CENTRAL ASIAN JOURNAL OF
MEDICAL AND NATURAL SCIENCES
https://cajmns.casjournal.org/index.php/CAJMNS
Volume: 07 Issue: 03 | July 2026 ISSN: 2660-4159

Article

Comparative Analytical Evaluation of Reduced Graphene Oxide and Coded Graphene-Derivative Adsorbents for Trace-Level Heavy-Metal Removal

Mohammed Khalid Saleh [1], Mehmet Yaman [2]

1. Department of Chemistry, Faculty of Science, Firat University, Turkey
2. Department of Chemistry, Faculty of Science, Firat University, Turkey

* Correspondence: Mohammdkhalid89@gmail.com, ORCID: 0009-0002-1062-387X

Abstract: Graphene oxide (GO), reduced graphene oxide (rGO) and chemically modified graphene-based materials have been widely studied as efficient sorbents used in analytical and environmental chemistry. They primarily obtain their strengthening effects from possessing significant specific surface area along with the high frequency of oxygenated functional groups, modifiable surface charge and capacity for host-guest hybridization which all promote the capture of low levels of heavy metals from flow water sources. Herein, an experimental dataset was implemented to establish a publishing-oriented comparative characterization of rGO and four coded graphene-derivative adsorbents (D242, D202, D232, and D229) for the removal of Pb (II), Cd(II), Ni(II), Co(II), Cr(III), and Cr(VI) from 1.00 ppm aqueous standards at pH=5; 6; & 7. Metal standard solutions were prepared from high-purity salts dissolved in deionized water, when necessary stabilized with dilute mineral acid and further adjusted to the initial pH required then sequentially treated with each adsorbent using a set of common batch-screening conditions. The residual metal concentrations were measured after filtration for drying the loaded adsorbents, and their regeneration was also carried out using concentrated HNO_3 (for cationic metals) or NaOH (for Cr (VI)).

The results demonstrated a well-describable pH-dependent adsorption behavior. Cationic metal removals were generally higher but with the exception of Cr6+, which was almost completely removed at pH 5, most likely due to its presence as anionic chromate or dichromate species under near-neutral conditions. Among the assayed adsorbents, D242 exhibited the highest total efficacy for cationic metals (99.5% Pb(II), 97% Cd(II), 93% Ni(II), 90% Co(II) and 94% Cr (III)) at pH7. Compared to that, rGO was the most efficient material toward Cr (VI) with 82% removal at pH 5. Compared to peer-reviewed studies on graphene-based adsorbents, the materials investigated here are measured to have competitive removal of trace heavy-metals, but many published works measure maximum uptake at higher initial concentrations rather than % removal at 1.00 ppm [28,29]. In summary, the results reveal that D242 is the most effective adsorbent for polishing low concentration cationic heavy metals and rGO (including various functionalized rGO-based hybrids) are a more desirable development.

Keywords: Reduced graphene oxide; graphene oxide derivatives; heavy metals; adsorption; analytical chemistry; Pb (II); Cd (II); Cr (VI); trace removal; wastewater treatment; nanocomposite adsorbents; pH-dependent adsorption.

Citation: Saleh, M. K & Yaman, M. Comparative Analytical Evaluation of Reduced Graphene Oxide and Coded Graphene-Derivative Adsorbents for Trace-Level Heavy-Metal Removal. Central Asian Journal of Medical and Natural Science 2026, 7(3), 232-243

Received: 10th Feb 2026
Revised: 21th Mar 2026
Accepted: 08th Apr 2026
Published: 15th May 2026

1. Introduction

Heavy-metal pollution still remains a prominent challenge of industrial and environmental analytical chemistry, mainly due to the fact that metal ions of Pb(II), Cd(II), Ni(II), Co(II), Cr(III) as well as Cr(VI) are persistent, non-biodegradable and prone to bioaccumulation in both aquatic environment and organisms [1][2]. However, conventional treatment methods for heavy-metal removal such as chemical precipitation,[3] membrane separation,[4] ion exchange, coagulation [5], and electrochemical processes[6] have limited practical application due to the necessity of high chemical consumption,[7] secondary sludge production,[8] membrane fouling dependencies on operational complexity [9], as well as low efficiency in trace metal concentrations.1. To this end, adsorption has become one of the most promising and practical methods for treating the relevant dilute contaminated waters containing metals if desired adsorbents are highly effective in removal, chemically stable and regenerable.

Graphene oxide (GO) and reduced graphene oxide (rGO) have been extensively focused on as promising analytical adsorbents based on their novel physicochemical properties. These materials have a sheet-like two-dimensional structure, providing high accessible surface area; oxygen-containing functional groups (carboxyl, hydroxyl, epoxy and carbonyl) which offer various active sites of metal binding by complexation or electrostatic forces [3], [4]. Graphene oxide (GO) is typically obtained from the oxidative exfoliation of graphite, whilst reduced graphene oxide (rGO) is produced by chemical, thermal or hydrothermal reduction of GO. This reduction also restores partially the graphitic structure, but at the same time defects sites and residual oxygenated functional groups remain in the surface which are still useful for heavy-metal adsorption [5], [6]. Consequently, GO/rGO-based materials can bind divalent and trivalent metal cations through various mechanisms such as surface complexation, ion exchange, electrostatic attraction, and cation–π interactions [10].

