hydrophilicity, have become ideal materials for the adsorption of heavy metal ions. Research has shown that graphene oxide exhibits superior adsorption performance for copper, zinc, nickel, chromium, cobalt, and other heavy metal ions. The adsorption mechanism primarily involves the coordination between the surface oxygen-containing groups (such as carboxyl and hydroxyl groups) and the metal ions [4].The selective adsorption capacity for specific metals can be further improved by functional modification (such as the introduction of amino groups and thiol groups) [5].The macroscopic structure of 3D graphene shows significant advantages in practical applications due to its easy recycling characteristics.

3.2 Removal of organic pollutants

Graphene-based materials primarily adsorb organic pollutants, including dyes, drug residues, and phenols, through π-π stacking and hydrophobic interactions. The layered structure of graphene oxide can effectively intercept drug pollutants such as ibuprofen and ciprofloxacin [6]. For complex pollution systems, graphene-based photocatalysts can achieve the synergistic removal of heavy metal-organic pollutants (HM/OP) by generating electron-hole pairs [7]. Research indicates that when graphene is combined with semiconductor materials, it can extend the light-responsive range to the visible light region, significantly enhancing photocatalytic efficiency.

3.3 Membrane separation technology

Graphene-based separation membranes, with their controllable two-dimensional nanochannels (0.3-0.8 nm) and surface charge characteristics, can achieve precise screening of heavy metal ions [8]. By adjusting the interlayer spacing and surface charge density, both water flux and ion retention rates can be enhanced simultaneously. Graphene oxide membranes exhibit excellent performance in dye molecule retention for nanofiltration applications. The newly developed graphene-polymer composite membranes, through functional modification, have achieved efficient and rapid removal of pollutants [9]. However, long-term stability and large-scale production remain significant challenges in practical applications.

3.4 Catalytic degradation and advanced oxidation process

The graphene-based catalytic membrane reactor integrates membrane separation with chemical catalytic degradation, significantly enhancing the efficiency of pollutant removal [10]. The metal oxide/graphene composite catalyst can activate peroxymonosulfate to produce sulfate radicals, effectively degrading organic pollutants [11]. The nitrogen-doped graphene and carbon nanotube composite catalytic system exhibits excellent catalytic activity in the activation of peroxymonosulfate. Moreover, the combination of graphene and TiO_2 can broaden the visible light response range of photocatalysts, thereby improving the degradation efficiency of pollutants such as phenol.

4 Structural stability and control strategy

One of the primary challenges in the application of graphene and its derivatives in water treatment is the issue of structural stability. In complex solution environments, such as strong acids, strong bases, or high salt conditions, or during prolonged operation, graphene materials are prone to structural degradation, leading to performance decline or even failure. Therefore, it is essential to gain a deep understanding of the degradation mechanisms and to develop effective control strategies.

4.1 Common structural degradation mechanisms

The stability of graphene materials is mainly manifested by interlayer peeling, agglomeration and loss of functional groups. Interlayer peeling is usually caused by electrostatic repulsion or solvent intercalation, especially in graphene oxide, the hydrophilicity of oxygen-containing functional groups (such as carboxyl and hydroxyl) will exacerbate this phenomenon. Recombination occurs more frequently in hydrophobic reduced graphene oxide, where the nanosheet layers are prone to irreversible stacking due to π-π interactions and van der Waals forces, reducing the specific surface area and active site [12]. In addition, under extreme pH or high temperature conditions, the functional groups on the surface of graphene may decompose or reduce, further weakening its adsorption or catalytic performance.

4.2 Surface functionalization and chemical modification

The structural stability of graphene can be significantly improved by covalent or non-covalent modification. Covalent modifications, such as amidation and silanization, can introduce stable chemical bonds, thereby reducing the loss of functional groups (for example, by cross-linking graphene oxide sheets with ethylenediamine). In terms of non-covalent modifications, branched polyethyleneimine (BPEI) and polyacrylic acid (PAA) can stabilize the dispersion of reduced graphene oxide (rGO) through electrostatic interactions [13]. Additionally, sulfonic acid group modifications can enhance the material's resistance to acidic environments.

4.3 Solid loading and support body construction

Membrane carrier immobilization is a common strategy. GO membranes can selectively remove pollutants through interlayer nanochannels and oxygen functional groups, but the swelling problem in long-term use needs to be solved. The novel graphene-coated sand (GCS) filter medium shows stable performance in sewage treatment by regulating its surface morphology [14].

In conclusion, the structural stability of graphene materials can be significantly improved by multi-scale regulation, which lays a foundation for its long-term application in water treatment.

