Comparative Evaluation of Graphene Oxide and Ethylenediamine-functionalized Graphene Oxide for Heavy Metal Removal from Contaminated Water

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***Abstract*—This study investigated the comparative adsorption performance of Graphene Oxide (GO) and Ethylenediamine-Functionalized Graphene Oxide (EDA/GO) for the removal of Arsenic (As), Copper (Cu), and Zinc (Zn) from contaminated water. Removal efficiency was systematically evaluated under acidic, neutral, and alkaline conditions (pH 3, 7, and 9), representing environmentally relevant pH ranges. GO was synthesized using the modified Hummers' method and subsequently functionalized with ethylenediamine to introduce nitrogen-containing coordination sites. Structural characterization using X-Ray Diffraction (XRD) and Fourier Transform Infrared Spectroscopy (FTIR) confirmed the successful oxidation of graphite to GO and the subsequent incorporation of amine functionalities into EDA/GO. Batch adsorption experiments conducted with an adsorbent dosage of 0.5 wt.% revealed distinct pH-dependent removal behavior. EDA/GO exhibited higher adsorption efficiencies across all conditions, achieving near-quantitative removal of Cu and Zn (~100% and 95%, respectively) under neutral to alkaline conditions, and enhanced As removal (~96%) even under acidic conditions. The improved adsorption performance of EDA/GO is due to increased surface disorder, amine-metal coordination, and enhanced charge tunability, which collectively facilitate electrostatic interactions and chelation. These findings demonstrate that EDA functionalization enhances the metal-binding affinity and kinetics of GO, supporting its potential applicability for multi-metal remediation in groundwater treatment systems.**

***Keywords*—graphene oxide, ethylenediamine, heavy metals, adsorption, remediation**

I. Introduction

Heavy metals, including Arsenic (As), Copper (Cu), and Zinc (Zn), are ubiquitous environmental pollutants that exist in diverse physicochemical forms, such as free ions and complexed species in air, soil, and water matrices. Their persistence, bioaccumulation, and toxicity pose severe risks to human health and ecological integrity [1, 2]. Sources of heavy metal contamination arise from both natural geogenic processes, such as the mobilization from geological strata, and anthropogenic activities, including mining, electroplating, metal processing, agriculture, and rapid industrialization [3, 4]. In Taiwan, intensive land use and urban-industrial expansion have accelerated the release of these heavy metals into groundwater, emphasizing the need for efficient and sustainable remediation strategies.

Among the available treatment technologies, adsorption has emerged as a highly effective approach due to its operational simplicity, cost-effectiveness, and superior removal efficiency for trace-level contaminants [5, 6]. In recent years, carbon-based nanomaterials, particularly Graphene Oxide (GO), have received significant attention as advanced adsorbents. GO possesses a large specific surface area and oxygen-containing functional groups (hydroxyl, carboxyl, and epoxy), which enhance its hydrophilicity and provide multiple active sites for metal-ion coordination [7, 8]. However, pristine GO experiences aggregation and exhibits limited selectivity for specific metal ions, diminishing its adsorption efficiency in complex aqueous environments.

Chemical modification of GO with nitrogen-rich ligands enhances its binding affinity and selectivity toward metal ions [9, 10]. Ethylenediamine (EDA), a bidentate amine compound, possesses two terminal $–NH_2$ groups capable of chelating metal ions through coordination interactions. Functionalization of GO with EDA (forming EDA/GO composites) not only introduces additional nitrogen-containing sites but also alters the surface charge and structural properties of GO, leading to improved adsorption capacity and stability under aqueous conditions [11].

Although EDA/GO is an effective adsorbent for heavy metal removal, most existing studies focus on single-metal systems, optimizing adsorption capacity, or idealized laboratory conditions. In contrast, this study emphasizes a comparative assessment of pristine GO and EDA/GO under multi-metal conditions, involving cationic (Cu^{2+}, Zn^{2+}) and oxyanionic (As species) contaminants across environmentally relevant pH ranges. By systematically examining pH-dependent removal behavior at concentrations representative of contaminated groundwater, we aim to provide insight into the differential adsorption mechanisms governing amine-functionalized GO surfaces. Rather than pursuing extensive material optimization, this study aims to elucidate how ethylenediamine functionalization alters surface chemistry and metal-binding interactions relative to unmodified GO, thereby advancing a mechanistic understanding relevant to practical groundwater remediation.