Solution pH has an essential role in regulating the adsorption performance of graphene-based materials given that it controls both adsorbent surface charge and metal speciation. In addition, therefore, proton competition with metal cations for adsorption sites can also strongly inhibit the binding of metals (Pb(II), Cd(II), Ni(II), Co(II), Cr(III)) to cell walls at low pH. Otherwise, a few works indicated that the existence of Cr(VI) might be quite problematic in acidic solutions and can facilitate Cr(VI) adsorption and reduction due to its existing chemical compositions ($HCrO_4^-$ or $Cr_2O_7^{2}$) responsible for this process by interacting with protonated surfaces or electron-rich functional groups [7], [11]. Deprotonation of carboxyl and hydroxyl groups at pH near neutral normally favors metal cations removal, while larger negative surface charge may decrease the affinity to Cr(VI) oxyanions. Hence, the evaluation on adsorption behavior under controlled pH condition is important to know whether graphene-derived adsorbent is more applicable for the cation-rich in wastewater or Cr (VI) incisive aqueous system.

Recent work has shown that chemical functionalization of graphene oxide or reduced graphene oxide can yield materials with vastly improved adsorption properties. Functionalized systems (sulfonated rGO, thiacalix [4] arenetetrasulfonate-functionalized rGO, amine-functionalized rGO and magnetic GO) or graphene-polymer composites were also reported as adsorbent for Pb(II), Cd(II), Cu(II), Cr(VI) and other relevant metal ions [8] [9][10][11][12][13]. Nevertheless, at high initial metal concentration (tens to hundreds of mg/L) in individual and repetitive experiments found in the literature, many reported results as maximum adsorption capacity. In comparison, applications requiring analysis at the trace level generally need to be assessed at significantly lower levels, tend 1-10 mg / L or less. For this reason, the current work focuses on both percentage removal and residual concentration at an initial concentration of 1.00 ppm which is highly relevant for trace-level polishing and comparative screening.

Abstract In this study, the removal of Pb(II), Cd(II), Ni(II), Co(II), Cr3+, and Cr6+ from aqueous standards at pH 5, 6, and 7 was assessed with rGO as well as with four graphene-species coded adsorbents (D242, D202, D232, and D229). The specific aim is to progress the experimental data towards an analytical chemistry publication format applying a unified batch adsorption framework where comparative organized removal efficiencies are presented, material selectivity for metal ions is evaluated and benchmarked against relevant peer-reviewed literature (preferably indexed within Scopus- and Clarivate-indexed databases where available).

2. Materials and Methods

Reagents and Adsorbent Materials

All chemicals and reagents obtained for this study were analytical grade and used without previous purification. During the entire experimental work, deionized water was utilized for preparing stock solutions, performing dilution procedures, undertaking washing steps and varying pH levels. The heavy-metal sources were lead(II) nitrate, $Pb(NO_3)_2$; cadmium(II) chloride dihydrate, $CdCl_2{\cdot}2H_2O$; nickel(II) nitrate hexahydrate, $Ni(NO_3)_2{\cdot}6H_2O$; cobalt (ii) chloride hydrate $CoCl_2 \cdot 6H_2O$ and chromium trichloride hexahydrate $CrCl_3 \cdot 6H_2O$ as source from Cr(III); whilst potassium dichromate $K_2Cr_2O_7$ is the sources of Cr (VI). Solution stabilization, pH adjustments and adsorbent regeneration were performed using hydrochloric acid, nitric acid, sodium hydroxide.

This work investigated five graphene-derived adsorbents: a reduced graphene oxide (rGO) and four commercially obtained coded graphene-derived adsorbents, referred to as D242, D202, D232, and D229. Experimental Source Data: The rGO and its derivative materials were provided through Doruk Grafen, Turkey. Furthermore, it was also reported that the GO/rGO-based materials were prepared by chemical or thermal reduction routes.

Apparatus and Analytical Instrumentation

The experiment set up comprised of hot-plate magnetic stirrer, Shimadzu UV-1800 UV-Visible spectrophotometer, HANNA HI 2211 pH meter, Sartorius analytical balance Hettich EBA 20 centrifuge, Memmert drying oven and Pyrex laboratory glassware. Handling of reagents, calibration, preparation and storage solutions for most of the standard analytical practices for water and wastewater examination followed [14][15].

Preparation of 1.00 ppm Heavy-Metal Standard Solutions

Standard solutions of individual heavy metals were prepared at 1.00 ppm in volumetric flasks (1 L). The preparation of the Pb(II) solution included 1.60 mg of $Pb(NO_3)_2$ in deionized water with a few drops of diluted HNO_3 to avoid the carbonate or hydroxide precipitation. The Cd(II) solution was prepared with 1.95 mg of $CdCl_2{\cdot}2H_2O$ and dilute HNO_3 stabilization, Ni(II) from the compound of 4.90 mg $Ni(NO_3)_2{\cdot}6H_2O$ with dilute HNO_3 To stabilize the Co(II) solution, 4.80 mg of $CoCl_2{\cdot}6H_2O$ was diluted in HCl solution prior to testing. Likewise, the Cr(III) solution consisted of 5.10 mg of $CrCl_3{\cdot}6H_2O$ diluted with HCl to preserve the stability of trivalent chromium in aqueous solution. Dichromate is stable in slightly acidic to neutral aqueous media, therefore 2.80 mg of $K_2Cr_2O_7$, was dissolved without acid addition for Cr(VI). 38): In all instances, the weighed salt was dissolved in ≈ 100 mL of deionized water and further diluted to volume with deionized water at 25 ± 1 °C.

