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Green Synthesis and Functionalization of Graphene-like Nanocomposites for Heavy Metal Remediation: A Systematic Review of Mechanisms and Performance Prospects

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Abstract

In recent years, the search for heavy metal sorbents with high capacity, selectivity, regenerability, and sustainability has been intensified, as heavy metal contamination of water resources remains one of the most persistent environmental threats to ecosystem integrity and public health. Graphene-like materials, comprising graphene oxide (GO), reduced graphene oxide (rGO), and their composites, offer an ultrahigh theoretical surface area, a tunable surface chemistry, and an abundance of oxygenated binding sites, all of which are ideal for capturing metal ions. However, conventional production routes rely on strong oxidants and hazardous reducing agents such as hydrazine, which conflict with the environmental purpose of the resulting materials. Using a PRISMA-guided systematic approach filtering 412 records down to 57 included studies (2013–2026), this review synthesizes recent progress in the green synthesis of graphene-like nanomaterials, including plant extract-mediated and microbial reduction of GO, biomass- and waste-derived graphene, and reductant-free electrochemical routes. It examines how these materials are functionalized with magnetic phases, biopolymers, metal oxides, layered double hydroxides, and chelating ligands to produce nanocomposites tailored for heavy metal remediation. The molecular mechanisms of metal uptake, spanning electrostatic attraction, surface complexation, chelation, ion exchange, precipitation, and redox-mediated immobilization, are analyzed alongside the

isotherm, kinetic, and thermodynamic models used to describe them. A comparison of reported performance for priority contaminants such as Pb(II), Cd(II), Cr(VI), Hg(II), As, and others is presented, with capacities in the range of 100–560 mg/g for magnetic or macroscopic formats, which facilitate separation and re-use. Finally, the review examines challenges that hinder translation, such as aggregation, regeneration efficiency, cost, scalability, and nanomaterial release, as well as research pathways most likely to yield deployable, environmentally viable graphene-like adsorbents.

Keywords: Adsorption; Functionalization; Graphene; Graphene Oxide; Green Synthesis; Heavy Metals; Mechanism; Nanocomposite; Water Remediation

1. Introduction

Heavy metals such as, Pb, Cd, Hg, Cr and As are among the most significant pollutants, due to their persistent nature, toxicity at low concentrations and high capacity for bioaccumulation in food chains (Baby et al., 2019; Singh et al., 2021; Abbas et al., 2024). Chronic exposure to Pb(II) is associated with neurodevelopmental damage, Cd(II) with renal dysfunction and bone disease, Hg(II) with neurological impairment, Cr(VI) with carcinogenicity and arsenic with a spectrum of cancers and cardiovascular effects. Consequently, regulatory limits for these contaminants in drinking water lie in the low microgram-per-liter range and continue to tighten (Singh et al., 2021; Tripathy et al., 2024).

The sources of contamination are diverse and expanding. Mining and mineral processing, electroplating, tanning, battery and electronics manufacture, pigment production, and fertilizer-intensive agriculture discharge metal-bearing effluents that are frequently treated inadequately, particularly in rapidly industrializing regions (Baby et al., 2019; Abbas et al., 2024). Unlike organic pollutants, heavy metals cannot be degraded. They can only be transferred between phases, immobilized or recovered, making separation and capture technologies the core of any remediation plan (Singh et al., 2021).

Traditional treatment technologies all have proven limitations. Chemical precipitation is low cost but produces large quantities of hazardous sludge and is ineffective at low residual concentration; coagulation and flocculation have the same sludge problem; ion exchange resins are expensive and foul in complex matrices; membrane processes require high energy and generate concentrate streams; and electrochemical methods involve high electrode costs and passivation problems (Tripathy et al., 2024). In this context, adsorption is considered to be the most flexible, simple, and economical method, especially for the treatment of effluents to trace levels (Baby et al., 2019; Wang & Guo, 2023).

Although activated carbon has been most prevalent in adsorptive treatment, its limited capacity, poor selectivity for competing ions, and need for energy-intensive

regeneration have spurred a search for better sorbents (Baby et al., 2019; Tripathy et al., 2024). Nanomaterials have emerged as the leading candidate due to an ultrahigh surface-area-to-volume ratio, abundant binding sites, and a tunable surface chemistry that easily translates into higher capacities and faster kinetics (Wang & Guo, 2023; Abbas et al., 2024).

Within this class, graphene-like materials have drawn tremendous attention over the past decade (Chang & Wu, 2013; Upadhyay et al., 2014). Graphene oxide, a product of oxidative exfoliation of graphite, possesses high densities of hydroxyl (–OH), epoxide (C–O), and carboxyl (C=O) groups capable of binding metal cations, while rGO features a partial restoration of graphitic conductivity and π-domains for cation–π interactions with hydrated metal cations (Chang & Wu, 2013; Peng et al., 2017; Ghugare & Jadhav, 2026). Sitko et al. (2013) established GO as an outstanding sorbent for a series of divalent metal ions, with capacities exceeding those of previously reported sorbents, and subsequent reviews have documented the rapid expansion of graphene-based adsorption chemistry as a major remediation platform (Peng et al., 2017; Xu & Wang, 2017; Asghar et al., 2022; Abbas et al., 2024).

The sustainability of the sorbent, however, has emerged as a major problem. Traditional Hummers-type oxidation uses concentrated sulfuric acid, permanganate, and nitrate, while conventional chemical reduction of GO relies on hydrazine or sodium borohydride. These agents are toxic and corrosive, rendering them incompatible with the environmental goals of the resulting materials (Asghar et al., 2022; Ghugare & Jadhav, 2026). To address this contradiction, green synthesis focuses on using plant extracts, microbial and biomolecular reductants, waste biomass feedstocks, and reductant-free electrochemical routes for adsorbent manufacture, aligning the process with the principles of green chemistry (Suresh et al., 2015; Dautov et al., 2026; Rai et al., 2026; Swaroop et al., 2026).