II. Literature Review

The removal of toxic heavy metals such as As, Cu, and Zn from water has been a primary focus of environmental research. Adsorption techniques are widely used for their simplicity, low cost, and effectiveness in removing trace contaminants [5, 6]. GO has gained attention as a promising

adsorbent due to its high surface area and oxygenated functional groups that facilitate metal ion binding [12–14].

Studies by Stankovich *et al.* [7] and Dreyer *et al.* [8] reported the chemical basis for GO reactivity, revealing that functional groups such as hydroxyl, epoxy, and carboxyl play dominant roles in metal-ion binding. Earlier research demonstrated a strong affinity of GO for transition metals; however, its tendency to aggregate in aqueous environments and its limited selectivity for specific ions significantly impaired its performance in complex systems [15, 16]. Additionally, the aggregation and partial reduction of GO during synthesis could compromise the accessibility of active adsorption sites, prompting investigations into chemical modification strategies to enhance its adsorption capacity and selectivity.

From the mid-2010s, research focused on surface modification of GO to improve selectivity and adsorption efficiency. Nitrogen-rich ligands emerged as particularly effective modifiers due to their strong electron-donating and coordination ability [17, 18]. Among these, Ethylenediamine (EDA) was widely used as a functionalizing agent. Zawisza *et al.* [19] demonstrated that EDA-modified GO significantly enhances the adsorption of Cu^{2+}, Zn^{2+}, and Ni^{2+} via amine-metal complexation. Later studies by Meng *et al.* [20] and White *et al.* [21] reported that EDA intercalation expands interlayer spacing and increases surface disorder, resulting in higher dispersion stability and greater accessibility of reactive sites.

Subsequent research focused on elucidating the structural and mechanistic aspects of EDA/GO composites. Characterization studies using XRD, FTIR, and SEM confirmed successful amine grafting and an increase in basal spacing [22, 23]. The introduction of N–H and C–N stretching bands in FTIR spectra indicated strong covalent and hydrogen bonding interactions between EDA and GO [24]. Adsorption behavior commonly followed pseudo-second-order kinetics and Langmuir isotherms, indicating monolayer chemisorption dominated by surface coordination. More recent studies have further demonstrated that functionalized GO materials exhibit enhanced removal efficiencies for a broad range of heavy metals and other aqueous contaminants under environmentally relevant conditions, highlighting the continued advancement and applicability of these materials in water treatment applications [25–28].

Despite considerable progress in the development of functionalized graphene oxide-based adsorbents, critical gaps persist in the literature. First, most existing studies have focused on single-metal systems under ideal laboratory conditions, which may not adequately reflect the complexity of natural groundwater containing coexisting ions and organic matter. Second, there is a limited understanding of the competitive adsorption behavior between cationic and anionic metals on bifunctionalized GO surfaces, particularly under environmentally relevant pH conditions. Future research should thus emphasize:

(1) Systematic multi-metal adsorption studies to understand synergistic and antagonistic interactions;
(2) Mechanistic modeling, integrating surface chemistry and thermodynamic data; and
(3) Scalable synthesis approaches that retain the structural integrity and functionality of EDA/GO composites for practical deployment in groundwater remediation.

III. Materials and Methods

A. Synthesis of Graphene Oxide

Graphene oxide was synthesized using the modified Hummers' oxidation method [29], which is known for its established reproducibility, well-characterized performance, and widespread use. While more recent greener synthesis approaches are available, the primary focus of this study was comparative adsorption behavior rather than synthesis optimization.

In the modified method (Fig. 1), commercial graphite powder (Sigma-Aldrich; particle size <20 μm; purity 100%) was gradually added to a mixture of sodium nitrate ($NaNO_3$) and concentrated sulfuric acid (H_2SO_4) under continuous stirring in an ice bath to control exothermic heat generation. Potassium permanganate ($KMnO_4$) was then introduced as the primary oxidizing agent to promote the intercalation and oxidation of graphite layers. The reaction was carried out at a controlled temperature and agitation to ensure uniform oxidation.

Upon completion, the reaction was quenched using hydrogen peroxide (H_2O_2) to decompose excess permanganate and terminate oxidation. The suspension was treated with dilute hydrochloric acid (HCl) to remove metal-ion residues. The resulting GO slurry was repeatedly washed with deionized (DI) water until the supernatant reached neutral pH, ensuring the complete removal of residual salts and acids. The purified GO was subsequently dried at 60 °C, ground to a fine powder, and stored in a desiccator prior to characterization and adsorption experiments.

