

# Simultaneous adsorption of heavy metals from aqueous matrices by nanocomposites: A first systematic review of the evidence

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## Abstract

**Background:** Nanocomposites have received remarkable attention as effective adsorbents for removal of coexisting pollutants over the last decades. The presence of heavy metals (HMs) in wastewater has caused a global health concern. Therefore, the aim of this study was to review the most relevant publications reporting the use of nanostructures to simultaneous adsorption of HMs in mixed aqueous systems.

**Methods:** In this systematic review, 9 studies were included through a systematic search in the three databases (ISI, Scopus, and PubMed) during 1990-2021. The optimal value of simultaneous adsorption parameters such as initial concentration, contact time, adsorbent dosage, and pH was discussed.

**Results:** Findings indicate that the Langmuir and Freundlich models and the pseudo-second-order kinetic model have been widely used and the most popular models to describe the equilibrium of HMs by nanoadsorbents. This study confirmed that the simultaneous removal rate of HMs decreased with an increase in pH value. It was found that the major mechanisms of HMs adsorption onto nanostructures were electrostatic interactions and precipitation.

**Conclusion:** Nanocomposites have remarkable adsorption performance for HMs with the highest adsorption capacity ( $q_e$ (mg/g)).

**Keywords:** Adsorption, Wastewater, Heavy metals, Nanocomposites, Kinetics

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## Introduction

Heavy metals (HMs) are the chemical elements with a high density more than 5 gr/cm<sup>3</sup> and atomic weights between 63.5 to 200.6 g/mol (1-3). This group of chemical compounds are used in mining, ceramics, automobile industries, electronics, battery manufacturing industries, agricultural production, and to improve home care (4-6). HMs are widely distributed in the ecosystems due to human activities. The chemical structures and other characteristics for some of HMs are listed in Table 1. As HMs have long half-life and are non-biodegradable in the environment, they have been considered as toxic substances for the public health and environment even at ppb levels (4, 6, 7). These toxic metals affect cardiovascular systems, immune, digestive system, and the normal functioning of neurological via accumulation in the soft tissues and living organisms (8, 9). In fact, some of them

like Cu and Zn are proposed as carcinogens by the United States Environmental Protection Agency (USEPA) (7, 10). Therefore, in recent years, the remediation of very low concentrations of HMs from aqueous solutions media has become a very hot topic (9, 10).

The results of bibliographic searches show that the adsorption process is a cost-effective technology with high efficiency, enabling large-scale application to remove HMs from drinking water, groundwater, and wastewater (11-14). During the last decade, nanocomposites have been the most widely used fundamental and suitable adsorbents due to its large surface area, excellent surface activities, its robust efficiency and rapid reactivity (13,15-18).

Generally, nanoparticles are particles with a size ranging from smaller than 100 nm (18). Therefore, in many researches, it is attempted that the modification



**Table 1.** Characteristics of the heavy metals discussed in the study

Name	Abbreviation	CAS Number	Molecular Weight (g/mol)	Density (g.cm <sup>-3</sup> ) at 20°C	Atomic Number
Zinc	Zn	7440-66-6	65.4	7.1	30
Chromium	Cr	7440-47-3	51.996	7.19	24
Arsenic	As	As <sup>+3</sup> = 22569-72-8 As <sup>+5</sup> = 17428-41-0	74.9216	5.7	33
Cadmium	Cd	7440-43	112.4	8.7	48
Nickel	Ni	7440-02	58.71	8.9	28
Copper	Cu	7440-50-8	63.546	8.9	29
Mercury	Hg	7439-97-6	200.59	13.6	80
Fluorine	F	7782-41-4	18.998403	1.8*10 <sup>-3</sup>	9

of various adsorbents (such as activated carbon, zeolite, montmorillonite, etc) with nanostructures to show a very high efficiency of simultaneous adsorption of HMs from wastewater due to the creation of very high affinity to simultaneous adsorption of water pollutants.