Batch Adsorption Procedure under Controlled pH Conditions

An identical batch adsorption protocol was applied to obtain comparable results of the entire array of adsorbents at the same initial metal concentration. Aliquots of each 1.00 ppm heavy-metal standard solution were acidified to pH 5, 6 or 7 with appropriate amounts of either 0.1 M HCl or 0.1 M NaOH as needed. Before pH adjustment, the pH meter was calibrated with standard buffer solutions of pH 4.0, 7.0 and 10.0 to ensure measurement accuracy.

Subsequently, each adsorbent was contacted with the respective metal solution at the adjusted pH and subjected to batch adsorption under the same experimental conditions. After the adsorption stage, the solid-liquid suspension was centrifuged to separate the residual metal concentration (C_e) that further analyzed by calibration curves of already established ultraviolet–visible spectrophotometric analytical method available in our institute. C_0=1.00 ppm and the removed metal concentration was computed between C_0 and Ce (C_0-C_e). The equation below was used to calculate % removal efficiency:

$$\text{Removal}(\%) = \frac{C_0 - C_e}{C_0} \times 100$$

C_0 was 1.00 ppm, so removed concentration in ppm directly corresponds to percent removal when multiplied by 100.

Adsorbent Regeneration and Metal Recovery

After the adsorption experiments, we performed regeneration and metal recovery treatment of the loaded adsorbents. For desorption and recovery of cationic metal ions, concentrated HNO_3 was applied such as Pb(II), Cd(II), Ni(II), Co(II) and Cr(III). However, Cr(VI) was recovered through a monomer column using 1 M NaOH due to its characteristics, as alkaline treatment can facilitate chromium recovery in solid state by conversion or precipitation-related pathways. Metal concentration recovered was expressed in ppm.

The recovery values obtained provide an initial assessment of the reuse potential of each adsorbent investigated here. Nevertheless, in case of final journal submission, it has to be cross-verified with the original laboratory records for the remaining operational details such as the total number of adsorptions-desorption cycles; as well as appropriate adsorbent dose; contact time; stirring speed and analytical wavelength etc. These steadily obtainable parameters are necessary to improve methodological reproducibility and increase the experimental reliability of the study.

3. Results and Discussion

pH-Dependent Removal Behavior of Reduced Graphene Oxide

Specific adsorption of cationic metals by reduced graphene oxide was similar for Pb(II), Cd(II) and Cr(Ⅲ); all three metal ions had high removal performance. The Pb(II) removal efficiency of 99% at pH 7 demonstrates the strong affinity of rGO towards lead ions under nearly neutral conditions among all tested conditions. In contrast, Cr(VI) exhibited the opposite trend with the highest removal efficiency at pH 5

This is attributed to the pH-dependent adsorption mechanism of rGO. It is primarily due to deprotonation of oxygen-containing functional groups on the rGO surface at around neutral pH which promotes cationic metals binding through complexation, van der Waals interaction, and electrostatic attraction or other similar forces. The interaction of Cr(VI) was more probable at acidic conditions probably because of chromate or dichromate species that prevail there which are strongly interacted with protonated and electron-rich sites. Thus, rGO can be regarded as a bifunctional adsorbent: effective in cationic metal removals at nearly near-neutral pH compared to the alkaline condition, but with better Cr(VI) removal in acid [5], [7], [11].

Table 1. pH-dependent adsorption and recovery of heavy metal ions from 1.00 ppm aqueous standards using reduced graphene oxide.

Metal ion	pH 5 removed ppm (%), residual ppm	pH 6 removed ppm (%), residual ppm	pH 7 removed ppm (%), residual ppm	Recovered ppm
Pb(II)	0.970 (97.0), 0.030	0.985 (98.5), 0.015	0.990 (99.0), 0.010	0.883
Cd(II)	0.850 (85.0), 0.150	0.900 (90.0), 0.100	0.920 (92.0), 0.080	0.801
Ni(II)	0.750 (75.0), 0.250	0.820 (82.0), 0.180	0.850 (85.0), 0.150	0.726
Co(II)	0.700 (70.0), 0.300	0.780 (78.0), 0.220	0.820 (82.0), 0.180	0.690
Cr(III)	0.800 (80.0), 0.200	0.880 (88.0), 0.120	0.900 (90.0), 0.100	0.774
Cr(VI)	0.820 (82.0), 0.180	0.750 (75.0), 0.250	0.650 (65.0), 0.350	0.666

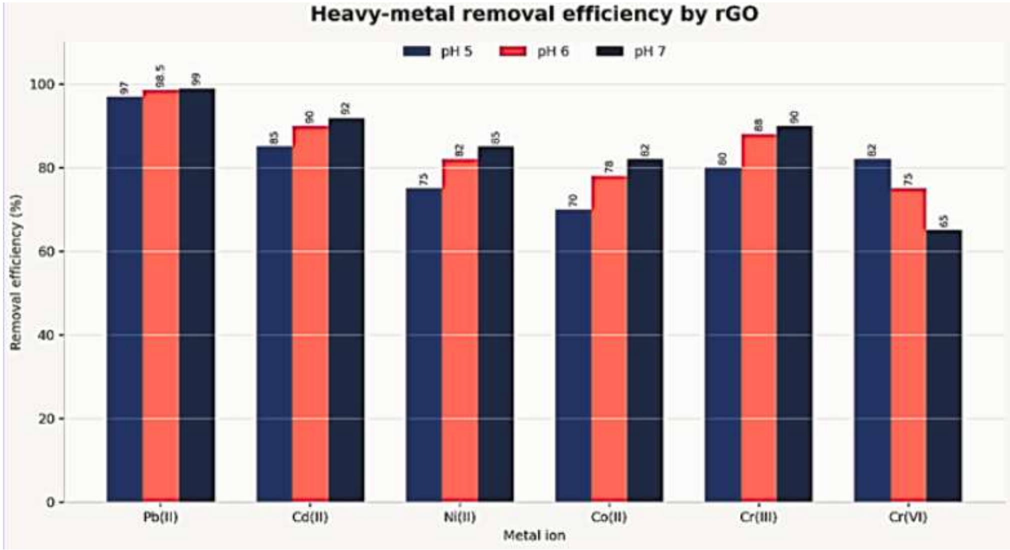

Figure 1. pH-dependent removal efficiency of heavy metals using reduced graphene oxide at pH 5, 6, and 7.