A second problem is practical applicability. Pristine GO disperses too well; post-treatment separation is difficult due to its colloidal stability, metal cations induce sheet aggregation and morphological transformations, and the uncontrolled release of nanomaterials into treated water creates secondary ecological concerns (Yang et al., 2016; Asghar et al., 2022; Jassim et al., 2025). Functionalization addresses these issues by anchoring graphene-like sheets within magnetic, biopolymeric, or inorganic hybrid architectures. These setups add selective binding chemistries, enable magnetic or gravitational recovery, and permit shaping into beads, hydrogels, and monoliths suitable for real reactors (Mokoena & Mofokeng, 2023; Mubarak et al., 2026).

Despite a growing number of reviews on graphene-based adsorbents, the literature remains fragmented. Green synthesis, functionalization chemistry, adsorption mechanisms, and contaminant-specific performance are typically treated in isolation, and the field has expanded so rapidly since 2020 that earlier syntheses no longer reflect the state of the art (Peng et al., 2017; Xu & Wang, 2017;

Baby et al., 2019; Abbas et al., 2024). The present review therefore applies a systematic methodology (Section 2) to integrate the full value chain—from sustainable precursor chemistry to regenerable engineered adsorbents. Specifically, it identifies available green routes for producing graphene-like materials, evaluates functionalization strategies that convert these materials into practical nanocomposites, analyzes the mechanisms driving heavy metal capture, reviews performance against priority contaminants, and outlines the emerging challenges facing their widespread deployment.

2. Methodology

This review was designed and conducted as a systematic review, following a protocol defined before screening began and reported in alignment with the PRISMA 2020 statement for transparent systematic reviews. The methodology comprises six elements: review design, research questions, search strategy, eligibility criteria, screening and selection, and data extraction and synthesis.

2.1 Review Design and Reporting Standard

The present review followed a systematic, PRISMA-style workflow (Figure 1), utilizing a qualitative and thematic synthesis rather than a meta-analytic approach (Wang & Guo, 2023).

2.2 Research Questions

The review is organized around four research questions. RQ1: which green synthesis routes are available for producing graphene-like materials, and what trade-offs in yield, quality, and sustainability do they entail? RQ2: Which functionalization strategies convert these materials into practical nanocomposite adsorbents, and through what design logic? RQ3: Through which molecular mechanisms do green functionalized graphene-like nanocomposites capture heavy metals, and how is uptake described by isotherm, kinetic, and thermodynamic models? RQ4: What performance has been demonstrated against priority contaminants, and which barriers and prospects govern translation to real-world water treatment?

2.3 Search Strategy

Relevant literature was identified through systematic searches of Google Scholar, ScienceDirect, SpringerLink, ACS Publications, RSC Publishing, Wiley Online Library, Taylor & Francis Online, and MDPI, with the search finalized in October 2026. Search strings combined the following categories using the Boolean operators “OR” and “AND”: (i) material terms ("graphene", "graphene oxide", "reduced graphene oxide", "graphene-like nanocomposite"); (ii) synthesis and modification terms ("green synthesis", "biosynthesis", "plant extract", "biomass-derived",

"electrochemical exfoliation", "functionalization", "magnetic", "chitosan", "biochar"); and (iii) application terms ("heavy metal", "adsorption", "Pb(II)", "Cd(II)", "Cr(VI)", "Hg(II)", "arsenic", "wastewater remediation"). Reference lists of retrieved reviews were further searched to capture additional primary studies missed by keyword searches.

2.4 Eligibility Criteria

Studies were screened against the pre-specified inclusion and exclusion criteria summarized in Table 1. The 2013 start date reflects the maturation of graphene-based environmental adsorption research (Chang & Wu, 2013).

Table 1. Pre-specified eligibility criteria applied during screening

Inclusion criteria	Exclusion criteria
Peer-reviewed journal article, review, or scholarly book chapter	Conference abstracts and preprints without full text
Published in English between 2013 and October 2026 (priority to 2020-2026)	Published before 2013, or not in English
Reports green or low-impact synthesis of graphene-like materials, or functionalized graphene-like nanocomposites	No graphene-like component, or no green synthesis / functionalization element
Applies the material to heavy metal removal from water or wastewater	Non-aqueous matrices only, or no heavy metal application
Reports at least one quantitative or mechanistic descriptor (capacity, removal efficiency, isotherm/kinetic fitting, regeneration)	Insufficient performance or mechanistic data

2.5 Screening and Selection

Database searching returned 412 records; after the removal of 116 duplicates, 296 unique records were screened by title and abstract, of which 198 were excluded as clearly irrelevant. The remaining 98 full texts were assessed for eligibility, and 41 were excluded for the reasons detailed in Figure 1: publication before 2013 (n = 9), no heavy metal application (n = 12), no green synthesis or functionalization component (n = 11), and insufficient quantitative or mechanistic performance data (n = 9). The final corpus comprised 57 studies, with the vast majority published within the last five years.