Therefore, the aim of this study was to collect information on the associations with the use of different nanocomposites for simultaneous adsorption of HMs from aqueous solutions by kinetics and isotherms analyses. The optimization value of simultaneous adsorption parameters such as initial concentration, contact time, adsorbent dosage, and pH was discussed. Furthermore, the adsorption mechanism was investigated. To the best of our knowledge, this research is the first systematic review (SR) that evaluated simultaneous adsorption of multiple HMs by nanocomposites.

## Materials and Methods

This SR was conducted on May 9, 2021 and based on the Preferred Reporting Items for Systematic reviews and Meta-Analyses (PRISMA) statement, to review studies on simultaneous adsorption of HMs by nanocomposites in aqueous solutions (19-21). The peer-reviewed literature was systematically searched in all available electronic information databases including Institute for Scientific Information (ISI), Scopus, and PubMed during 1990-2021. The keywords were used based on the Medical Subject Headings (MeSH) as follows: [(removal) OR (adsorption) OR (sorption) OR (treatment) OR (simultaneous adsorption) OR (simultaneous removal) AND (aqueous) OR (wastewater) OR (water) AND (nanoparticle) OR (nanocomposite) OR (nanostructure) OR (nanoadsorbent) OR (nanomaterial) AND (heavy metal) OR (lead) OR (Pb) OR (cadmium) OR (Cd) OR (arsenic) OR (As) OR (mercury) OR (Hg) OR (zinc) OR (Zn) OR (copper) OR (Cu) OR (chromium) OR (Cr) OR (nickel) OR (Ni)].

Firstly, duplicate papers were excluded using Mendeley software. Then, some papers were included and excluded based on the title/abstract assessment. In this study, inclusion (a) and exclusion (b) criteria as follows:

(a) Articles published in English in peer-reviewed

journals with a focus on simultaneous adsorption of HMs through different types of nanocomposites in aqueous solutions.

(b) Articles published as reviews, presentations, letters, books, conference papers, theses, guidelines, and short communications. Finally, the full texts of the remaining studies were downloaded and checked by two reviewers (ZN and SM). The flowchart of the paper search is illustrated in Figure 1. The information extracted from each original paper include study ID, type of HM, type of nanocomposite, operational parameters such as initial concentration, contact time, adsorbent dosage and pH, adsorption kinetic, adsorption isotherm, maximum adsorbent capacity, and possible adsorption mechanism (Table 2).

## Results

Firstly, 210 records were identified through a systematic search in the mentioned three databases, as shown in Figure 1. Out of them, 190 records were excluded because review articles, duplicates and adsorbents were not related to nanocomposites. After reading full-texts of 20 records, 11 records were excluded because simultaneous adsorption is not related to HMs together. Then, 9 records were included based on the inclusion and exclusion criteria and investigated for this review (4-10,22,23). The summarized information of the included records are listed in Table 2. Among 9 screened records, 5 records were on As; 6 records on Cd; 5 records on Pb, 3 records on Cu, one record on Zn, one record on Hg, one record on Cr, one record on Ni, and finally, one record on F (Table 2).

## Discussion

### Type of adsorbents and their efficiencies

Nine types of nanocomposite as adsorbents have been used by different studies from 2013 to 2020 for simultaneous adsorption of HMs in aqueous solutions (Figure 1). The nanocomposites including hydroxy ferric phosphate and hydroxy ferric sulfate coating on *Aspergillus*, biochar-supported zero-valent iron, polymer-based hydrated iron oxide, Fe-Ti oxides, graphene oxide modified with 2,20-dipyridylamine, zeolite-supported zero-valent iron,