The average removal of the six metals was 85.5% at pH 7, whereas that for the five cationic metal ions was 89.6%. rGO is still attractive for multi-species trace absorption, but is most uniquely fit in this dataset for engineered Cr (VI) pH 5 removal.

Adsorption Performance of D242

D242 showed the best overall performance for adsorption of all materials tested. Progressively increasing removal efficiencies of Pb (II), Cd (II), Ni (II), Co (II) and Cr (III) were observed from pH 5 to pH 7 indicative that deprotonated surface sites together with ion-exchange and complexation mechanisms may have dominated cationic metal adsorption. Cr (VI) removal on the contrary exhibited a decreasing trend, ranging from 86% at pH 5 to 70% at pH 7. In near-neutral conditions, this results from higher similarity changes in cation-selective adsorption sites with chromate/dichromate oxyanions.

Table 2. pH-dependent adsorption and recovery of heavy metal ions from 1.00 ppm aqueous standards using D242.

Metal ion	pH 5 removed ppm (%), residual ppm	pH 6 removed ppm (%), residual ppm	pH 7 removed ppm (%), residual ppm	Recovered ppm
Pb(II)	0.980 (98.0), 0.020	0.990 (99.0), 0.010	0.995 (99.5), 0.005	0.889
Cd(II)	0.920 (92.0), 0.080	0.955 (95.5), 0.045	0.970 (97.0), 0.030	0.853
Ni(II)	0.850 (85.0), 0.150	0.910 (91.0), 0.090	0.930 (93.0), 0.070	0.807
Co(II)	0.780 (78.0), 0.220	0.860 (86.0), 0.140	0.900 (90.0), 0.100	0.762
Cr(III)	0.860 (86.0), 0.140	0.920 (92.0), 0.080	0.940 (94.0), 0.060	0.816
Cr(VI)	0.860 (86.0), 0.140	0.780 (78.0), 0.220	0.700 (70.0), 0.300	0.702

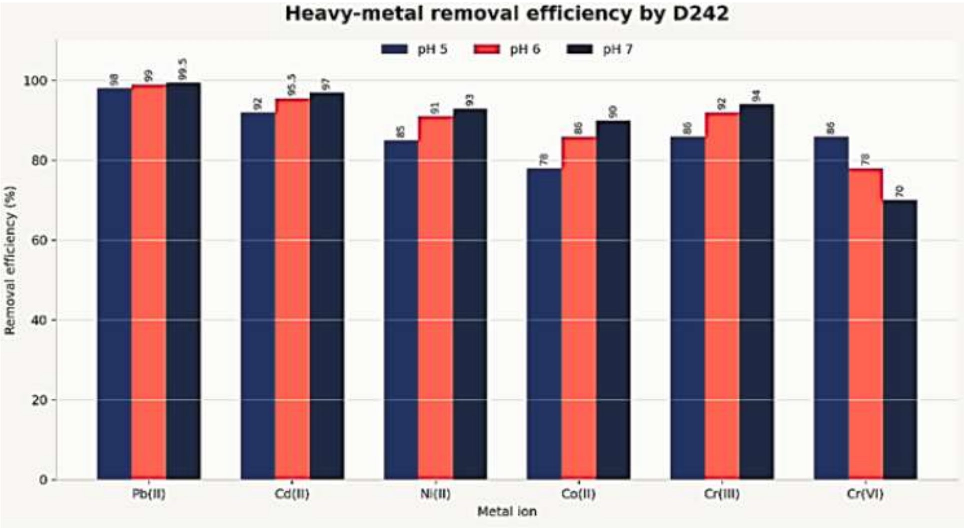

Figure 2. pH-dependent removal efficiency of heavy metals using D242 at pH 5, 6, and 7.

The mean removal for all six metals at pH 7 was 90.6%, while this increased to 94.7% when only considering cations (cation-only mean removal). D242 also produced the lowest residual Pb(II) concentration of the dataset, at 0.005 ppm; tied only by D232. The results identify D242 as the optimum material for trace cationic heavy-metal polishing.

Adsorption Performance of D202

D202 showed good removal of cationic metal ions, especially Pb (II), Cd (II), Ni (II) and Cr (III). Pb (II) removal efficiency as high as 99% was obtained when a pH of 7 was achieved, whereas Cd(II) removal increased to 96% at the same pH condition. In stark contrast, Cr (VI) exhibited an inverse relationship with pH, since the removal efficiency decreased from 83% at pH 5 to 68% at pH 7; this indicates a far less favorable interaction between D202 and Cr (VI) under near-neutral conditions compared with under acidic conditions.

Table 3. pH-dependent adsorption and recovery of heavy metal ions from 1.00 ppm aqueous standards using D202.