5 Regeneration and recycling strategies

5.1 Regeneration method of adsorbent

For the regeneration of adsorbents, magnetite/reduced graphene oxide (FeO/RGO) nanocomposites have achieved efficient regeneration through chemical leaching and magnetic separation. However, their cyclic stability is still affected by a decline in adsorption capacity with each regeneration cycle. The heat treatment method has shown advantages in the regeneration of graphene-based photocatalysts. Ag/TiO/graphene nanocomposites not only maintain their microstructural stability under visible light irradiation but also exhibit excellent cyclic stability, attributed to the synergistic effect of graphene's adsorption properties and electron-hole pair separation efficiency [15].

5.2 Regeneration performance and optimization of functional membrane

Graphene oxide (GO) membranes, due to their reversible physicochemical properties and nano-channel structure, can restore flux through alternating acid and alkali washing. However, studies have highlighted significant stability issues with GO-coated membranes, necessitating cross-linking modification or three-dimensional structural design to enhance interlayer bonding [16]. Performance evaluations indicate that after 10 cycles, the graphene-coated sand (GCS) filter medium retains over 85% of its initial efficiency in removing emerging pollutants, thanks to the stable integration of the graphene coating with the sand particle surface morphology [14].

5.3 Influence of regeneration on material structure

Chemical regeneration may lead to partial loss of oxygen functional groups on the surface of graphene derivatives, while thermal regeneration may cause the accumulation of carbon skeleton defects. It is worth noting that functionalized reduced graphene oxide (frGO) can maintain more than 90% heavy metal adsorption capacity after multiple regeneration by precisely controlling the number of residual oxygen-containing groups, which confirms the key role of structural stability regulation on cycling performance [5].

6 Current challenges and research trends

The primary challenge for graphene-based water treatment materials is the coordinated regulation of structural stability and adsorption capacity. Research indicates that while graphene oxide (GO) boasts a high specific surface area (2630 m^2/g) and a rich array of oxygen-containing functional groups, its structure tends to degrade in acidic or alkaline environments, leading to a decline in adsorption performance. For instance, magnetic graphene oxide composites can maintain an Ho(III) adsorption capacity of 72.1 mg/g after five regeneration cycles, but prolonged exposure to extreme pH conditions can cause the layered structure to collapse [17]. In practical applications, the complex components of industrial wastewater, such as competitive ions and organic matter, can occupy the active sites of graphene, resulting in a 40-60% decrease in adsorption capacity when treating saline wastewater [14]. Multifunctional composite materials can partially address this issue through synergistic effects, but their large-scale production is still constrained by the uniform dispersion of graphene and the stability of the carrier.

The key technical barriers in the process of industrialization include: (1) Mass transfer optimization of continuous processing devices; (2) Durability of anti-bacterial coating; (3) Batch stability control of ton-level preparation.

The future development direction will be to build an intelligent composite material system with self-monitoring function, and realize the real-time regulation of adsorption-degradation synergistic effect and regeneration process.

7 Conclusion and Prospect

This article provides a systematic review of the research progress on graphene and its derivatives in water treatment, focusing on their structural stability and recycling strategies. It also explores future development directions. Graphene-based materials, owing to their unique physical and chemical properties, have demonstrated outstanding performance in areas such as heavy metal adsorption, organic pollutant removal, membrane separation, and catalytic degradation.

However, there are still key challenges in practical applications:

(1) Insufficient structural stability: In complex water environment, graphene materials are prone to agglomeration, interlayer peeling or loss of functional groups, resulting in decreased adsorption capacity and catalytic activity.

(2) Regeneration efficiency and cost: Although the existing regeneration methods can partially restore the material properties, the adsorption capacity still decreases significantly after multiple cycles.

(3) Influence of actual water body complexity: Competitive ions and organic matter in industrial wastewater will occupy active sites, resulting in a decrease of 40%-60% in the adsorption efficiency of materials in salt-containing wastewater.

(4) Obstacles to large-scale application: including mass transfer optimization of continuous processing devices, durability of anti-bacterial coating, and batch stability control of ton-level preparation.

In view of the above problems, this paper proposes the following future research directions:

(1) Green synthesis and low-cost preparation: Develop environmentally friendly graphene-derived methods to reduce the use of toxic reagents and reduce the cost of large-scale production.

(2) Design of intelligent composite materials: Construct graphene-based materials with self-monitoring function, regulate the adsorption-degradation process in real time, and optimize the regeneration strategy.

(3) Multi-technology collaborative application: Combine membrane separation, catalytic oxidation and adsorption technology to develop modular water treatment system and improve the comprehensive treatment efficiency.

(4) In-depth study of mechanism: in situ characterization technology is used to reveal the structural evolution law of materials in the regeneration process, and guide the optimization of stability.

In summary, the further development of graphene-based water treatment materials requires interdisciplinary collaboration, integrating technologies such as materials science, environmental engineering, and artificial intelligence to facilitate their transition from laboratory research to practical engineering applications. By addressing issues of stability and regeneration, graphene materials are poised to become the core of the next generation of efficient and sustainable water treatment technologies.

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