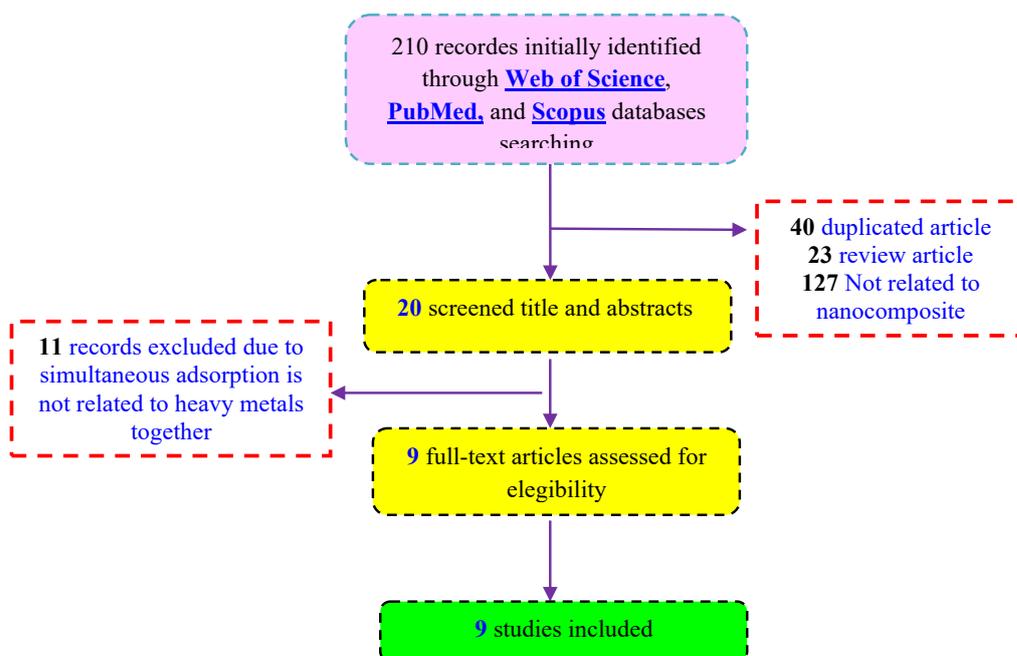


Figure 1. Summary of a standard four-step protocol for literature review.

Table 2. Summary of the included studies based on the simultaneous adsorption of heavy metals on nanocomposite

Possible Adsorption Mechanism	Isotherm Type	Kinetic Type	Removal (%)	Adsorption Capacity (mg/g)	Operational Parameters	Nanocomposite	Heavy Metal	Ref.
Transformation of hydroxyl groups, precipitation	Freundlich	Pseudo-second	As=77 Cd=74 Pb=94	As=162.00 Cd=205.83 Pb=730.79	Cons=100 mg/L pH = 4.0 Time=48 h Sorbent=0.01 g (n-HFP) 0.09 g (n-HFS)	Hydroxy ferric phosphate (HFP) & hydroxy ferric sulfate (n-HFS) coating on Aspergillus	As, Cd, Pb	(22)
Electrostatic interaction, formation of ternary surface complexes, and precipitation	Redlich–Peterson	Pseudo-second	98 for two metals	Cd=179.9 As=158.5	pH = 4.0 Time=1 h Cons=60 mg/L	Biochar-supported zero-valent iron	Cd, As	(5)
Electrostatic interactions and the coordination of the hydrated iron oxide nanoparticles	Freundlich	Pseudo-second	94 for two metals	As=71.56 p-ASA=41.63	Cons=50 mg/L Sorbent=100mg	Polymer-based hydrated iron oxide	Organic/inorganic As	(23)
Presence of multiple oxides in the synthesized material	Freundlich	-	As=98 Cu=94 F=43 Cd=100 Pb=98	As=2.8 Cd=1.86 Pb=2.98 F=7.2 Cu=118	Cons=0.15-4 mg/L Time=10-30 min pH = 7 Sorbent =0.005 g/L	Fe-Ti oxides	As, Cd, Cu, Pb, F	(8)
Electrostatic interactions with oxygen-containing groups and the strong surface complexation	Langmuir	Pseudo-second	87 for four metals	Pb=369.749, Cd=257.201, Ni=180.893 Cu=358.824	Sorbent =8 mg Cons=20 mg/L pH =5 Time=4 min	Graphene oxide modified with 2,20-dipyridylamine	Pb, Cd, Ni, Cu	(6)
Electrostatic adsorption, ionic exchange, oxidation, reduction, co-Precipitation	Langmuir	Pseudo-second	-	As=11.52 Cd=48.63 Pb=85.37	pH=6 Time=10 h	Zeolite-supported zero-valent iron	Cd, Pb, As	(4)
chemisorption between Cd and Fe surface	-	-	Cr=100 Cd=100	Cr=60 Cd=60	Cons=70 mg/L pH =6 Fe dose: 0.15 g/L	Silica-Coated Fe <sup>0</sup>	Cr, Cd	(7)
Electrostatic interaction	Langmuir	Pseudo-second	Hg=98 Pb=92	Hg=200 Pb=142.85	pH = 6-7 Sorbent =0.4 g Time=1 h Cons=0.1 mg/L	Ceria entrapped in tamarind powder	Hg, Pb	(9)
Electrostatic interaction	-	Pseudo-second	Cu=93 Zn=58	-	Cons=100 mg/L	Zero-valent iron impregnated with clays	Zn, Cu	(10)