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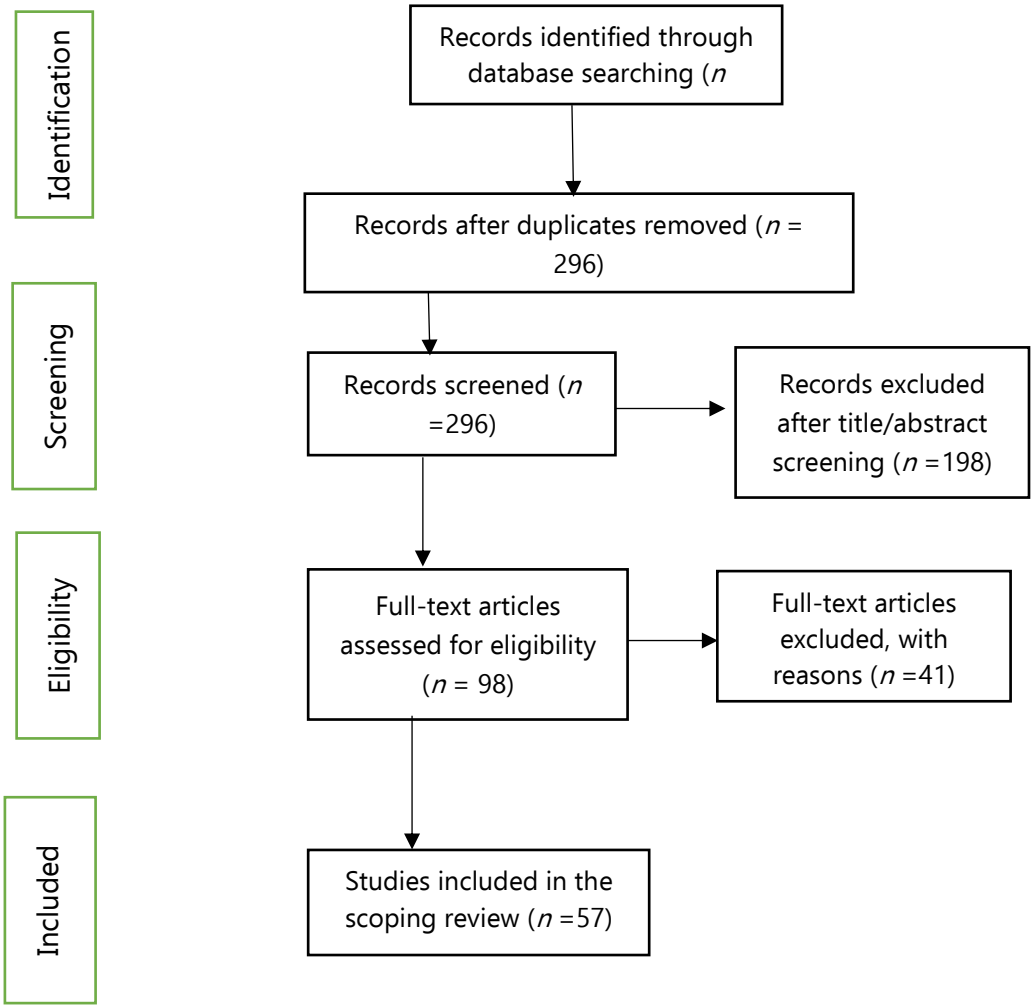


Figure 1. PRISMA Flow diagram

2.6 Data Extraction, Quality Assessment, and Synthesis

From each included study, the following data were extracted where available: the synthesis route and its green attributes, the functionalization strategy, target metal ion(s), the reported adsorption capacity or removal efficiency, isotherm and kinetic models, regeneration cycles, and the removal mechanism(s) proposed by the authors. Included studies were evaluated for methodological adequacy based on the clarity of synthesis reporting, the adequacy of material characterization, the rigor of isotherm and kinetic fitting, and evidence of regeneration or real-matrix testing. Findings were synthesized thematically along the remediation value chain: green synthesis (Section 4), functionalization (Section 5), removal mechanisms (Section 6), contaminant-specific performance (Section 7), and regeneration and deployment readiness (Sections 8–9); representative systems are compared in Table 2. Because operational parameters such as pH, initial

concentration, adsorbent dose, temperature, and water matrix varied widely across the evaluated studies, capacities were compared qualitatively and by orders of magnitude rather than through formal meta-analytic pooling. Limitations of the methodology include single-reviewer screening, restriction to English-language sources, and the possibility of publication bias toward high-performing materials.

3. Graphene-like Nanomaterials: Structures and Remediation-Relevant Properties

The remediation utility of graphene-like materials is based on several structural features. GO is a highly oxidized, hydrophilic derivative of graphite with –OH, C–O, C=O, and –COOH groups on the basal planes and edges which deprotonate with increasing pH to generate negatively charged metal binding sites (Peng et al., 2017; Ghugare & Jadhav, 2026). The same groups render GO highly hydrated and colloidally stable, which is beneficial for dispersibility but not desirable for solid-liquid separation. After reduction to rGO, some oxygen functionalities are removed, thus restoring part of the conjugated sp^2 network; the oxygen functionalities and defects that remain anchor metal ions, while the graphitic domains contribute to π-cation interactions (Chang & Wu, 2013; Peng et al., 2017; Jassim et al., 2025).

Holey graphene oxide, with in-plane nanopores, reduces the diffusion path length and exposes more edge sites, which enhances the uptake kinetics of heavy metal ions (Kahve Yildirim et al., 2025). Three-dimensional assemblies such as aerogels, hydrogels, and cryogels can transform two-dimensional sheets into flow-compatible, interconnected monolithic architectures with high microporosity (Nasiri et al., 2018; Pinelli et al., 2023; Sharma et al., 2023; Mubarak et al., 2026). In all of the variants, the same performance levers exist: the density and chemical speciation of binding sites, available surface area, colloidal or hydraulic stability and separability from treated water (Asghar et al., 2022; Tripathy et al., 2024).

Notably, the graphene-like adsorbents are not inert scaffolds in solution. Yang et al. (2016) found that the aggregation and morphological transformation of GO are induced by the adsorption of metal cations through charge screening and bridging, suggesting that adsorption performance, transport behavior, and environmental fate are coupled phenomena. Therefore, it is important to take into account the engineered function and colloidal behavior of graphene-like nanocomposites in realistic water chemistries (Yang et al., 2016; Singh et al., 2021).

4. Green Synthesis of Graphene-like Materials

Green synthesis seeks to replace hazardous oxidants and reductants with benign alternatives, valorize waste feedstocks, and reduce energy and water intensities, while preserving the surface chemistry that makes graphenic materials effective adsorbents (Ghugare & Jadhav, 2026; Swaroop et al., 2026). Four primary synthesis routes dominate the recent literature:

4.1 Plant Extract-Mediated Reduction

Plant extracts containing polyphenols, flavonoids, terpenoids, and ascorbic acid serve as both reducing and capping agents by donating electrons to GO and inhibiting the restacking of GO sheets (Ghugare & Jadhav, 2026). Common, inexpensive botanicals can be used in place of synthetic reductants, as demonstrated by Suresh et al. (2015) using spinach, and by Tayade et al. (2019) using plant seed extracts. More recent work utilizes *Mimosa pudica* leaf extract to produce rGO with tunable structure-property relationships (Kindalkar et al., 2026), neem (*Azadirachta indica*) leaf extract to produce green rGO applied directly to the removal of heavy metals and dyes from industrial wastewater (Rai et al., 2026), and *Eucalyptus globulus* leaf extract to mediate the one-pot green synthesis of iron oxide-GO nanocomposites for Cd(II) and Pb(II) removal (Bhushan & Gupta, 2025).

