

Graphene Oxide–Based Materials for Heavy Metal Ion Adsorption: A Comprehensive Review

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Abstract: Accelerating global industrialization and urbanization, coupled with the proliferation of emerging technologies, have rendered heavy metal contamination an environmental and public health crisis of growing international concern. The recalcitrance of heavy metals to degradation and their propensity for bioaccumulation pose severe threats to ecological integrity and human health. Conventional remediation strategies—principally chemical precipitation, ion exchange, and membrane separation—are inherently constrained by prohibitive costs, limited removal efficiencies, and poor regenerability, rendering them inadequate for the treatment of increasingly complex heavy metal-laden wastewaters. Graphene oxide (GO), distinguished by its exceptionally high specific surface area, abundant oxygen-containing functional groups, outstanding adsorption capacity, and versatile chemical modifiability, has emerged as a promising platform for overcoming these limitations and has attracted intensive research interest as a next-generation heavy metal adsorbent. Drawing upon high-impact literature from Web of Science and other mainstream databases, this review employs bibliometric and systematic review methodologies—screening publications by date, impact factor, and data integrity—to critically examine the research progress of GO and its composites in heavy metal remediation. The enhancing effects of key modification strategies on adsorption performance are evaluated, including magnetic functionalization, amino functionalization, chitosan hybridization, and membrane fabrication. Furthermore, persistent technical bottlenecks are identified, including insufficient selectivity, poor adaptability to complex aqueous matrices, and limited scalability. The review concludes with a forward-looking discussion on precision recognition, green and sustainable material

design, and other promising research directions.

Keywords: Graphene Oxide; Heavy Metal Adsorption; Functionalized Composites; Magnetic Separation; Membrane Filtration; Water Remediation

1. Introduction

Since the Second Industrial Revolution, the large-scale extraction of coal and the rapid expansion of non-ferrous metal smelting, electroplating, and chemical manufacturing have resulted in the uncontrolled discharge of untreated heavy metal-bearing wastewater from mines and industrial facilities. Consequently, concentrations of lead (Pb), cadmium (Cd), mercury (Hg), arsenic (As), and chromium (Cr) in aquatic environments have risen precipitously. Owing to their non-biodegradability and capacity for bioaccumulation, heavy metals inflict persistent and cumulative damage on ecosystems. Moreover, these contaminants bioaccumulate through dietary intake and inhalation, ultimately compromising the nervous system, hepatic and renal function, and immune responses [1].

Current approaches to heavy metal wastewater treatment rely predominantly on chemical precipitation, ion exchange, and membrane separation [2,3,4]. Among these, adsorption has garnered considerable attention by virtue of its operational simplicity, cost-effectiveness, minimal secondary pollution, and broad applicability [5]. The development of high-performance adsorbent materials has accordingly become a critical frontier in environmental remediation research.

Since its landmark isolation by Geim and Novoselov in 2004—building upon theoretical foundations laid by Wallace in 1947—graphene has attracted widespread interest owing to its remarkable mechanical and chemical properties. As an oxidized derivative of graphene, graphene oxide (GO) bears abundant oxygen-containing

functional groups, predominantly hydroxyl moieties, on its basal plane and edges. This functionalization not only confers excellent hydrophilicity but also enables strong interactions with heavy metal ions through surface complexation, ion exchange, electrostatic attraction, and physisorption, thereby endowing GO with formidable adsorption capabilities. Zhao et al. (2011) provided seminal evidence that few-layered graphene nanosheets exhibit exceptionally high adsorption capacities for heavy metal ions, establishing a foundational benchmark for subsequent investigations [6]. In recent years, researchers have pursued functional modification of GO through the introduction of magnetic nanoparticles, amino groups, and metal oxides, substantially enhancing its adsorption capacity, selectivity, and recyclability [7,8,9,10]. Concurrently, the field has increasingly embraced the integration of theoretical computation with experimental validation; density functional theory (DFT) and molecular dynamics (MD) simulations are now widely employed to elucidate the interaction mechanisms between heavy metal ions and surface functional groups at the atomic level [11].

Notwithstanding the superior adsorption performance demonstrated by GO-based materials under idealized laboratory conditions, their deployment in complex real-world environments remains fraught with formidable challenges. A systematic review of GO-based adsorbents for heavy metal ion removal—one that critically assesses current limitations and projects future trajectories—is therefore of considerable theoretical significance and practical relevance.