silica-coated Fe<sup>0</sup>, ceria entrapped in tamarind powder, and zero-valent iron impregnated with clays (4-10, 22, 23). The maximum adsorption capacities ( $q_e$  (mg/g)) of nanocomposites for multiple HMs were investigated and it was found that all of the adsorbent have good adsorption capacity for simultaneous adsorption of As, Cd, Pb, F, Ni, Cu, Cr, Hg, and Zn. Among included adsorbents in SR, hydroxy ferric sulfate has the highest adsorption capacity for Pb (730.79 mg/g), while the minimum adsorption capacity was related to zeolite-supported zero-valent iron for As (11.52 mg/g). Many researches have shown that the simultaneous adsorption capacities of nanocomposites were greatly improved compared to adsorbents without nanostructures for multiple HMs. Modification of different adsorbents with nanomaterial increases the surface area of material and improves the affinity of adsorbents to simultaneous adsorption of multiple HMs (22). Generally, nanocomposites exhibited remarkable adsorption performance for HMs.

#### ***Influence of key factors (pH, initial concentration, adsorbent dosage and contact time) on adsorption***

Optimization of many factors such as contact time, adsorbent dose, pH, and initial dye concentration can affect the development of industrial-scale adsorption process (12,24). The solution pH is an important factor in adsorption process of pollutants. The results of the included records show that solution pH affects the simultaneous adsorption performance of different nanocomposites for HMs via the changes of the characteristics and the functional groups of surface adsorbent (with changes in  $pH_{pzc}$  of adsorbent). Furthermore, the solution pH can affect the surficial charge of HMs within the solution. This process can create the electrostatic or repulsive force between HMs and surface of the adsorbent. Therefore, the optimal pH depends on the types of nanocomposites and target HMs. The maximum and minimum pHs in studies were 6 and 4, respectively, as shown in Table 2. Thus, the simultaneous removal rate of HMs decreased with an increase in pH value (4, 5, 23).

Initial dye concentration has a great effect on adsorption process. Literature has shown that the removal efficiency of adsorbed HMs was decreased with increasing initial concentration of HMs. It can be concluded that the increase of initial concentration can affect available binding sites and active sites of the adsorbent surface. This was attributed to the fact that the simultaneous removal of HMs is a surface-mediated process. When more ions of HMs approached the surface of adsorbent, the available active sites decreased for per mole ions. Thus, lower removal rates of HMs were obtained with increasing initial concentration of HMs (7, 10).

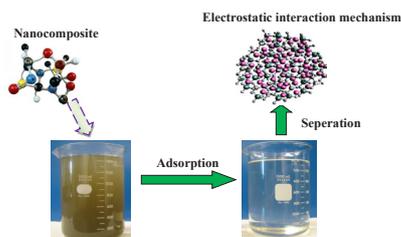
The effect of the adsorbent dosage is another key parameter in the adsorption of HMs. According to the extracted data in SR, effectiveness and efficiency (%) of

the HMs removal improve with increasing the adsorbent mass. As the adsorbent dosage grows, contact and interaction between the adsorption sites and the target molecule in the solution can increase due to the greater accessibility of surface binding sites.

The equilibrium time is known as a critical factor that substantially influences the adsorption process. Based on Table 2, the adsorption studies have been conducted in a period between 4 minutes to 48 hours. The results of records show that the adsorption rate and removal efficiency increases drastically, and then, increases gradually until the sorption system reached equilibrium. This issue can be explained by the fact that the plenty of the active sites available on the adsorbent exist at the beginning of the sorption process for simultaneous adsorption of metallic molecules. While, vacant sites on the adsorbents are occupied and saturated by ions of HMs with the increase of contact time (12).

