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REVIEW ARTICLE

Removal of heavy metals from the aqueous solution by nanomaterials: a review with analysing and categorizing the studies

Shervin Adabi[1] · **Ahmadreza Yazdanbakhsh**[1,2] · **Abbas Shahsavani**[1] · **Amir Sheikhmohammadi**[3] · **Mahdi Hadi**[4]

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Abstract

With the development of nanotechnology and its application in various sciences, scientists have investigated the use of nanoparticles as adsorbents to remove heavy metals from aqueous solutions all over the world. So far, the results of many of these studies have been published in reputable journals. Obviously, reviewing these articles and summarizing the results of these studies from different aspects will provide new perspectives for the development of this technology for heavy metals removal from water. So the current study was performed to review the results of the published studies between 1/January/1980 to 1/January/2022. The focus of the study is on the analysis of these studies and their classification. In addition, a more detailed investigation was carried out. Among the 5155 articles, 576 articles were included based on Cochrane protocols. Results show that most of the studies (90.8%) were conducted on a laboratory scale and used synthetic solutions. Most studies were performed for Pb, Cd and Cu, removal respectively. Compared to other countries, authors with affiliation from China and Iran have published more articles. The ranking of the use of various nanomaterials were: nanocomposites > metal oxide nanomaterials > metal-based nanomaterials > carbon-based nanomaterials > dendrimers, with the wide range of sizes from less than 10 nm to several hundreds of nanometers. The required amount of carbon-based nanoparticles to remove many heavy metals were lower than other nanoparticles. In most studies, $pH \leq 7$ has been reported as optimal. Most studies have been followed pseudo second-order and pseudo first-order reactions and have been more agreement with Langmuir and Freundlich adsorption isotherms respectively. The results of studies show that the synthesis and optimization of new nanomaterials can be considered as a new and competitive technology. However, more studies are needed to investigate the removal of heavy metals in real samples and to overcome some challenges in the full-scale application.

Keywords Heavy metals · Aqueous solutions · Nanomaterials · Adsorbtion · Systematic review

✉ Ahmadreza Yazdanbakhsh
yazdanbakhsh@sbmu.ac.ir

Shervin Adabi
Sh_adabi@sbmu.ac.ir

Abbas Shahsavani
ashahsavani@gmail.com

Amir Sheikhmohammadi
asheikh1359@gmail.com

Mahdi Hadi
hadi_rfm@yahoo.com

1 Department of Environmental Health Engineering, School of Public Health and Safety, Shahid Beheshti University of Medical Sciences, Tehran, Iran

2 Workplace Health Promotion Research Center, Shahid Beheshti University of Medical Sciences, Tehran, Iran

3 Department of Environmental Health Engineering, School of Public Health, Khoy University of Medical Sciences, Khoy, Iran

4 Center for Water Quality Research (CWQR), Institute for Environmental Research (IER), Tehran University of Medical Sciences, Tehran, Iran

Introduction

The increase in world population and industrialization can be a serious threat to the supply of healthy drinking water sources. This issue is one of the most challenging issues that make the supply of safe drinking water one of the main concerns around the world. One of the main reason for the scarcity of safe and healthy drinking water in the world, is the presence of the various pollutants in the water sources. These pollutants can enter water sources in the different ways, especially through industrial wastewater [1, 2].

Meanwhile, heavy metals are among the most common and important pollutants entering water environments. Like many pollutants, the important source of contamination of water sources with heavy metals are industrial wastewaters, in addition, there are other ways of their entry into water sources. Many mineral springs contain heavy metals (including arsenic), which may come into contact with different substrates, and threaten the water sources due to passing through the depths of the earth. In addition, these metals pollute the environment through mining (production of ingots, etc.), agricultural activities (toxins used for pest control), as well as fuel and energy production [3, 4].

The presence of heavy metals in water and soil, in addition to threatening the public health (causing carcinogenicity and short-term genetic effects), can threaten the aquatic and agricultural ecosystems. The importance of heavy metals is due to their special characteristics, such as accumulation in body tissues, non-degradability in the environment, toxicity to living organisms at low concentration, and increasing the toxicity of some of them in the environment over time (for example mercury) [5].