2. Structural Characteristics and Adsorption Advantages of GO-Based Materials

2.1 Structure and Functional Groups of Graphene Oxide

Graphene oxide is synthesized via the vigorous oxidation of graphite, yielding a two-dimensional layered architecture comprising hydrophobic sp^2 -hybridized carbon domains interspersed with sp^3 -hybridized carbon regions bearing oxygen-containing functional groups. This structural motif preserves the graphene backbone while introducing a high density of oxygen functionalities. Epoxy and hydroxyl groups distributed across the basal plane, together with carboxyl groups located at the sheet edges, collectively confer pronounced hydrophilicity

upon GO and furnish lone-pair electrons capable of coordinating with heavy metal ions to form stable complexes. As Ahmad et al. have emphasized, these functional groups constitute the chemical core of GO's adsorptive function; their speciation, surface density, and protonation state directly govern adsorption behavior, and the reactive sites they present are amenable to further chemical modification through both covalent grafting and physical adsorption [12].

2.2 Advantages of GO for Heavy Metal Adsorption

Relative to conventional adsorbent materials, GO offers several distinct advantages.

Exceptional adsorption capacity. Zhao et al. demonstrated that few-layered graphene exhibits adsorption capacities for heavy metal ions such as Pb(II) that substantially exceed those of activated carbon and natural mineral adsorbents [6].

Abundant and tunable surface chemistry. As discussed above, the profusion of oxygen-containing functional groups on the GO surface permits targeted chemical modification to markedly augment adsorption performance.

Outstanding structural modifiability. The unique two-dimensional morphology of GO, combined with its rich surface functionality, affords remarkable versatility in material design. Grafting strategies involving amino groups, chitosan, magnetic nanoparticles, and other moieties have been successfully employed to enhance selectivity and facilitate recovery [7,8].

Robust mechanical properties and chemical stability. GO maintains structural integrity across a wide range of environmental conditions—from acidic to alkaline media—thereby sustaining adsorption functionality under diverse operational parameters [13].

In summary, pristine GO exhibits considerable adsorption potential by virtue of its chemically modifiable surface. Both Chen et al. and Wu et al. have documented the superior adsorption performance of GO toward common heavy metal ions; however, the intrinsic difficulty of rapidly separating and recovering GO from aqueous suspensions remains a critical limitation that constrains its practical utility [5,8]. Accordingly, the development of GO-based composite materials has emerged as an imperative research direction.

3. Research Progress on GO-Based Composite Materials

3.1 Magnetic GO Composites

Magnetic GO composites are fabricated by anchoring magnetic nanoparticles—predominantly Fe_3O_4 —onto GO nanosheets. This design paradigm retains the high adsorption capacity intrinsic to GO while enabling rapid separation and recovery via an external magnetic field, thereby addressing the longstanding challenge of GO recovery from aqueous media [8,9,14].

Guo et al. [14] synthesized three-dimensional Fe_3O_4 –graphene macroscopic composites for the removal of arsenic species from aqueous solution. Their results demonstrated favorable adsorption capacity coupled with excellent structural stability, effectively overcoming the separation difficulties that plague conventional adsorbents. In a complementary investigation, Wu et al. [8] revealed that magnetic GO composites are capable of simultaneously adsorbing heavy metal ions such as Pb(II) and co-existing organic pollutants. Notably, a synergistic relationship was observed between the two adsorption processes, suggesting that the concurrent removal of multiple contaminant classes by a single material may open new avenues for integrated wastewater treatment. To achieve selective adsorption of specific heavy metal ions, Kumar et al. prepared amino-functionalized magnetic graphene oxide (AMGO) and evaluated its performance for the selective capture of Pb(II) , Hg(II) , and Cd(II) from aqueous solution [10]. The grafted amino groups significantly enhanced the material's affinity toward all three target ions; remarkably, superior selectivity was maintained even when the concentrations of competing background ions substantially exceeded those of the target species. Although magnetic GO composites have demonstrated the capacity for rapid magnetic separation and regeneration—with negligible decline in adsorption capacity over multiple cycles—the long-term stability of the magnetic component remains a concern. Detachment and oxidation of magnetic nanoparticles during prolonged use may progressively degrade adsorption performance, rendering the enhancement of interfacial bonding between the magnetic phase and the GO matrix a priority for future research.