A systematic analysis by Ghugare and Jadhav (2026) shows that extract composition, such as polyphenol content, is a key factor in the degree of reduction, morphology, and conductivity of the product, thereby offering a rational basis for extract selection.

Furthermore, the advantages of this method are significant for remediation applications: the reductants used are renewable and non-toxic, reactions occur in water at moderate temperatures, and the phytochemicals on the sheets' surface provide metal-chelating hydroxyl and carbonyl groups (Ghugare & Jadhav, 2026; Rai et al., 2026). Conversely, the major limitations include batch-to-batch variability of natural extracts, incomplete reduction relative to chemical agents, and the need for inter-laboratory standardization (Ghugare & Jadhav, 2026).

4.2 Biomass- and Waste-Derived Graphene-like Materials

A more radical green approach completely avoids graphite in favor of graphenic carbon derived from biomass or industrial waste. For instance, the use of agricultural residues to produce materials for adsorption and remediation was demonstrated by Swaroop et al. (2026), who synthesized GO and rGO from sugarcane bagasse. Similarly, Chee et al. (2025) produced GO from sago waste for Pb(II) removal from water, while Akhtar et al. (2026) developed bio-based GO-magnetite nanocomposites as a single remediation platform for arsenic removal. To target Cd(II) capture, Ghugare et al. (2024) modified GO sorbents with waste orange peel biochar. Highlighting a notable circular-economy application, Karim and Uddin (2026) fabricated gelatin-GO hydrogels from tannery waste to treat Cr-contaminated tannery wastewater. While these synthesis routes lower precursor costs, minimize waste generation, and enhance life-cycle sustainability, they typically suffer from lower-quality graphitization and poorer reproducibility compared to graphite-derived GO (Swaroop et al., 2026).

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4.3 Microbial, Biomolecular, and Electrochemical Routes

Alternative benign routes to rGO utilize microbial respiration and biomolecular reductants at ambient temperature and pressure with water as the solvent, as reviewed in detail elsewhere (Baby et al., 2019; Ghugare & Jadhav, 2026). Similarly, the electrochemical exfoliation of graphite—wherein graphite is expanded and delaminated via ion intercalation at an applied potential—completely eliminates the need for strong chemical oxidants. Instead, oxygen functional groups are generated directly on the exfoliated surface, thereby facilitating ion adsorption (Dautov et al., 2026). Leveraging this mechanism, Ithnin and Liu (2024) combined electrochemical exfoliation with plant-extract reduction to synthesize graphene without conventional chemical oxidants. Ultimately, its reagent-free operation, precise potential control, and exclusive reliance on electricity render electrochemical methods excellent candidates for the scale-up of sustainable remediation processes (Dautov et al., 2026).

5. Functionalization Strategies for Heavy Metal Capture

Pristine GO and rGO are rarely deployed alone. Functionalization pursues four goals: adding high-affinity or selective binding chemistries, imparting magnetic or macroscopic separability, preventing sheet aggregation to preserve the accessible area, and improving mechanical and chemical robustness for regeneration (Chang & Wu, 2013; Asghar et al., 2022; Mokoena & Mofokeng, 2023). The strategies below are organized by the functional partner's type rather than by the chemistry of the graphene sheet itself.

5.1 Magnetic Functionalization

Decorating graphene-like sheets with Fe_3O_4 or spinel ferrite nanoparticles is the single most common functionalization strategy, because it converts a colloidally stable, difficult-to-separate sorbent into a magnetically recoverable one without sacrificing surface chemistry (Jassim et al., 2025; Akhtar et al., 2026). Magnetic GO and rGO composites have been applied across the priority metals: rGO-Fe_3O_4@polydopamine for Pb(II) (Mehdinia et al., 2020), Fe_3O_4-polypyrrole-GO for Hg(II) (Zhou et al., 2017), glutathione-functionalized $NiFe_2O_4$/GO for Hg(II), Cu(II), and Pb(II) (Khorshidi et al., 2020), lanthanum sulfide-decorated magnetic GO for Pb(II) (Rezania et al., 2022), and $NiFe_2O_4$-decorated chitosan-GO for Co(II) (Guo & Bulin, 2024).

Although the magnetic phase is generally non-reactive toward the target ion, it provides hydroxylated oxide surfaces for the adsorption process while facilitating solid–liquid separation via an external magnet. Because this separation has a low energy requirement, it eliminates the need for centrifugation or membrane filtration (Zhou et al., 2017; Huang et al., 2018). A trade-off between saturation magnetization

and oxide loading exists, given that excess ferrite reduces the available graphene-like binding sites. To address this, the three-dimensional graphene/chitosan/$NiFe_2O_4$ composite developed by Nasiri et al. (2018)—which features a saturation magnetization of ~11 emu/g—demonstrates a practical compromise that allows efficient magnetic recovery.

5.2 Biopolymer-Based Composites

Biopolymers serve as ideal matrices for graphene-like sheets (Mokoena & Mofokeng, 2023), combining inherent eco-friendliness with robust metal-binding functionality. Specifically, the chelation efficiency of chitosan toward Pb(II), Cd(II), and Cu(II) is driven by its abundant amine and hydroxyl groups, which actively coordinate heavy metal ions. When fabricated as cross-linked chitosan–GO cryogels, these materials form macroporous monoliths well-suited for simultaneous multi-metal removal in continuous flow-through formats (Mubarak et al., 2026). Magnetic chitosan/GO composites extend the approach to rare-earth recovery, as shown for neodymium (Al-Salem et al., 2024). Ternary chitosan/GO/polypyrrole composites have also been validated in applications ranging from batch systems to real industrial effluents (Maity et al., 2026).