Although many heavy metals are essential to life, but entering the body beyond the permissible limit can cause chronic poisoning and acute problems. Excessive accumulation of each of the metals in different tissues of the body will cause specific problems, for examples, kidney injuries (through mercury), liver problems (through cadmium), respiratory diseases (through chromium), skin problems (through arsenic), digestive problems (through copper and cadmium) and adverse effects on the nervous system function (through lead) [6–9]. According to the guidelines of the United States Environmental Protection Agency, the maximum allowed concentrations of mercury, cadmium, chromium, arsenic, copper and lead in drinking water are suggested 0.002 mg L^{-1}, 0.005 mg L^{-1}, 0.1 mg L^{-1}, 0.01 mg L^{-1}, 1.3 mg L^{-1} and 0.015 mg L^{-1} respectively [4, 10].

According to the mentioned points, in order to prevent toxic effects and environmental hazards of heavy metals, their removal from wastewater and polluted waters should be considered as an important and urgent measure. Different methods have been utilized to remove heavy metals from the contaminated water, such as chemical precipitation, membrane filtration, adsorption, and ion exchange. [11–13].

The adsorption Technique is one of the common methods for removing pollutants, including heavy metals from water environments. This process has some benefits, including good efficiency, simplicity low cost and availability of different absorbents. However, due to the excessive waste products, challenges for the recovery and regeneration of adsorbents, the low absorption capacity of some absorbents, researchers have tried to produce high capacity adsorbents with multiple reuse capabilities.[14–16].

In recent years, the successful development of nanotechnology has led to its application in various other fields, including the removal of pollutants from water and wastewater. [17, 18] Researches on the synthesis and application of nanomaterials as new adsorbents such as modified (CNTs), magnetic nanoparticles (MNPs) and other nanomaterials to remove pollutants from water environments are one of the actions that have been carried out along with the development of nanotechnology [19, 20]. Until now, many studies have been conducted using nanomaterials such as carbon-based nanomaterials, metal-based nanomaterials, metal oxide nanomaterials, dendrimers and nanocomposites to remove various metals such as cadmium, mercury, and zinc from aqueous solutions, which the results of them have been published in scientific journals [21–23]. The goals of these studies were to introduce adsorbent nanoparticles with new properties and to investigate various aspects affecting the process, such as the type and concentration of heavy metal, adsorbent dose, reusability of adsorbent, adsorbent capacity, kinetic and adsorption isotherm. The systematic review of these studies can provide valuable scientific information for further research. In this regard Rajeev in a review study was examined the removal of heavy metals by the absorption process. In this study, the type of adsorbent, contact time, pH and some other parameters were examined [24]. Zito et al. conducted an overview of the subject of "mineral nanoparticles for removing heavy metals and arsenic". The focus of this study has been the use of nano - engineed mineral adsorbents with the emphasis on the benefits and limitations of nanostructured type. ([25]. In Other study by Hua et al., in one review study, the results of the research on the removal of heavy metals from water and sewage using nanometal oxides were analyzed [26]. A review study is focused on summarizing the results of researches on the removal of various types of heavy metals by nanoparticles. Effective parameters and optimal conditions for their removal are reported [27].

In general, the review of the literature shows that, in each of the reviewed studies, certain aspects of the nanoparticles application for the removal of heavy metals have been considered. The review of studies based on the classification of studies and their analysis is limited or have been reviewed in a short time interval. So the main purpose of this study was to review the research conducted in the field of using various types of nanomaterials to remove heavy metals from aqueous solutions. The distinction of this study compared to other studies can be considered the following; provided a detailed overview of the articles in a long period of time, the classification of studies based on the type of water sources under investigation, whether the study was conducted in a laboratory or full scale, the trend of the number of studies conducted during the last two decades, the publisher of the articles, the ranking of the countries based on the number of published articles, the ranking of the metals and nanoparticles examined, the dosage of the nanoparticles used, and the examination of the absorption process parameters such as kinetics and absorption isotherms.

Materials and methods

Search strategy

The search process was performed according to Cochrane protocols [28]. A schematic of the search process is shown in Fig. 1. In the search process, articles were considered that examined the removal of heavy metals in aqueous solutions using nanomaterials. The searching was accomplished among the two international databases Scopus and Web of Knowledge, were examined between January 1, 1980 to January 1, 2022.