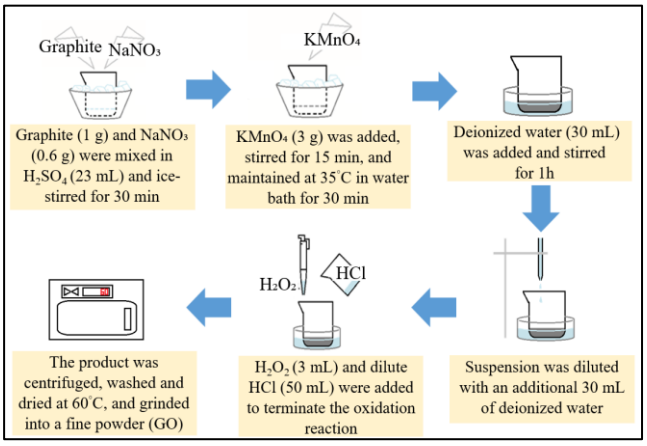

Fig. 1. Graphene oxide synthesis procedure.

B. Functionalization of Graphene Oxide with Ethylenediamine

To enhance the adsorption capacity and selectivity of GO for heavy metal ions, its surface was modified with EDA, a nitrogen-donor functionalizing agent. For the synthesis, GO powder was dispersed in anhydrous ethanol via ultrasonication to obtain a uniform suspension. A stoichiometric amount of EDA was added dropwise, and the mixture was heated and stirred continuously at a controlled temperature (80 °C) for 6 h to promote surface functionalization. The resulting product was filtered,

thoroughly washed with DI water and ethanol until neutral, dried at 60 °C, and ground to obtain the EDA/GO adsorbent.

C. Characterization

Commercial graphite powder, synthesized GO, and EDA/GO were characterized by XRD to evaluate structural modification and interlayer spacing changes. XRD measurements were carried out using a Rigaku MiniFlex 300 diffractometer equipped with Cu Kα radiation (λ = 1.5406 Å). Diffraction patterns were collected over a 2θ range of 10–90° to capture characteristic reflections associated with basal plane expansion and structural disorder. Data acquisition was conducted using a step width of 0.01° and a scanning rate of 10° min^{-1}, consistent with standard practices for phase identification and comparative analysis of graphene-based materials. Samples were ground to ensure homogeneity and mounted to obtain flat, uniform surfaces before analysis. The XRD results were used for comparative structural evaluation, including phase identification and an estimation of interlayer spacing, rather than detailed crystallographic refinement.

Similarly, FTIR spectroscopy was used for qualitative identification of oxygen- and nitrogen-containing functional groups and to verify the successful incorporation of amine functionalities. FTIR spectra were acquired using a Thermo Scientific Nicolet iS5 spectrometer over the wavenumber range of 4000–400 cm^{-1} at a spectral resolution of 4 cm^{-1}, with 32 scans averaged per sample to enhance signal quality. Before measurement, GO and EDA/GO powders were dried to remove residual moisture and prepared according to standard solid-sample handling procedures.

D. Preparation of Heavy Metal Solutions and Experimental Conditions

Standard stock solutions of As, Cu, and Zn (1000 mg/L; Merck Millipore) were prepared from analytical-grade reagents and subsequently diluted to working concentrations of 0.25, 5.0, and 25.0 mg/L, respectively. These concentrations were selected to be approximately 10 times the maximum permissible limits specified by Taiwan's groundwater quality standards, thereby simulating conditions representative of polluted scenarios. Differences among metal concentrations also reflect their distinct toxicity thresholds and regulatory standards.

To evaluate the comparative adsorption behavior of GO and EDA/GO, controlled batch experiments were conducted at three pH levels (3, 7, and 9) to represent acidic, neutral, and alkaline conditions. This pH range also reflects conditions commonly encountered in natural and contaminated groundwater systems across varying geochemical environments. pH was adjusted using dilute HCl (0.1 M) and NaOH (0.1 M) solutions. Batch experiments were conducted with an adsorbent dosage of 0.5 wt.%. Before the main experiments, a preliminary dosage screening was done at adsorbent loadings of 0.5 and 1.0 wt.% to evaluate the effect of dosage on removal performance. Based on these initial tests, 0.5 wt.% was identified as the optimized dosage, providing high removal efficiencies while minimizing material usage. Increasing the dosage to 1.0 wt.% did not demonstrate a proportionate improvement in removal performance, indicating partial saturation of available adsorption sites and reduced adsorption efficiency per unit mass of adsorbent.