3.2 GO-Based Membrane Materials

Membrane separation constitutes a cornerstone technology in modern water treatment, offering advantages such as high automation relative to

conventional batch adsorption processes. Drawing inspiration from mussel adhesive proteins, Zhang et al. developed a polydopamine-reinforced GO/polyvinylidene fluoride (PVDF) composite membrane [15]. This membrane exhibits super-hydrophilic and underwater super-oleophobic characteristics, conferring exceptional resistance to organic fouling. The incorporation of polydopamine not only enhances the hydrophilicity of the base membrane but also reinforces the stability of the interlayer architecture. During filtration of heavy metal-bearing solutions, the membrane captures target ions through the adsorption sites provided by GO while maintaining high water flux through its low-resistance channel structure. Despite their impressive laboratory performance, GO-based membrane materials confront substantial barriers to practical deployment, including membrane fouling, mechanical fragility under operational conditions, and the engineering challenges inherent in scaling membrane fabrication to industrial dimensions.

3.3 Chitosan/GO Composites

Chitosan, a naturally occurring polysaccharide, is characterized by its wide availability, biodegradability, and environmental benignity. Rich in amino ($-\text{NH}_2$) and hydroxyl ($-\text{OH}$) functional groups, chitosan, when hybridized with GO, yields composites with enhanced mechanical strength and a synergistic increase in adsorption-active sites [16]. Fan et al. provided a comprehensive review of the preparation methodologies and heavy metal adsorption–separation performance of chitosan/GO composites [16]. Their analysis revealed that the incorporation of GO effectively improves the acid resistance and thermal stability of chitosan, while chitosan reciprocally prevents the restacking of GO nanosheets, thereby increasing the effective specific surface area and mechanical robustness of the composite. Benefiting from the complementary functionality of $-\text{COOH}$ and $-\text{NH}_2$ groups, the composite exhibits markedly enhanced adsorption capacities for heavy metal ions. The chitosan–magnetic GO composite employed by Sherlala et al. for arsenic adsorption represents a successful exemplar of this hybridization strategy, underscoring its potential for rational adsorbent optimization [7]. Moreover, compared with pristine GO, chitosan-based composites align more closely with the principles of green chemistry and sustainable development,

positioning them as promising candidates for future environmental remediation applications.

3.4 Other Functionalized Composites

Beyond the categories discussed above, a diverse array of purpose-designed functional GO-based materials has been extensively investigated. Ruan et al. reviewed studies in which GO was modified through the grafting of thiol ($-SH$) and polyamine moieties to reinforce adsorption performance; these modifications substantially enhanced the material's affinity for specific soft metal ions, with commensurate improvements in adsorption efficiency [4]. In a separate contribution, Zhang et al. highlighted that GO and its composites can serve not only as adsorbents but also as platforms for heavy metal ion detection when coupled with fluorescence or electrochemical sensing modalities, thereby demonstrating the potential of GO-based materials as integrated adsorption–detection systems [17].

4. Adsorption Mechanisms and Model Analysis

A thorough understanding of adsorption kinetics is indispensable for the rational design of adsorbent materials. Wang and Guo provided a systematic review of adsorption kinetic models, elucidating their physical underpinnings, boundary conditions, and applicability, thereby establishing a rigorous methodological foundation for the interpretation of adsorption data [18].

4.1 Adsorption Kinetics

Adsorption kinetics investigations seek to elucidate the rate-governing mechanisms and the underlying physicochemical processes that dictate adsorption dynamics. The three most widely applied kinetic models are: the pseudo-first-order model, the pseudo-second-order model, and the Weber–Morris intra-particle diffusion model. Extensive experimental evidence indicates that the adsorption of heavy metal ions by GO and its composites is predominantly described by the pseudo-second-order kinetic model [18,19]. However, the good fit to this model alone does not inherently prove that chemisorption is the rate-limiting step; definitive mechanistic identification requires complementary evidence from XPS, FTIR, activation energy analysis, and diffusion data to confirm the underlying rate-controlling process.

4.2 Adsorption Isotherms

Adsorption isotherms, which describe the functional relationship between the equilibrium concentration of adsorbate and the amount adsorbed per unit mass of adsorbent at constant temperature, constitute a fundamental tool for evaluating adsorption capacity. The two most representative models are the Langmuir and Freundlich equations. The Langmuir model postulates a homogeneous adsorbent surface in which each active site accommodates a single adsorbate molecule, yielding monolayer adsorption; it is primarily employed to estimate the maximum adsorption capacity. The Freundlich model, by contrast, assumes surface heterogeneity and is applicable to multilayer adsorption, providing parameters that characterize adsorption intensity and surface heterogeneity. The preponderance of published data indicates that the adsorption behavior of heavy metal ions on GO and its composites conforms more closely to the Langmuir model [18,20]. This observation suggests a relatively uniform energy distribution among adsorption-active sites and an adsorption mechanism that favors monolayer surface coverage.