The following terms were used as key words: Scopus: (TITLE-ABS-KEY ({water treatment} OR {wastewater} OR {aqueous solution} OR {drinking water} OR {surface water} OR {water reuse} OR {groundwater} OR {potable water}) AND TITLE-ABS-KEY ({nanotechnology} OR {nanofilter} OR {nanoparticle} OR {nanofiber} OR {nanotube} OR {nanocomposite} OR {nanostructure} OR

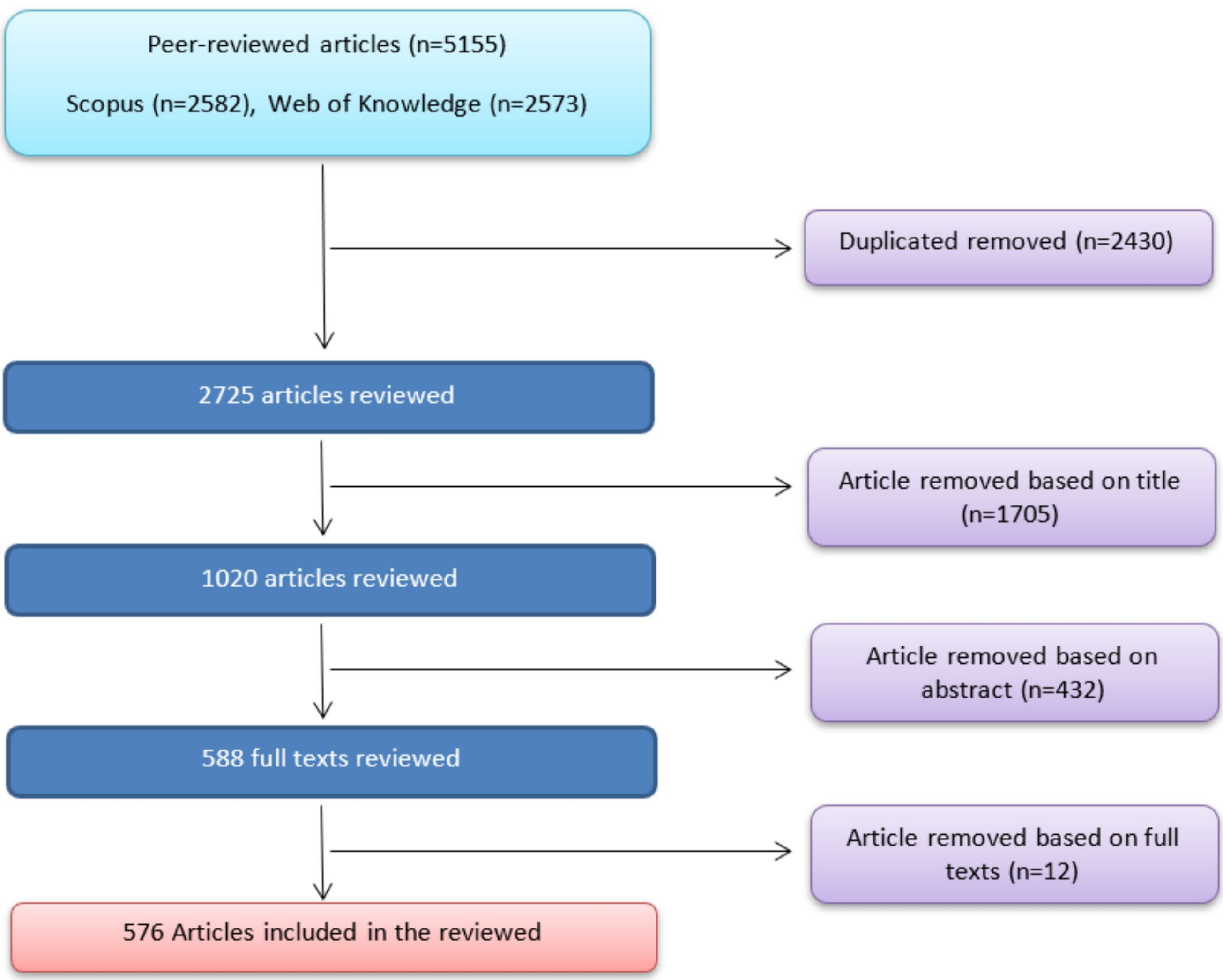


Fig. 1 Selection process studies conducted on removing of heavy metals from aqueous solution by nanomaterials

{nanomaterial} OR {dendrimer} OR {nanobiocide} OR {nanoscale} OR {nanozeolite} OR {nano-} OR {nanocrystal})) AND TITLE-ABS-KEY ({heavy metals}); Web of Knowledge: (TS=("water treatment" OR "wastewater" OR "aqueous solution" OR "drinking water" OR "surface water" OR "water reuse" OR "groundwater" OR "potable water") AND TS= ("nanotechnology" OR "nanofilter" OR "nanoparticle" OR "nanofiber" OR "nanotube" OR "nanocomposite" OR "nanostructure" OR "nanomaterial" OR "dendrimer" OR "nanobiocide" OR "nanoscale" OR "nanozeolite" OR "nano-" OR "nanocrystal")) AND TS= ("heavy metals") Also, the references lists of articles were assessed in order to retrieve additional citations.