After equilibrium mixing under specified conditions, the supernatant was filtered and analyzed for residual metal concentration using Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES) (Thermo Scientific, iCAP Q). Instrument calibration, quality control measures, and heavy metal quantification were performed in accordance with the procedures specified in Standard Method 3125 B [30].

IV. Results and Discussion

A. Characterization of Graphite, GO, and EDA/GO

The XRD pattern of graphite exhibited a sharp diffraction peak at $2\theta = 26°$, representing the (002) plane associated with a well-ordered crystalline structure. After oxidation, this peak disappeared and shifted to approximately $2\theta = 10°$, indicating an increase in interlayer spacing due to the introduction of oxygen-containing groups such as hydroxyl, epoxy, and carboxyl functionalities (Fig. 2). The interlayer spacing (d-spacing) of GO was determined using Bragg's law and found to be approximately 0.88 nm compared to 0.34 nm in the case of pristine graphite. This shift confirms successful oxidation and exfoliation of graphite into graphene oxide [22, 23]. The resulting GO exhibited reduced crystallinity, indicative of a layered, amorphous structure with disrupted stacking order.

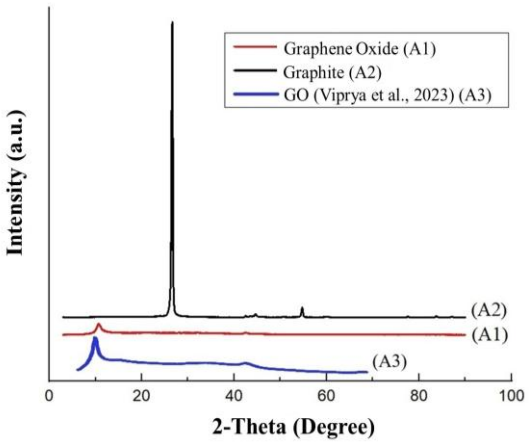

Fig. 2. XRD patterns of graphene oxide.

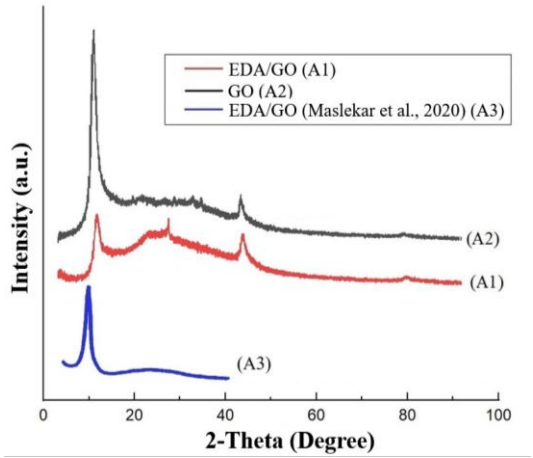

Fig. 3. XRD patterns of EDA/GO and GO.

Following surface modification with ethylenediamine (Fig. 3), the EDA/GO XRD pattern showed further broadening and a reduction in peak intensity. This change indicates structural disorder resulting from EDA intercalation between GO layers and the formation of hydrogen or covalent bonds with oxygen functional groups [20]. Such interlayer disruption expands the basal spacing, introducing flexibility and improving the accessibility of adsorption sites [24]. The resulting partial amorphization enhances the material's reactivity and is useful for adsorption processes, as it exposes more active sites for metal-ion coordination.

In addition, diffraction features observed near $2\theta = 40°$ in both GO and EDA/GO samples are attributed to residual graphitic domains or short-range ordering within partially oxidized graphene layers. The persistence of this feature suggests incomplete disruption of graphitic regions, a phenomenon commonly reported for GO-based materials.