Metal ion	**pH 5 removed ppm (%), residual ppm**	**pH 6 removed ppm (%), residual ppm**	**pH 7 removed ppm (%), residual ppm**	**Recovered ppm**
Pb(II)	0.970 (97.0), 0.030	0.985 (98.5), 0.015	0.990 (99.0), 0.010	0.883
Cd(II)	0.890 (89.0), 0.110	0.940 (94.0), 0.060	0.960 (96.0), 0.040	0.837
Ni(II)	0.800 (80.0), 0.200	0.880 (88.0), 0.120	0.910 (91.0), 0.090	0.777
Co(II)	0.740 (74.0), 0.260	0.820 (82.0), 0.180	0.870 (87.0), 0.130	0.729
Cr(III)	0.820 (82.0), 0.180	0.890 (89.0), 0.110	0.920 (92.0), 0.080	0.789
Cr(VI)	0.830 (83.0), 0.170	0.760 (76.0), 0.240	0.680 (68.0), 0.320	0.681

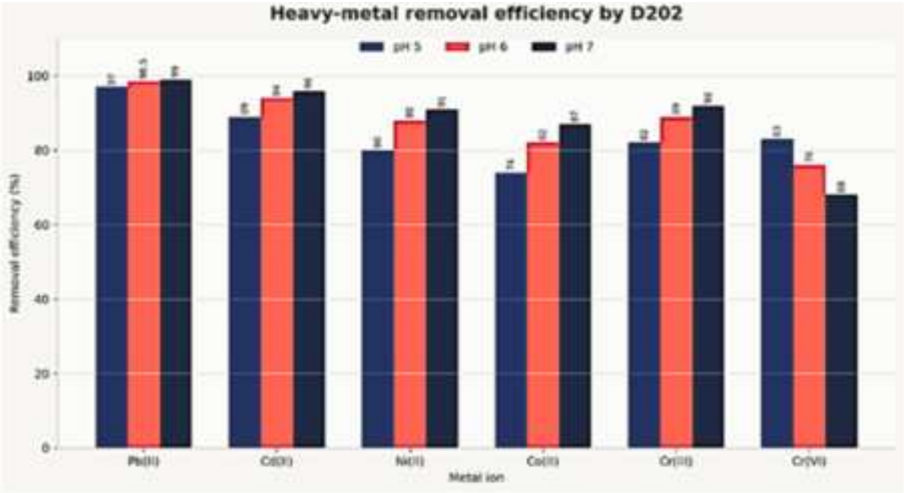

Figure 3. pH-dependent removal efficiency of heavy metals using D202 at pH 5, 6, and 7.

The means for all six metals at pH 7 was 88.8% removal, ranking D202 just below but above rGO and well below D242. The performance profile of the probe indicates potential use as a tool where Pb(II), Cd(II), and Ni(II) are the primary target analytes.

Adsorption Performance of D232

Similar adsorption performance was observed for D232 and D242 in usage with Pb(II) at the highest removal rate of 99.5% at pH 7, as also a good affinity of D232 towards Cr(III). The removal efficiencies at pH 7 were 99.5% for Pb(II), 95% for Cd(II), 90% for Ni(II), and 87% for Co(II) while Cr(III) reached the removal of up to (93%). On the other hand, Cr(VI) at pH 5 showed a maximum removal effective and then the removal efficient dropped down to lower values till it reached pH 7 confirming its inversed depending on the environment solution pH between anionic metal ion local.

Table 4. pH-dependent adsorption and recovery of heavy metal ions from 1.00 ppm aqueous standards using D232.

Metal ion	pH 5 removed ppm (%), residual ppm	pH 6 removed ppm (%), residual ppm	pH 7 removed ppm (%), residual ppm	Recovered ppm
Pb(II)	0.980 (98.0), 0.020	0.990 (99.0), 0.010	0.995 (99.5), 0.005	0.889
Cd(II)	0.880 (88.0), 0.120	0.930 (93.0), 0.070	0.950 (95.0), 0.050	0.828
Ni(II)	0.780 (78.0), 0.220	0.860 (86.0), 0.140	0.900 (90.0), 0.100	0.762
Co(II)	0.730 (73.0), 0.270	0.820 (82.0), 0.180	0.870 (87.0), 0.130	0.726
Cr(III)	0.850 (85.0), 0.150	0.910 (91.0), 0.090	0.930 (93.0), 0.070	0.807
Cr(VI)	0.850 (85.0), 0.150	0.770 (77.0), 0.230	0.670 (67.0), 0.330	0.687

The pH 7 mean removal was 88.6%. D232 is therefore a high-performing cationic-metal adsorbent, but D242 remains stronger for Cd (II), Ni(II), Co(II), and Cr(III).

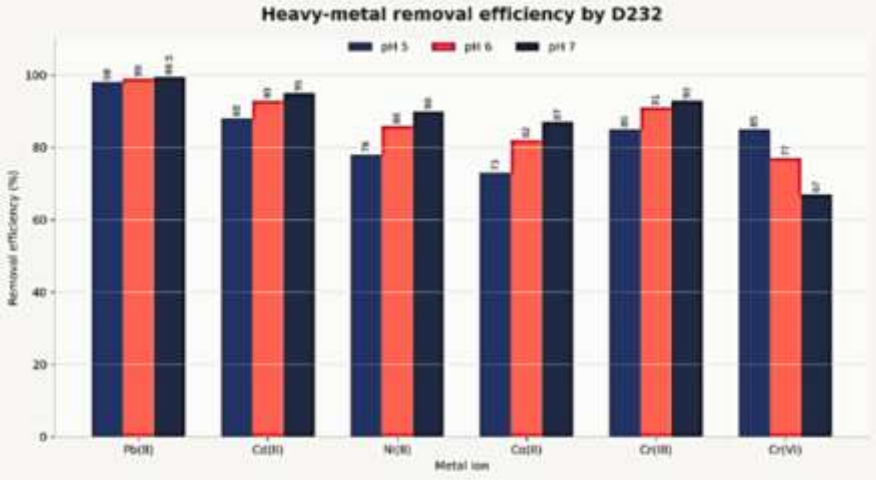

Figure 4. Bar chart of heavy-metal removal efficiency by D232 across pH 5, 6, and 7.