4.3 Adsorption Thermodynamics

Thermodynamic analysis of adsorption reveals the influence of temperature on adsorption equilibrium, as well as the spontaneity and energy transformations inherent to the adsorption process. This is accomplished by analyzing experimental data obtained at varying temperatures and computing three fundamental thermodynamic parameters: the standard Gibbs free energy change (ΔG°), the standard enthalpy change (ΔH°), and the standard entropy change (ΔS°). For GO-based materials, the adsorption of heavy metal ions is characteristically a spontaneous endothermic process. The negative values of ΔG° confirm the thermodynamic spontaneity of adsorption, while the positive values of ΔH° indicate that the process is endothermic, implying that moderate temperature elevation enhances adsorption capacity [11,20]. The observation of a positive ΔS° —seemingly counterintuitive for an adsorption process—admits a well-established explanation: heavy metal ions in aqueous solution exist as hydrated species, and upon coordination with oxygen-containing functional groups on the GO surface, the hydration sheath is disrupted through desolvation. The liberation of previously constrained water

molecules into the bulk solution generates a substantial entropy gain that more than compensates for the loss of translational entropy accompanying ion immobilization, thereby driving a net increase in the total entropy of the system.

4.4 Integration of Theoretical Computation

Although the classical models discussed above effectively describe macroscopic adsorption capacity, kinetics, and energetics, they offer limited mechanistic insight into the origin of pronounced differences in adsorption capacity among different heavy metal ions on the same adsorbent material. Deshwal et al. have emphasized the growing trend of integrating experimental investigation with theoretical computation—principally density functional theory (DFT)—to address this gap [11]. DFT calculations enable the determination of preferential adsorption configurations, binding energies, and the specific interaction modes between functional groups and metal ions at the molecular and atomic levels, thereby providing a mechanistic rationale for experimentally observed selectivity sequences such as $\text{Pb}^{2+} > \text{Cu}^{2+} > \text{Cd}^{2+}$.

5. Challenges and Future Perspectives

Despite the exceptional performance demonstrated by GO-based materials under controlled laboratory conditions, a substantial gap persists between bench-scale promise and full-scale engineering deployment. The principal challenges include, but are not limited to, the following:

Discrepancy between simulated and real water matrices. Laboratory studies typically employ synthetic aqueous solutions that bear limited resemblance to real-world water bodies. Actual wastewaters contain complex mixtures of co-existing ions, dissolved organic matter, and microorganisms, all of which can profoundly compromise adsorption performance [11].

Environmental risk considerations. The potential ecological risks associated with nanomaterial release necessitate a comprehensive life-cycle assessment. Ahmad et al. have specifically cautioned that the ecotoxicity of GO itself is non-negligible, underscoring the imperative that remediation strategies avoid introducing derivative environmental hazards in the course of pollutant removal [12].

Scalability and cost-effectiveness. The majority

of current composite materials entail elaborate fabrication protocols and elevated production costs. The development of environmentally benign and scalable synthesis routes is a prerequisite for the translation of laboratory innovations to practical water treatment infrastructure.

Looking forward, it is proposed that research efforts be strategically directed along the following trajectories: (i) the deep integration of high-throughput experimentation with DFT and molecular dynamics simulations to accelerate the discovery and optimization of novel functionalized GO adsorbents; (ii) the fabrication of multifunctional GO-based membranes that integrate adsorption, sensing, and catalytic degradation capabilities within a single platform; and (iii) the development of technologies for the deep purification and highly selective enrichment of trace heavy metals, in alignment with increasingly stringent drinking water safety standards [11,15,17].

6. Conclusions

Graphene oxide-based composite materials constitute a remarkably versatile platform for the remediation of heavy metal-contaminated water. Through multi-scale regulation-spanning chemical modification to impart molecular-level "recognizability" to functional groups, the engineering of magnetic or structural strategies for nanomaterial recovery, and the optimization of process-level operability—researchers are progressively transforming this two-dimensional nanomaterial into a comprehensive adsorption toolkit. Although the pathway from laboratory discovery to engineering practice necessitates overcoming multiple hurdles related to long-term stability and cost-effectiveness, the convergence of multidisciplinary advances in materials science, environmental chemistry, and computational modeling is steadily rendering the prospects for GO-based remediation technologies increasingly compelling.

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