Similarly, FTIR spectra (Fig. 4) further validated the structural transformation and functionalization of the materials. The GO spectrum exhibited characteristic absorption peaks corresponding to O–H stretching (3420 cm^{-1}), C=O stretching of carbonyl groups (1730 cm^{-1}), C=C skeletal vibrations (1630 cm^{-1}), and C–O stretching (1058 cm^{-1}). These peaks confirm the presence of hydroxyl, carboxyl, and epoxy groups on the GO surface. Such oxygen functionalities contribute to GO's hydrophilicity and provide reactive sites for heavy metal adsorption [31] via electrostatic interactions and surface complexation [32].

In contrast, the EDA/GO spectrum showed distinct modifications, including a broader absorption band at 3200–3500 cm^{-1}, attributed to overlapping O–H and N–H stretching vibrations, which confirm the successful introduction of amine groups from EDA. A new peak near 2900 cm^{-1} corresponded to C–H stretching of methylene groups in EDA, while peaks around 1600–1650 cm^{-1} indicated N–H bending and possible amide bond formation. Additional peaks in the 1350–1050 cm^{-1} range corresponded to C–N and C–O stretching, confirming chemical bonding between EDA and GO surface functionalities [19].

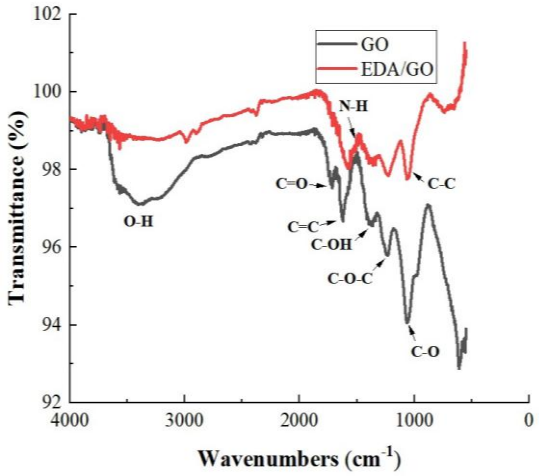

Fig. 4. FTIR images of GO and EDA/GO.

The appearance of nitrogen-related peaks in EDA/GO demonstrates successful surface modification, transforming the GO surface into a nitrogen-rich, multifunctional adsorbent. The introduced amine groups provide strong coordination sites for cationic heavy metals (e.g., Cu^{2+}, Zn^{2+}) via chelation, while enhancing the overall surface charge tunability and dispersion stability in aqueous systems.

B. Removal Efficiency of GO and EDA/GO

Fig. 5 shows the removal efficiency of GO and EDA/GO for As, Cu, and Zn as a function of pH and reaction time. Findings revealed distinct adsorption behaviors dependent on both pH and material surface chemistry.