Beyond chitosan, other biopolymer platforms are equally active. Gelatin–GO hydrogels derived from tannery waste target Cr in the same effluent stream from which the gelatin was recovered (Karim & Uddin, 2026). Other polysaccharide platforms are equally active: agar–GO nano-biocomposites adsorb Cd(II) with outstanding reuse capacity (Gomes et al., 2024), bacterial cellulose–GO composite films provide eco-friendly Pb(II) removal (Nguyen, 2025), magnetic hydrogel beads based on modified gum tragacanth/GO remove both heavy metals and dyes with high capacities and uptake rates (Sahraei et al., 2017), and pH-tunable GO/chitosan beads have been demonstrated in fixed-bed columns with breakthrough modeling (Duong et al., 2025).

Ultimately, these biopolymer matrices solve a critical shaping problem. They convert nanomaterial sheets into practical macroscopic forms such as beads, hydrogels, aerogels, and cryogels that can be efficiently packed into engineering columns for continuous water treatment (Pinelli et al., 2023; Mubarak et al., 2026).

5.3 Metal Oxide, Sulfide, and Layered Double Hydroxide Hybrids

Incorporating inorganic phases into the carbon scaffold provides additional adsorption mechanisms. Metal oxides such as ZnO, NiO, and iron oxides introduce hydroxylated surfaces and, in some cases, photocatalytic properties that enable co-contaminants to be oxidized or reduced upon adsorption (Upadhyay et al., 2014; Mohamed et al., 2023; Ahmad et al., 2026). Sulfide phases exploit soft–soft interactions: lanthanum sulfide on magnetic GO strongly complexes Pb(II), a soft Lewis acid (Rezania et al., 2022). In a one-pot solvothermal fabrication of magnetic

GO/MgAl-LDH nanocomposites, Huang et al. (2018) immobilized lead, cadmium, and copper ions via simultaneous complexation, precipitation, and isomorphic substitution mechanisms, while achieving magnetic separability. Overall, although inorganic hybrids generally exhibit higher capacities than the parent GO, their long-term chemical stability must be carefully evaluated, as the dissolution of oxides or the oxidation of sulfides during acid regeneration may generate secondary contaminants (Huang et al., 2018; Ahmad et al., 2026).

5.4 Chelating Ligands, Amine Polymers, and Molecular Grafts

Molecular functionalization installs coordination chemistry tuned to the target ion. Carboxyl-rich ligands such as EDTA grafted onto magnetic GO/cellulose hydrogels markedly increase Pb(II) capacity by providing multidentate chelation sites (Dacrory et al., 2022), while simply enriching GO with carboxyl groups (carboxylated GO) enhances Pb(II) and Cu(II) uptake, including in real wastewater matrices (Van Tran et al., 2025). Amine-bearing modifiers are particularly effective. Piperazine-modified magnetic GO achieved a Langmuir capacity of 558.2 mg/g for Pb(II), which is among the highest reported for a magnetically separable graphene-like sorbent (Alboghbeish et al., 2022). Similarly, 3,5-diaminobenzoic acid-GO composites selectively captured Pb(II) and Al(III) from wastewater (Farag et al., 2022). Amine-functionalized GO integrated with the metal–organic framework MIL-88A also showed enhanced Cr(VI) removal (Hammad et al., 2024).

In contrast, capturing softer heavy metals requires distinct surface chemistry. Thiolated ligands such as glutathione confer selectivity for soft metal ions including Hg(II) (Khorshidi et al., 2020), while catechol-rich polydopamine coatings provide a universal adhesive layer whose amine and phenolic groups bind diverse metals (Mehdinia et al., 2020). The design logic follows hard–soft acid-base theory: carboxyl and amine donors suit borderline ions such as Pb(II) and Cd(II), whereas sulfur donors target Hg(II) and other soft cations (Peng et al., 2017; Wang & Guo, 2023).

5.5 Emerging Hybrid Architectures

Recent work has begun to integrate GO with metal–organic frameworks, covalent organic frameworks, and three-dimensional porous assemblies to combine molecularly designed porosity with graphene-like surface chemistry (Mahmoodi et al., 2023; Hammad et al., 2024; Mubarak et al., 2026). Green synthesis variants of such hybrids, including magnetic Al-fumarate MOF–GO composites prepared under mild conditions, indicate that framework chemistry can be reconciled with sustainable processing (Mahmoodi et al., 2023). Fungal-hyphae biochar/rGO/iron oxide composites—which combine biotemplating with magnetic functionality and have undergone testing in real wastewater—represent a further convergence of green feedstocks with hybrid design (Deveci et al., 2025). These architectures remain at a nascent stage; their increased synthetic complexity will require commensurate

improvements in selectivity or reusability before they can compete at scale with simpler magnetic or biopolymer composites.

6. Mechanisms of Heavy Metal Removal

Heavy metal capture by graphene-like nanocomposites is rarely driven by an isolated interaction. Rather, spectroscopic analyses, molecular modeling, and theoretical calculations consistently identify a multi-mechanistic pathway whose relative importance depends heavily on pH, ionic strength, metal speciation, and the surface functional groups present (Peng et al., 2017; Elgengehi et al., 2020; Singh et al., 2021; Wang & Guo, 2023).

6.1 Electrostatic Attraction and pH Dependence

Above the pH point of zero charge, the deprotonation of surface carboxyl and hydroxyl groups imparts a negative charge to graphene-like surfaces. This shifts the mechanism toward stronger electrostatic attraction for metal cations, driving a rapid, pH-dependent increase in the adsorption of ions such as Pb(II), Cd(II), and Cu(II) until the onset of hydrolysis or precipitation (Sitko et al., 2013; Peng et al., 2017; Van Tran et al., 2025). Furthermore, the observation that metal sorption in GO systems remains relatively insensitive to ionic strength suggests that a fraction of the bound metal exhibits inner-sphere coordination rather than a purely outer-sphere, electrostatic association (Sitko et al., 2013; Peng et al., 2017). Ultimately, within multi-component cryogel architectures, these electrostatic forces operate synergistically with chemical chelation (Mubarak et al., 2026).