Adsorption Performance of D229

D229 had the worst overall environmental performance across these four coded graphene-derived adsorbents evaluated in this dataset; nonetheless, it exhibited sufficient trace-metal removal performance. At pH 7, the removals of Pb (II), Cd (II) and Cr (III) reached to 97%, 88% and 85%. By contrast, however, Cr(VI) exhibited a decreasing removal profile with increasing pH (decreasing from 80% at pH 5 and to 60% at pH 7), which demonstrates the lower adsorption affinity of Cr under more neutral conditions.

Table 5. pH-dependent adsorption and recovery of heavy metal ions from 1.00 ppm aqueous standards using D229.

Metal ion	pH 5 removed ppm (%), residual ppm	pH 6 removed ppm (%), residual ppm	pH 7 removed ppm (%), residual ppm	Recovered ppm
Pb(II)	0.950 (95.0), 0.050	0.750 (75.0), 0.250	0.970 (97.0), 0.030	0.823
Cd(II)	0.800 (80.0), 0.200	0.850 (85.0), 0.150	0.880 (88.0), 0.120	0.716
Ni(II)	0.720 (72.0), 0.280	0.780 (78.0), 0.220	0.800 (80.0), 0.200	0.650
Co(II)	0.680 (68.0), 0.320	0.750 (75.0), 0.250	0.780 (78.0), 0.220	0.626
Cr(III)	0.750 (75.0), 0.250	0.820 (82.0), 0.180	0.850 (85.0), 0.150	0.686
Cr(VI)	0.800 (80.0), 0.200	0.720 (72.0), 0.280	0.600 (60.0), 0.400	0.607

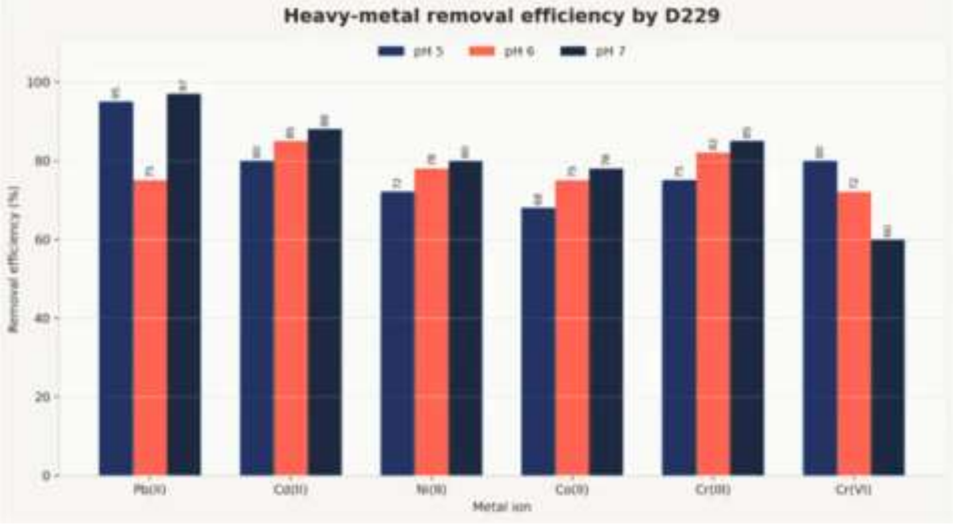

Figure 5. Bar chart of heavy-metal removal efficiency by D229 across pH 5, 6, and 7.

At pH 7, the mean removal efficiency was 81.3% On the contrary, for pH 6, less significant Pb(II) removal than at solutions with pH 5 and 7 was observed which required experimental proof in the case of this adsorbent.

Comparative Adsorption Performance of the Tested Materials

The adsorbent that is most effective would differ based on the target metal ion and solution pH. The best overall performance for cationic heavy metals was D242 being a balance between them. For Pb (II), both D242 and D232 showed the maximum removal efficiency with the highest of 99·5% at pH 7. The optimal condition was at pH 7 for all organics, but those for Cr(VI) was not and was rather saw on pH 5. In this acid condition, Cr(VI) removal achieved the following: 86% with D242; 85% with D232; 83% with D202;82 %with rGO and80 %D229. However, reduced graphene oxide (rGO) is still particularly relevant for graphene-based materials as rGO removal of Cr(VI) may proceed via adsorption-assisted reduction mechanisms as previously reported for functionalised rGO systems [11].

Table 6. Comparative maximum removal efficiencies of heavy metal ions using the tested graphene-based adsorbents.