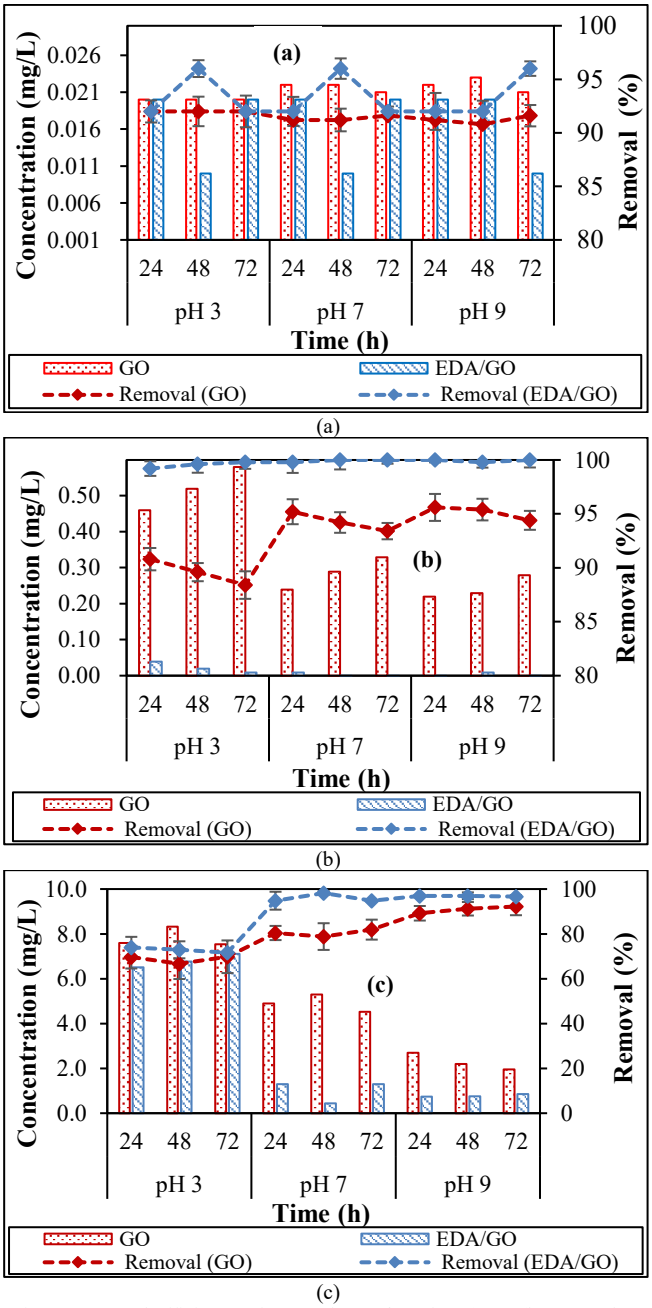

Fig. 5. Removal efficiency of GO and EDA/GO for (a) As, (b) Cu, and (c) Zn as a function of pH and reaction time.

Under acidic conditions (pH 3), both GO and EDA/GO demonstrated appreciable adsorption of As (Fig. 5(a)), but the rate and stability of removal were higher for EDA/GO. The As removal reached approximately 92% for GO and increased to about 96% with EDA/GO within 24 h, indicating the role of amine groups in enhanced electrostatic interactions even under acidic conditions. Protonation of amino sites in EDA/GO generates localized positive charges

that attract negatively charged arsenate or arsenite species, improving adsorption [33]. However, for Cu (Fig. 5b) and Zn (Fig. 5c), acidic conditions suppressed adsorption for both materials due to competition between H^+ and metal cations for active sites, as well as the absence of metal hydroxide precipitation. Nevertheless, EDA/GO exhibited faster Cu uptake than GO, suggesting a chelation advantage from amine-metal coordination.

At neutral pH (pH 7), the adsorption of Cu and Zn increased sharply for both adsorbents. GO achieved around 95% Cu (Fig. 5(b)) and 80% Zn (Fig. 5(c)) removal, while EDA/GO nearly reached 100% and 95%, respectively. The figures show rapid adsorption within the first 24 h, followed by gradual stabilization, indicating saturation of active sites. The enhanced efficiency of EDA/GO arises from its amine groups, which introduce additional coordination sites and improve the overall surface reactivity [28, 33]. For As (Fig. 5(a)), both adsorbents-maintained removal efficiencies above 90%, showing minimal pH dependence. However, EDA/GO exhibited a faster adsorption rate, consistent with its enhanced surface functionality. At both pH 3 and 7, As adsorption exhibited a non-linear time-dependent trend characterized by an initial increase followed by a gradual decrease. This behavior is due to a combination of factors, including progressive saturation of available adsorption sites, competitive interactions between arsenic species and coexisting metal ions, and possible partial desorption under dynamic surface charge conditions. At acidic to near-neutral pH, changes in arsenic speciation and surface protonation of GO and EDA/GO may influence electrostatic interactions and coordination mechanisms over time, leading to transient adsorption maxima. These effects, coupled with surface site rearrangement and competitive binding, can result in the observed decrease in apparent removal efficiency at longer contact times.

Under alkaline conditions (pH 9), both Cu and Zn exhibited the highest removal efficiencies for GO and EDA/GO. Fig. 5(b) indicates that Cu removal approached 96% with GO and nearly 100% with EDA/GO, while Zn removal (Fig. 5(c)) increased to 92% and 95%, respectively. These high efficiencies are due to the formation of $Cu(OH)_2$ and $Zn(OH)_2$ precipitates, which promote co-precipitation and surface fixation. As removal remained stable (Fig. 5(a)), EDA/GO maintained ~96% compared to ~92% for GO, likely due to protonated amine sites enhancing the attraction of arsenate species even under high-pH conditions.