#### **Adsorption kinetics and isotherm**

The kinetic and isotherm studies in adsorption process were used to investigate the interactions between the adsorbent and the adsorbate in the solid-liquid phase. The kinetic and isotherm models can be used to describe the affinity strength of the adsorbent and adsorption capacity at different equilibrium concentrations. Five adsorption isotherms models (Langmuir, Freundlich, Redlich-Peterson, and Sips) and three adsorption kinetic models (pseudo-first-order, pseudo-second-order, and intraparticle diffusion) were employed to describe experimental adsorption data. All the adsorption experiments with mixed pollutants on the nanocomposites were performed under optimal experimental conditions. Based on the value of coefficient of the kinetic model ( $R^2$ ) and very close the theoretical  $q_e$  to experimental  $q_{exp}$  values for all of the ions, all of the simultaneous adsorption studies reported that the pseudo-second-order model was fitted better than the pseudo-first-order and intraparticle diffusion models. On the basis of these findings, there is a strong chemical force between metal ions and the functional groups on the nanocomposite, which creates a strong surface complexation between adsorbates and adsorbent. Langmuir and Freundlich models were more relevant to the simultaneous adsorption processes for the metals. The Langmuir and Freundlich models were assumed as monolayer and multilayer adsorption models, respectively. Redlich-Peterson model indicates that the adsorption process was a hybrid chemical reaction sorption process. Therefore, the isotherm model of an absorption process depends on the synergy among the multi-functional components strengthened the removal of HMs, and different types of adsorption sites on the adsorbent surface (4, 5, 22, 23)



**Figure 2.** Simultaneous adsorption mechanism of heavy metals from aqueous solution.

### Adsorption mechanism

Some studies have evaluated simultaneous adsorption pathways for ions of HMs on various adsorbents under different experimental conditions. Based on the literature review, multiple pathways can be utilized to the adsorption of metal species onto nanocomposites. The main mechanisms of HMs adsorption onto nanostructures were electrostatic interactions and surface complexation between hydroxyl and carboxyl groups and positively charged metal ion, precipitation (the formation of insoluble minerals) (Figure 2) (4, 5, 7, 9).

### Future perspectives

Metal ions can exert indirect and direct harm to the public health once discharged into the water resources due to their non-biodegradable nature. Today, in order to protect aqueous matrices, various nanomaterials are used to remove pollutants from drinking water, wastewater and groundwater due to good performance (4, 8, 9, 14, 18). Therefore, researchers are recommended to synthesize non-toxic and safe nanoadsorbents through green methods. In addition, researchers investigate the existence of synergistic effects of HMs in aqueous media.

### Conclusion

In this systematic review, the adsorption efficiency of different nanocomposites with high surface area were studied in the simultaneous adsorption of ions of HMs from aqueous matrices during 1990-2021. It is a big challenge to simultaneously adsorb of HMs efficiently and cost-effectively. It has been clearly demonstrated that nanocomposites exhibited remarkable adsorption performance for HMs and the simultaneous adsorption rates of HMs were higher than that of bulk materials. Generally, parameters such as initial concentration, contact time, and adsorbent dosage were discussed and this study confirmed that the simultaneous removal rate of HMs decreased with an increase in pH value. Findings indicate that the Langmuir and Freundlich models and the pseudo-second-order kinetic model have been widely used and the most popular to describe the equilibrium of HMs by nanoadsorbents.

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### Ethical issues

This study is not approved by the Research Ethics Committee of Ilam University of Medical Sciences. The authors certify that all data collected during the study are as presented in this manuscript, and no data from the study has been or will be published elsewhere separately.

### Competing interests

The authors declare that they have no known potential or actual competing financial interests.

### Authors' contributions

ZN and SAM contributed to conception and design, execution and data collection. ZN, SAM, SS, and NJ contributed to data analysis and interpretation. All authors critically reviewed, refined, and approved the manuscript.

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