6.2 Surface Complexation and Chelation

In the functionalized composites, the adsorption is dominated by the surface complexes formed by lone-pair donation of oxygen, nitrogen and sulfur functionalities into the empty orbitals of the metal ions. The performance of chitosan-GO, piperazine-MGO, and chitosan/GO/polypyrrole systems is attributed to amine chelation (Alboghbeish et al., 2022; Maity et al., 2026; Mubarak et al., 2026); carboxylate coordination is responsible for the performance of EDTA-grafted hydrogels and carboxylated GO (Dacrory et al., 2022; Van Tran et al., 2025); and thiol complexation is the basis for the selectivity of the composites towards Hg(II) (Khorshidi et al., 2020; Zhou et al., 2017). Energy calculations using density functional theory (DFT) confirm that oxygenated vacancies and functional groups are energetically preferred binding sites on GO (Elgengehi et al., 2020).

6.3 Ion Exchange, Precipitation, and Isomorphic Substitution

In hybrid composites, inorganic partners contribute additional removal pathways. Protons and interlayer cations exchange with target metals at oxide and LDH surfaces; hydroxide and carbonate precipitation immobilizes metals at high

local pH near basic surfaces; and isomorphic substitution can lock divalent metals into mineral lattices (Huang et al., 2018; Singh et al., 2021). Huang et al. (2018) showed that the magnetic GO/MgAl-LDH nanocomposite removes Pb(II), Cd(II), and Cu(II) through precisely this combination of complexation, surface precipitation, and lattice incorporation, which also explains the high stability of the captured metals against desorption. For ferrite-bearing biopolymer composites such as $NiFe_2O_4$-chitosan-GO, mechanistic analyses similarly resolve contributions from electrostatic attraction, complexation, and ion exchange acting in parallel (Guo & Bulin, 2024).

6.4 Redox-Mediated Immobilization

For redox-active contaminants, adsorption can be coupled to transformation. Cr(VI), present as anionic chromate species, is attracted to protonated amine and oxide surfaces and can be reduced to the far less mobile and less toxic Cr(III) by electron-rich graphene-like domains and coexisting functional groups, after which the Cr(III) is retained by complexation (Xu & Wang, 2017; Abdel Salam et al., 2020; Jibin et al., 2023). This adsorption-coupled reduction is a distinct advantage over inert sorbents because it addresses both mobility and toxicity in one unit operation; furthermore, it is a recurring feature of green composites whose residual phytochemical or biopolymer functionalities act as electron donors (Karim & Uddin, 2026). Recent binary-sorbate work on rGO-supported adsorbents confirms that Cr(VI) reduction–complexation can proceed alongside cationic Pb(II) uptake on the same material (Kanwal et al., 2025), and MOF-amine-GO hybrids exploit protonated amine sites to enhance anionic Cr(VI) capture (Hammad et al., 2024).

6.5 Adsorption Modeling: Isotherms, Kinetics, and Thermodynamics

Most published data on graphene-like nanocomposites utilize the Langmuir isotherm to describe equilibrium data and the pseudo-second-order model to model adsorption kinetics, a trend widely interpreted as evidence of chemisorption (Alboghbeish et al., 2022; Wang & Guo, 2023; Guo & Bulin, 2024). Conversely, Freundlich and Sips behaviors manifest in heterogeneous hydrogels and multilayer systems (Sahraei et al., 2017; Gomes et al., 2024). Furthermore, intraparticle diffusion analyses generally reveal that film and pore diffusion act sequentially rather than independently (Singh et al., 2021). Thermodynamic evaluations regularly yield negative Gibbs free energies and positive enthalpies, signifying spontaneous and endothermic sorption of divalent metal ions onto GO and its composites (Sitko et al., 2013; Guo & Bulin, 2024). Crucially, Wang and Guo (2023) caution against treating pseudo-second-order fits as conclusive proof of chemisorption, urging researchers to provide complementary spectroscopic data. This vital critique applies directly to the graphene-like literature examined in this review.

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7. Performance toward Priority Heavy Metals

Although performance outcomes vary significantly based on material architecture and experimental conditions, clear trends emerge from the studies summarized in Table 2. Specifically, among the investigated heavy metals, Pb(II) consistently exhibits the highest removal efficiency. Its borderline Lewis acidity, large ionic radius, and strong complexation with carboxyl and amine donors yield the highest capacities in the dataset, culminating in 558.2 mg/g for piperazine-modified magnetic GO under response-surface-optimized conditions (Alboghbeish et al., 2022), with additional Pb(II) successes reported for carboxylated GO in real wastewater (Van Tran et al., 2025), bacterial cellulose–GO films (Nguyen, 2025), and sago-waste-derived GO (Chee et al., 2025). Cadmium follows a similar chemistry at a generally lower capacity, with waste orange peel biochar-modified GO–chitosan sorbents (Ghugare et al., 2024), agar–GO biocomposites (Gomes et al., 2024), and green *Eucalyptus*-mediated iron oxide–GO nanocomposites (Bhushan & Gupta, 2025) demonstrating that low-cost green routes remain competitive.

Other metal and oxyanion targets exhibit distinct, mechanism-driven performance profiles. For instance, mercury performance is dictated by sulfur donor density, favoring thiolated and polypyrrole composites (Zhou et al., 2017; Khorshidi et al., 2020). Chromium remediation increasingly exploits adsorption-coupled reduction of Cr(VI) to Cr(III) on green hydrogels, biopolymer composites, and amine–MOF hybrids (Abdel Salam et al., 2020; Jibin et al., 2023; Hammad et al., 2024; Karim & Uddin, 2026), while arsenic removal has been demonstrated with bio-based GO–magnetite nanocomposites, extending the platform to oxyanionic contaminants (Akhtar et al., 2026). Cobalt, a less frequently studied target, is effectively captured by magnetic ferrite–biopolymer–GO composites (Guo & Bulin, 2024), and multi-metal simultaneous removal—essential for real effluents—has been shown for chitosan–GO cryogels treating combined Pb(II)/Cd(II)/Cu(II) streams (Mubarak et al., 2026) and for ternary chitosan/GO/polypyrrole composites in real industrial effluent (Maity et al., 2026).