Overall, pH strongly influenced the adsorption behavior by modulating both the adsorbent surface charge and metal speciation. At low pH, protonation of oxygen- and nitrogen-containing functional groups diminishes electrostatic attraction toward cationic metals while favoring interactions with anionic arsenic species. In contrast, neutral-to-alkaline conditions promote deprotonation of surface functional groups, increasing electrostatic attraction, coordination bonding, and hydroxide-mediated removal of Cu and Zn. Across all pH conditions examined, EDA/GO consistently outperformed pristine GO, with the greatest improvements observed for Cu and Zn, due to enhanced metal-ligand coordination and increased electron-donating capacity. Although arsenic removal was comparatively less sensitive to pH, EDA functionalization provided modest improvements in adsorption kinetics and stability. Collectively, these results demonstrate that neutral-to-mildly alkaline conditions (pH 7–9) are optimal for multi-metal removal, and that ethylenediamine functionalization significantly enhances the applicability of GO for groundwater remediation involving mixed-metal contaminants.

Table 1 presents a comparative summary of the adsorption results obtained in this study. The comparison includes activated carbon and representative functionalized graphene oxide-based adsorbents reported in the literature to contextualize the adsorption performance of EDA/GO.

Table 1. Comparison of EDA/GO with reported adsorbents

Adsorbent	Target Metal(s)	pH Ranges	Removal Efficiency (%)	References
Activated carbon (AC)	Pb^{2+}, Cu^{2+}, Zn^{2+}, Cd^{2+}, Cr(VI)	2–11	0–100	[34]
AC modified with fly ash	Cu^{2+}, Cd^{2+}	2–10	99.6–99.7	[35]
GO-NH_2	Cr(VI), Cu^{2+}, Pb^{2+}, Cd^{2+}	2–5	7.25–93.41	[28]
5-ATP-GO	Cd^{2+}, Hg^{2+}, As(III)	7.25–8.55	75.1–86.5	[10]
EDA/GO	As, Cu^{2+}, Zn^{2+}	3–9	80–100	**This study**

Activated carbon, a conventional adsorbent, generally exhibits moderate to high removal efficiencies; however, its performance is often strongly dependent on solution pH, metal speciation, and surface modification. In contrast, functionalized GO materials, particularly those incorporating nitrogen-containing groups, demonstrate enhanced adsorption capacities due to improved surface reactivity, metal-ligand coordination, and electrostatic interactions. As shown in Table 1, the EDA/GO synthesized in this study achieved removal efficiencies comparable to or exceeding those of reported functionalized adsorbents [10, 28] across a broad pH range (3–9) under multi-metal conditions involving both cationic and oxyanionic contaminants. This comparison highlights the advantage of EDA functionalization in achieving high, stable removal performance compared to conventional adsorbents. Notably, rather than emphasizing maximum adsorption capacity under idealized conditions, this study prioritizes the adsorption robustness and competitive behavior, which are critical for practical groundwater remediation.

V. Conclusion

Ethylenediamine functionalization significantly enhanced the adsorption efficiency and selectivity of graphene oxide toward As, Cu, and Zn across a wide pH range. Structural characterization confirmed successful amine grafting, which increased surface disorder, interlayer spacing, and the availability of reactive nitrogen coordination sites. These modifications strengthened metal-ligand interactions and charge tunability, leading to superior adsorption performance of EDA/GO compared to pristine GO. The adsorption behavior demonstrated that neutral to mildly alkaline

conditions (pH 7–9) were most favorable for the removal of Cu and Zn due to synergistic effects of electrostatic attraction, chelation, and hydroxide co-precipitation. In contrast, As removal remained effective even under acidic conditions, driven by protonated amine sites that electrostatically bind oxyanionic species.

This study provides experimental evidence that amine functionalization is an effective strategy to improve the structural and chemical reactivity of GO for simultaneous removal of cationic and anionic heavy metals. It should be noted, however, that the findings relating to characterization studies are based only on XRD and FTIR analyses. In future studies, advanced characterization techniques such as SEM/TEM, Raman spectroscopy, elemental analysis (XPS/EDX/CHNS), BET surface area analysis, and zeta potential measurements would be used to strengthen the structural and mechanistic interpretation of the material in a comprehensive materials-focused study. Future research should also focus on optimizing functionalization density, assessing regeneration and reuse potential, and scaling the synthesis for field applications to advance EDA/GO toward practical deployment in contaminated water treatment systems.

Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

SWCC: Conceptualization, Methodology, Resources, Writing—original draft, Visualization, Supervision, Project administration. YXH: Investigation, Data curation. SA: Formal analysis, Investigation, Methodology, Data curation, Writing—original draft, review and editing. All authors reviewed and approved the final version.

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