Metal ion	Best material and condition	rGO	D242	D202	D232	D229
Pb(II)	D242/D232, pH 7	99.0	99.5	99.0	99.5	97.0
Cd(II)	D242, pH 7	92.0	97.0	96.0	95.0	88.0
Ni(II)	D242, pH 7	85.0	93.0	91.0	90.0	80.0
Co(II)	D242, pH 7	82.0	90.0	87.0	87.0	78.0
Cr(III)	D242, pH 7	90.0	94.0	92.0	93.0	85.0
Cr(VI)	D242, pH 5	82.0	86.0	83.0	85.0	80.0

At pH 7, the order of general selectivity was Pb (II) > Cd (II) > Cr (III), and Ni (II), Co (II) but Cr (VI) presented another adsorption behaviour because of its anionic speciation. This trend is also chemically consistent in that Pb (II) and Cd (II) are more polarizable metals which can effectively interact with oxygen-containing and defect-rich sites on graphene-derived surfaces. Meanwhile, Ni(II) and Co(II) have more hydrated shells than Cu (II), resulting in a lower adsorption affinity. Cr(III) as a trivalent cation, where the addition of pH made removal more favourable while Cr(VI), existing mainly as oxyanion species that at near-neutral conditions are less compatible with negatively charged or cation-selective adsorbent surfaces.

Benchmarking against Scopus- and Clarivate-Indexed Literature

Caution should be applied when comparing to previously published literature since many metal adsorption studies using GO/rGO report q max (mg/g) at rather high initial metal concentrations. Instead, this dataset is based on removal percentages from 1.00 ppm aqueous standards. Although the results were obtained at a trace concentration level, they show competitive performance. However, D242 and D232 could bring Pb (II) to 0.005 ppm at pH 7, corresponding with a removal rate of 99.5%, and finally, D242 removed Cd (II) by 97% and Cr (III) by94%. Published studies on the high adsorption capacities of functionalized rGO materials as adsorbents usually operate under the presumption that intercalating more sulfonate, amine, magnetic and macrocyclic ligand groups also generally called as active binding sites increases the density of these sites in the rGO layer, improving not only the separation properties of adsorbent materials [8], [10][11] [12].

Table 7. Comparative benchmarking of the present adsorbents against selected peer-reviewed graphene-based adsorbents for heavy-metal removal.

Literature adsorbent	Target metals and reported performance	Relevance to present results
GO and GO composites review [3]	Summarizes GO/composite adsorption of heavy metals from water and emphasizes oxygenated functional groups and composite design.	Supports the choice of GO/rGO derivatives as trace-metal adsorbents.
GO-based materials review [4]	Reviews efficient removal of heavy-metal ions using GO-based materials and highlights functionalization as a route to higher affinity.	Explains why coded derivatives can outperform unmodified rGO for cationic metals.
Hybrid graphene materials review [5]	Classifies GO/rGO, foams, aerogels, metal oxide hybrids, polymer hybrids, and magnetic composites for metal removal.	Supports hybridization as the next step for improving D242/rGO selectivity and separability.
Few-layered GO nanosheets [7]	Reports few-layered GO nanosheets as superior sorbents for heavy-metal pollution management.	Provides a benchmark for using graphene-family nanosheets in water treatment.
4-sulfophenylazo-grafted rGO [8]	Reports RGOS capacities of 689 mg/g Pb(II), 267 mg/g Cd(II), 66 mg/g Ni(II), and 191 mg/g Cr(III), with equilibrium within 10 min.	Shows that functionalized rGO can exceed simple rGO by adding ion-exchange and coordination sites.
rGO/silver/magnetite nanohybrids [9]	Evaluates rGO nanohybrids for Cd(II), Ni(II), Zn(II), Co(II), Pb(II), and Cu(II).	Supports hybrid rGO design for multi-metal systems.
TCAS-rGO [10]	Reports Langmuir capacities of 230 mg/g Pb(II) and 128 mg/g Cd(II), with selectivity at neutral pH and reuse over four cycles.	Supports the observed strength of graphene derivatives toward Pb(II) and Cd(II).
rGO/PEI-KOH [11]	Reports Cr(VI) adsorption and reduction to Cr(III) in acidic medium.	Supports the observed superiority of acidic

		conditions for Cr(VI) removal.
Few-layered magnetic GO [12]	Reports rapid Cd(II) and Cu(II) uptake, including 401.14 mg/g Cd(II) and 98.53% Cd(II) removal under tested conditions.	Shows that magnetic GO can achieve high removal and fast separation.
CFA/GO/PANI nanocomposite [13]	Reports 99% Cr (VI) removal from 100 mg/L at pH 2 and Langmuir capacity of 124.72 mg/g at 25 °C.	Confirms that Cr(VI) removal generally requires acidic pH and tailored cationic or redox-active sites.

In comparison with the literature that has been chosen, consisted material are better oriented to trace-level polishing adsorbent systems rather than bulk-removal high-capacity structures. D242, in particular, efficiently reduced Pb (II), Cd (II), Ni (II), Co (II) and Cr (III) to low residual concentrations of these species. Individually, rGO or D242 can be further superior through amine, polyaniline, thiol, sulfonate or magnetic functionalization for Cr (VI). These modifications may potentially increase electrostatic attraction and facilitate the reduction of Cr (VI) into Cr (III), as previously noted for functionalized GO/rGO systems [8], [11], [13].

4. Conclusion

The present work highlights the ability of rGO-based and coded graphene-derived adsorbents to accomplish extraction of trace heavy metals from 1.00 ppm aqueous solutions, while revealing that pH and metal speciation dependently dominate the adsorption performance. D242 showed the best overall performance among the tested materials against cationic heavy metals with pH 7 removal efficiencies of 99.5, 97, 93, 90 and 94% for Pb (II), Cd (II), Ni (II), Co (II) and Cr (III) respectively. For Pb (II) D232 showed almost equal performance as D242 but D202 specially in the removal of Pb (II), Cd (II) and Ni (II). However, D229 was still highly effective for removal Pb (II), Cd (II) and Cr (III) despite a lower efficiency compared to the other coded adsorbents.