Table 2. Reported performance of representative green-synthesized and functionalized graphene-like nanocomposites for heavy metal removal

Green / functionalized adsorbent	Target metal(s)	Performance highlight	Source
Pristine GO nanosheets	Zn(II), Cd(II), Cu(II), Pb(II)	Capacities exceeding contemporary sorbents; strongly pH-dependent uptake	Sitko et al., 2013
Piperazine-modified magnetic GO (Pip@MGO)	Pb(II)	q_m 558.2 mg/g; Langmuir isotherm, pseudo-second-order kinetics; four reuse cycles	Alboghbeish et al., 2022

Magnetic 3D graphene/chitosan /$NiFe_2O_4$	Pb(II)	>98% removal under RSM-optimized conditions; saturation magnetization about 11 emu/g	Nasiri et al., 2018
rGO–Fe_3O_4@polydopamine	Pb(II)	Magnetically separable; catechol/amine-driven uptake with isotherm and kinetic characterization	Mehdinia et al., 2020
Glutathione–$NiFe_2O_4$/GO	Hg(II), Cu(II), Pb(II)	Thiol chelation confers high affinity for soft metal ions; magnetic recovery	Khorshidi et al., 2020
Fe_3O_4-polypyrrole-GO	Hg(II)	N-donor polypyrrole boosts Hg(II) binding; rapid magnetic separation	Zhou et al., 2017
Magnetic GO/MgAl-LDH	Pb(II), Cd(II), Cu(II)	One-pot solvothermal synthesis; complexation plus precipitation and isomorphic substitution	Huang et al., 2018
EDTA–Si/mGO/AmCMC hydrogel	Pb(II)	Grafted EDTA chelation sites raise capacity over unmodified hydrogel	Dacrory et al., 2022
chitosan–GO cryogel beds	Pb(II), Cd(II), Cu(II)	Simultaneous multi-metal removal; over 90% reusability across cycles; flow-compatible monolith	Mubarak et al., 2026
Ternary chitosan/GO/polypyrrole composite	Highly toxic metal ions	Validated from batch systems through to real industrial effluent; reusable	Maity et al., 2026
$NiFe_2O_4$-decorated chitosan–GO	Co(II)	Magnetic biopolymer composite with resolved adsorption mechanism	Guo & Bulin, 2024
Gelatin–GO hydrogel from tannery byproducts	Cr(VI)/Cr(III)	Waste-derived sorbent applied to Cr remediation in tannery effluent	Karim & Uddin, 2026
Green rGO (*Azadirachta indica* leaf extract)	Heavy metals, dyes	Hydrazine-free reduction with direct industrial wastewater application	Rai et al., 2026
Eucalyptus globulus leaf-	Cd(II), Pb(II)	One-pot plant-mediated green synthesis applied to Cd(II)/Pb(II) removal	Bhushan & Gupta, 2025

modified iron oxide-GO			
GO-orange peel biochar-chitosan (GO-OP–CS)	Cd(II)	Waste-biochar-modified sorbent; regenerated with 0.1 M HNO_3	Ghugare et al., 2024
Agar-GO nano-biocomposite	Cd(II)	Polysaccharide composite with outstanding reuse capacity	Gomes et al., 2024
Sago waste-derived GO	Pb(II)	Biomass-derived GO applied to Pb removal from water	Chee et al., 2025
Carboxylated GO (GO–COOH)	Pb(II), Cu(II)	Carboxyl enrichment enhances capacity; validated in real wastewater	Van Tran et al., 2025
CPBr-modified MIL-88A@amine-GO	Cr(VI)	MOF-amine-GO hybrid with enhanced Cr(VI) uptake and mechanistic study	Hammad et al., 2024
Holey GO	Heavy metals	In-plane nanopores enhance ion transport and adsorption uptake	Kahve Yildirim et al., 2025
Bio-based GO/magnetite nanocomposite	As(V)/As(III)	Biomass-derived sorbent for arsenic removal with kinetic study	Akhtar et al., 2026
La_2S_3-decorated magnetic GO (MGO@LaS)	Pb(II)	Soft sulfide sites afford high Pb(II) affinity and magnetic recovery	Rezania et al., 2022

8. Regeneration, Reusability, and Environmental Fate

Regeneration is the basis of economic viability. Cationic metals are well eluted from protonatable binding sites with acidic eluents (typically dilute HCl or HNO_3). Ghugare et al. (2024) regenerated orange peel biochar–GO–chitosan sorbents with 0.1 M HNO_3. Functionalized composites have also retained their useful capacity after 4 to 5 adsorption-desorption cycles (Alboghbeish et al., 2022; Kanwal et al., 2025). The chitosan–GO cryogel obtained by Mubarak et al. (2026) showed a reusability above 90% after several cycles, and agar–GO biocomposites had excellent reusability (Gomes et al., 2024).

Magnetic formats make the logistics of regeneration much easier because recovery can be done faster between treatments (Zhou et al., 2017; Deveci et al., 2025). Likewise, aerogel and bead forms allow for regeneration of the sorbent in a packed-bed without significant loss of sorbent (Pinelli et al., 2023; Sharma et al., 2023; Duong et al., 2025). These engineered designs facilitate better handling, easier recovery, and seamless process integration, thereby expanding the real-world scalability of graphene-like adsorbents.

Despite the progress, the adsorption capacity of the materials decreases with repeated use. This loss is typically attributed to the chemisorbed, irreversible, loss of functional ligands and dissolution of oxide components during regeneration. Furthermore, relatively few studies have evaluated performance beyond ten adsorption-desorption cycles. Consequently, a critical knowledge gap persists regarding the long-term stability and operational durability of many graphene-like nanocomposites (Baby et al., 2019; Verma et al., 2025).