Inclusion criteria and data extraction

The inclusion criteria included as (a) removing of heavy metals from the aqueous solutions by nanomaterial; (b) published in English language; (c) research papers; (d) articles that investigated and reported the parameters of heavy metal concentration, optimal pH value, type and size of nanomaterials and type of water source. It should be noted that review articles and books were excluded. The extracted characteristics of each study were consisted of the year of study, first author name, country, journal name, type of heavy metal and type of the nanomaterial, type of the water source, type of the water treatment method, size of the nanomaterial, concentration of heavy metal in the water solutions, the optimum pH and the heavy metal concentration in the treated solution, type of kinetic and isotherm adsorption.

Statistical analysis

In this study data preparation and visualization of all results were performed using Microsoft Excel. Excel's pivot table was used to generate summary tables of means, standard deviations, counts, etc.

Results and discussion

Retrieval studies process

It should be noted that the purpose of this study was to review articles on the removal of heavy metals from aquatic environments by nanomaterials, so other pollutants such as persistent organic pollutants (POPs) and other environments such as soil were not considered. Of the 5155 reviewed articles, 2582 articles from Scopus and 2573 articles from Web of Knowledge were collected (between 1/January/1980 to 1/January/2022). Then, in the first step due to repetition, 2430 articles were excluded via EndNote citation manager (vX7, Thomson Reuters, New York, USA). In second step based on the title of articles almost 63% of those which were residuum (n = 1705), were declassed thereupon 1020 articles were remained. Subsequently, based on abstract, 432 irrelevance articles were excluded and finally full text of 588 retrieved articles were downloaded and after studying of all of them, 12 inconsistent articles were excluded, so all 576 articles were included (Fig. 1).

Characteristics of studies

As shown in Fig. 2, most of the studies (90.8%) were conducted on a laboratory scale and synthetic solutions were used in them. Few studies have used real water samples to investigate this issue. It seems that researchers have used synthetic samples in their studies in order to have a better control in relation to investigating the effect of variables on the process. On the other hand, studies on synthetic samples cannot reveal the effects of natural water and wastewater interventions on process performance. Anyway, in some studies, they have tried to simulate the conditions of real samples as much as possible.

Figure 3 shows the ranking of countries based on the number of studies and the type of nanomaterials used. Most of the articles were published by China and Iran, followed by India and South Korea are ranked 3rd and 4th by a large margin. Also, most of the studies conducted in China have been based on metal nanomaterials, while in the studies of Iranian researchers, many studies with nanocomposites are observed.

Another finding of this chart is that dendrimers have been used mainly in studies conducted in Iran, Saudi Arabia, Malaysia, Thailand, India and Taiwan. It should be noted that these countries are interested in research in the field of nanotechnology [29, 30]. If the number of published articles are ranked by continent, the countries of the Asian continent are ranked first. While the American and European countries are ranked second and third respectively. The reason for most of the studies conducted in the Asian continent may be related to the larger population and size of this continent, especially in China and India, as well as many problems of water pollution in Asian countries.

Figure 4 shows the number of articles published by different publishers as well as by journals.

Elsevier publisher ranks first in publishing these articles. The next rank belongs to the Springer. Also, according to Fig. 4, nanocomposites have been used more than other nanomaterials in these studies. Nanocomposites have been frequently used in many applications due to their superior properties. Particularly, polymer-based nanocomposites often present superior physical, chemical and mechanical properties, as well as superior compatibility. Metal-based