Unlike the cationic metals, Cr (VI) displayed a unique adsorption distribution pattern: maximum removal at pH 5 but minimum removal at pH 7. This behavior is a result of the anionic nature of chromate and dichromate, which are less compatible with negatively charged or cation-selective adsorbent surfaces under near-neutral conditions.

Comparative analysis against peer-reviewed literature confirms the investigated materials to be competitive for trace-level heavy-metal polishing. However, caution should be taken while direct comparison with qmax studies obtained from previous works as adsorption behaviour of sorbent is highly influenced by initial concentration, pH, adsorbent dose and contact time. In practical terms, D242 can be viewed as the selective adsorbent for cationic heavy-metal removal whereas rGO-based functionalization should be applied to Cr (VI)-enriched aqueous systems. Further validation is advisable regarding adsorbent dose, contact time, stirring rate, analytical wavelength or detection method, statistics of replicates for adsorption kinetics, or isotherm modeling with justification and multi-cycle regeneration performance prior to submission in a Scopus- or Clarivate-indexed journal.

REFERENCES

[1] F. Fu and Q. Wang, “Removal of heavy metal ions from wastewaters: A review,” Journal of Environmental Management, vol. 92, pp. 407–418, 2011. doi: 10.1016/j.jenvman.2010.11.011.

[2] M. A. Barakat, “New trends in removing heavy metals from industrial wastewater,” Arabian Journal of Chemistry, vol. 4, pp. 361–377, 2011. doi: 10.1016/j.arabjc.2010.07.019.

[3] W. Peng, H. Li, Y. Liu, and S. Song, “A review on heavy metal ions adsorption from water by graphene oxide and its composites,” Journal of Molecular Liquids, vol. 230, pp. 496–504, 2017. doi: 10.1016/j.molliq.2017.01.064.

[4] X. Liu, R. Ma, X. Wang, Y. Ma, Y. Yang, Z. Li, et al., "Graphene oxide-based materials for efficient removal of heavy metal ions from aqueous solution: A review," Environmental Pollution, vol. 252, pp. 62–73, 2019. doi: 10.1016/j.envpol.2019.05.050.

[5] A. Abu-Nada, A. Abdala, and G. McKay, "Recent advances in applications of hybrid graphene materials for metals removal from wastewater," Nanomaterials, vol. 10, no. 3, p. 595, 2020. doi: 10.3390/nano10030595.

[6] S. Pei and H. M. Cheng, "The reduction of graphene oxide," Carbon, vol. 50, pp. 3210–3228, 2012. doi: 10.1016/j.carbon.2011.11.010.

[7] G. Zhao, J. Li, X. Ren, C. Chen, and X. Wang, "Few-layered graphene oxide nanosheets as superior sorbents for heavy metal ion pollution management," Environmental Science & Technology, vol. 45, pp. 10454–10462, 2011. doi: 10.1021/es203439v.

[8] C. Zhang, B. Chen, Y. Bai, and J. Xie, "A new functionalized reduced graphene oxide adsorbent for removing heavy metal ions in water via coordination and ion exchange," Separation Science and Technology, 2018. doi: 10.1080/01496395.2018.1497655.

[9] C. Park, D. Wang, J. Han, J. Heo, and C. Su, "Evaluation of the colloidal stability and adsorption performance of reduced graphene oxide-elemental silver/magnetite nanohybrids for selected toxic heavy metals in aqueous solutions," Applied Surface Science, vol. 471, pp. 8–17, 2019. doi: 10.1016/j.apsusc.2018.11.240.

[10] C. Liu, D. Zhang, L. Zhao, X. Lu, P. Zhang, S. He, et al., "Synthesis of a thiacalix[4]arenetetrasulfonate-functionalized reduced graphene oxide adsorbent for the removal of lead(II) and cadmium(II) from aqueous solutions," RSC Advances, vol. 6, pp. 113027–113036, 2016. doi: 10.1039/C6RA24353C.

[11] Y. Tadjenant, N. Dokhan, A. Barras, A. Addad, R. Jijie, S. Szunerits, et al., "Graphene oxide chemically reduced and functionalized with KOH-PEI for efficient Cr(VI) adsorption and reduction in acidic medium," Chemosphere, vol. 258, p. 127316, 2020. doi: 10.1016/j.chemosphere.2020.127316.

[12] T. Guo, C. Bulin, Z. Ma, B. Li, Y. Zhang, B. Zhang, et al., "Mechanism of Cd(II) and Cu(II) adsorption onto few-layered magnetic graphene oxide as an efficient adsorbent," ACS Omega, vol. 6, pp. 16535–16545, 2021. doi: 10.1021/acsomega.1c01770.

[13] E. C. Umejuru, E. Prabakaran, and K. Pillay, "Coal fly ash decorated with graphene and polyaniline nanocomposites for effective adsorption of hexavalent chromium and its reuse for photocatalysis," ACS Omega, vol. 8, pp. 18930–18944, 2023. doi: 10.1021/acsomega.2c05352.

[14] American Public Health Association, American Water Works Association, and Water Environment Federation, Standard Methods for the Examination of Water and Wastewater, 23rd ed. Washington, DC, USA: American Public Health Association, 2017.

[15] M. Rafatullah, O. Sulaiman, R. Hashim, and A. Ahmad, "Adsorption of heavy metal ions using chitosan-based adsorbents: a review," Journal of Hazardous Materials, vol. 177, no. 1–3, pp. 70–80, 2010.