Equally vital are the environmental fate and impact of spent adsorbents. Yang et al. (2016) demonstrated that metal cations induce the aggregation and morphological transformation of GO. Consequently, spent graphene-like sorbents tend to precipitate rather than remain dispersed within aquatic systems, thereby mitigating concerns regarding nanomaterial transport. Nevertheless, ecotoxicological data regarding green-synthesized and phytochemical-capped graphene-like materials remain scarce. To ensure that environmental remediation does not inadvertently cause ecological harm, future studies should focus on extensive field-scale performance testing and life-cycle risk assessments to avoid unintended ecological impacts from remediation (Singh et al., 2021; Ghugare & Jadhav, 2026) (Singh et al., 2021; Ghugare & Jadhav, 2026).

9. Challenges and Performance Prospects

Though laboratory results have been promising, there are a few limitations that need to be overcome for the translation of the green-synthesized graphene-like nanocomposites into practical water treatment technologies. Reproducibility is a critical consideration due to the seasonal, geographical, and processing variability of biomass feedstocks that may affect the material properties and the synthesis results. To allow for meaningful comparisons between different studies, standardized synthesis procedures, characterization methods, and reporting formats are therefore essential (Ghugare & Jadhav, 2026; Swaroop et al., 2026).

Other restrictions are the limited use of environmentally representative testing conditions. Most investigations use single-metal solutions, deionized water, and contaminant concentrations that are significantly greater than those found in natural waters. The ability of these materials to perform under conditions that are representative of the environment, such as in the presence of competing ions and mixed wastewater matrices, is therefore poorly understood, particularly in the context of environmentally relevant concentrations of trace contaminants. However, some recent studies have investigated their use in real wastewater systems, which represents an important development for practical validation (Peng et al., 2017; Wang & Guo, 2023; Deveci et al., 2025; Van Tran et al., 2025; Maity et al., 2026).

Another key challenge is nanosheet aggregation and its related hydraulic limitations. This aggregation can decrease the available adsorption sites and increase the pressure drop in treatment processes. A promising solution is

immobilization within macroporous structures such as cryogels, aerogels, and hydrogels. These designs enhance structural strength, thereby promoting efficient fluid flow, and material recovery (Sahraei et al., 2017; Pinelli et al., 2023; Mubarak et al., 2026).

Further investigation is needed to evaluate economic feasibility and production scalability. Utilizing biomass precursors and electrochemical techniques offers an attractive route toward more sustainable manufacturing, characterized by reduced reagent overhead and lower financial costs (Dautov et al., 2026; Chee et al., 2025; Swaroop et al., 2026). Scalable aerogel fabrication has already been demonstrated (Sharma et al., 2023). However, detailed techno-economic assessments and life-cycle analyses are still not widely available, limiting conclusions regarding the overall environmental and economic sustainability of graphene-like remediation technologies (Verma et al., 2025).

Managing the recovery and reuse of spent adsorbents presents critical hurdles for long-term operational sustainability. Secondary waste streams can be produced during acid-intensive desorption processes and may need additional treatment. Therefore, these factors should be taken into account when evaluating material reusability, overall process sustainability, and environmental performance (Ghugare et al., 2024).

Despite these constraints, green graphene-like nanocomposites hold significant promise as sustainable platforms for water treatment. Specifically, magnetically recoverable composites stabilized with biopolymers and synthesized via green reduction protocols appear uniquely positioned for imminent deployment. These materials feature reported adsorption capacities of up to several hundred mg/g for Pb(II), undergo facile magnetic separation from solution, and can be reused across multiple treatment cycles (Nasiri et al., 2018; Alboghbeish et al., 2022; Bhushan & Gupta, 2025; Mubarak et al., 2026).

Continuous-flow column studies, with breakthrough modeling, should be conducted in the future to evaluate performance under realistic operational conditions (Duong et al., 2025). Additionally, the establishment of standard sustainability measures to compare synthesis routes and design selective adsorbents is essential to recover valuable metals economically. Emerging work on neodymium recovery highlights the potential of graphene-like composites in this area (Al-Salem et al., 2024). Reductive detoxification processes coupled with adsorption are another research avenue of interest. Such approaches could improve the removal and stabilization of contaminants such as Cr(VI), thereby enhancing both treatment efficiency and environmental protection (Jibin et al., 2023; Karim & Uddin, 2026). Furthermore, the use of recovered waste as a synthesis feedstock and source of functional components for adsorbent fabrication would support the environmental and economic case for such materials within a circular production system. Continued progress in this direction may help establish green graphene-

like nanocomposites as sustainable and commercially viable technologies for advanced water remediation (Ghugare et al., 2024; Karim & Uddin, 2026).

10. Conclusion and Recommedation

Green synthesis is a well-established pathway for fabricating graphene-like adsorbents and is characterized by the use of less hazardous chemicals as compared to other synthesis methods. The method utilizes plant extracts and microbial or biomolecular reducing agents, waste-based biomass, and electrochemical exfoliation to obtain functional graphene-like materials.

By incorporating magnetic components, biopolymers, and selective chelating ligands, their adsorption efficiency, recovery, and reusability are further improved, leading to nanocomposites that can compete with traditional adsorbents for heavy metal removal.

The reported maximum adsorption capacities range from approximately 100 to 560 mg/g, with amine magnetic graphene oxide exhibiting lead adsorption capacities higher than 550 mg/g.

The uptake of metal is mediated by several interrelated processes such as electrostatic attraction, surface complexation, ion exchange, precipitation, and redox reactions. These processes are often modeled in terms of the Langmuir isotherm and pseudo-second-order kinetic behavior.

However, more effort needs to focus on standardized and reproducible green synthesis protocols, tests under realistic multi-contaminant scenarios and real wastewater matrices, long-term regeneration and re-use testing, and extensive techno-economic and ecotoxicological evaluations. Based on current evidence, the green synthesis of graphene-like nanocomposites is one of the most promising approaches to achieving sustainable and efficient remediation of heavy metals.

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