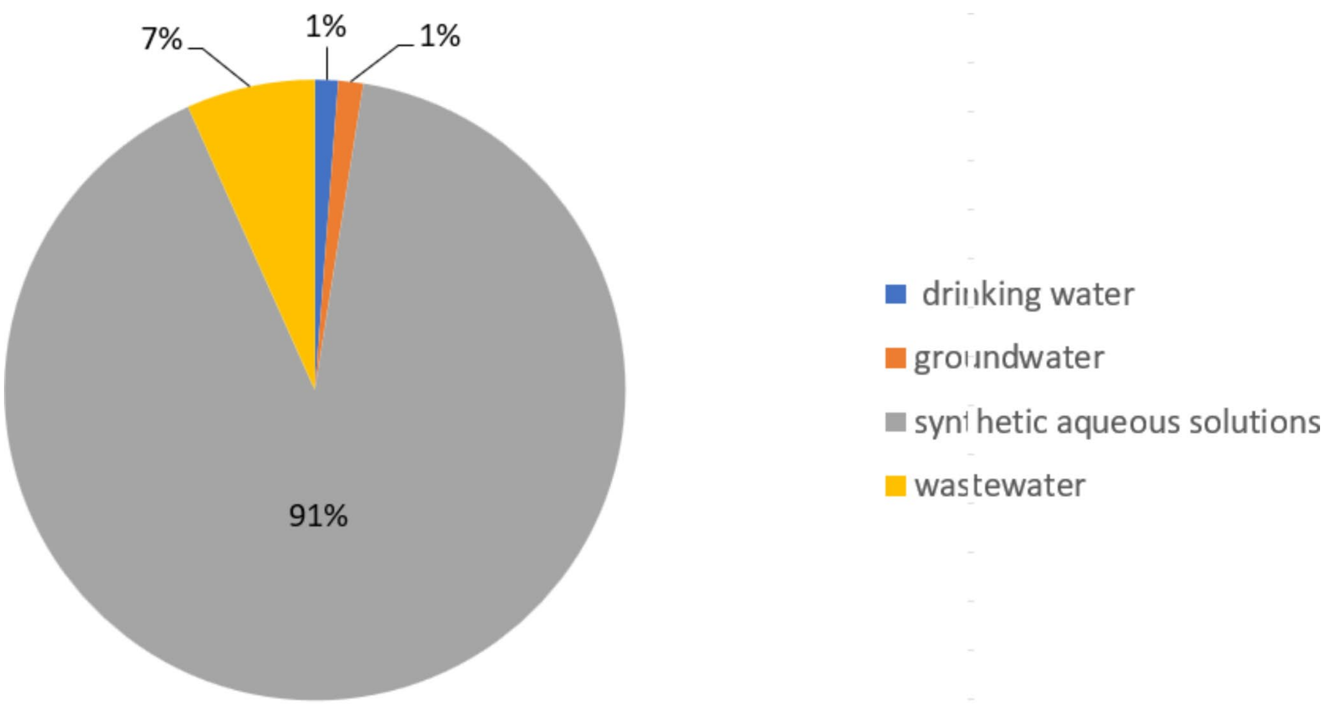


Fig. 2 Distribution of water samples used in studies

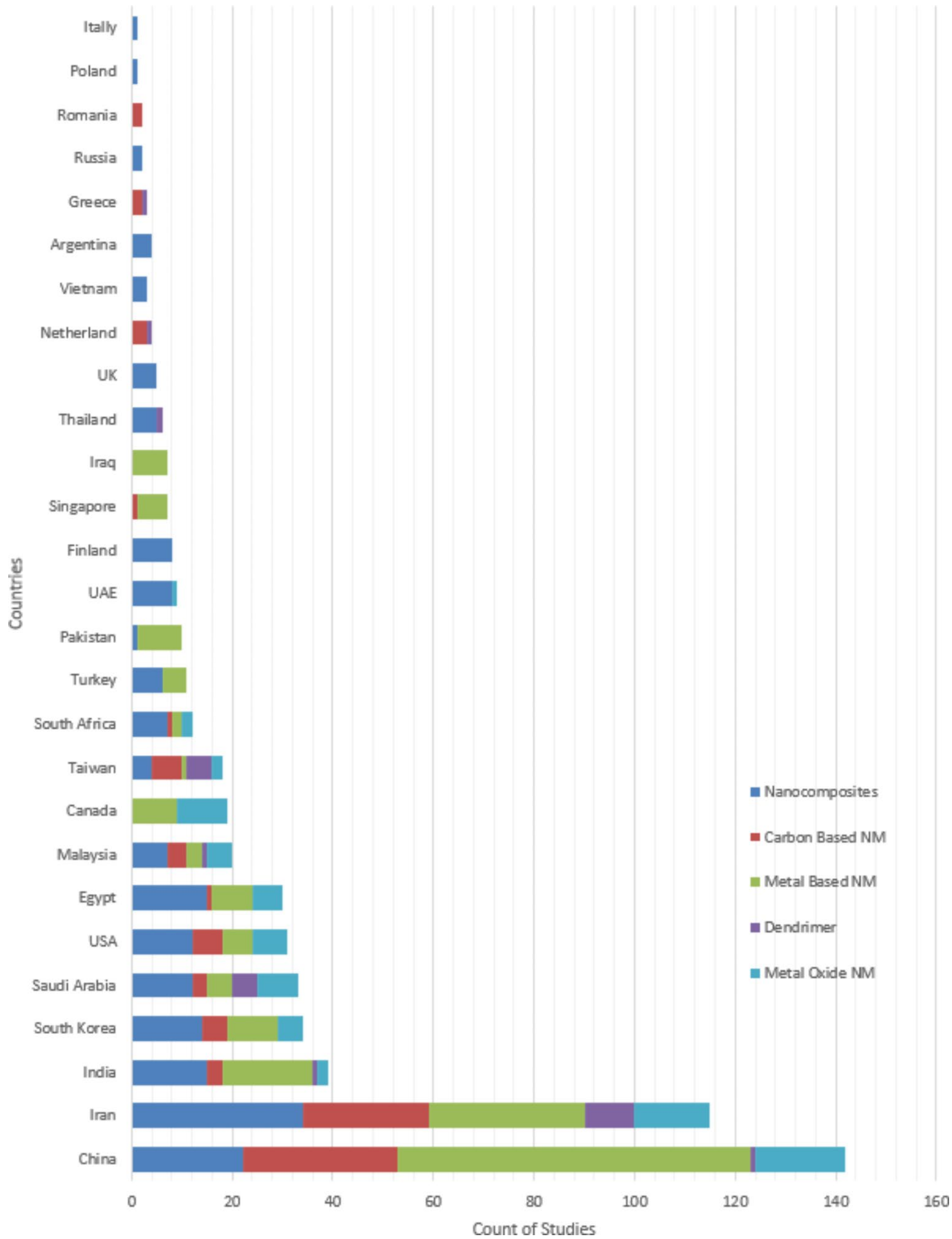
nanomaterials are the second class of nanomaterials that have been widely investigated. A possible explanation may be that iron and silver nanoparticles have been used in many studies. Because these nanoparticles with different properties are synthesized in the laboratory, so many studies have been conducted to investigate their performance in different conditions.[31–33].

Figure 5 shows the number of published articles using different nanomaterials based on the year of publication. The beginning of the science of nanotechnology was in the last decades of the 20th century, and therefore from the beginning, scientists paid attention to its use in the environment and the removal of pollutants. As the Fig. 5 shows, the first paper was published in 2003 in which nanocomposite was used to remove heavy metals [34]. The second article was published in 2004, which was the result of using carbon-based nanomaterials.[35]. The early years of 2010 was the beginning of the economic applications of nanotechnology. Figure 5 confirms that the number of published studies has increased sharply since 2010. The results of Fig. 4 also confirm that more metal based nanomaterials than other nanomaterials have been used for water treatment. Nanocomposites and metal-based nanomaterials have been extensively studied among a variety of nanomaterials [36]. Figure 5 shows that researchers have tried to design and develop new nanomaterials for versatile applications. Although the synthesis of new nanocomposites and metal-based nanomaterials with unique physical and chemical properties is still a challenging [37, 38]. One of the well-known types of metal-based nanomaterials is magnetic nanoparticles, which have many special properties such as supermagnetism, high coactivity, and high magnetic sensitivity, which make them very suitable for removing pollutants in aqueous solutions. It is clearly seen that the highest number of published articles was in 2020.

Type of heavy metals

Figure 6 shows the number of published articles based on the type of metals to be removed and the type of nanomaterials used. The results showed that most of the studies were for lead removal with more than 160 articles. The use of metal-based nanocomposites and nanomaterials to remove lead from aqueous solution has been more common than other nanomaterials. Lead exists naturally in the environment, but human activities such as the use of fossil fuels and metal mining have led to the increasing release of this metal in the environment. Lead is one of the most important systemic toxins that has destructive effects on the nervous system, liver, kidney, and endocrine system [39, 40]. Articles related to cadmium and copper, are in the second and third positions, respectively. Also, to remove these two metals, the use of metal-based nanocomposites and nanomaterials has been the most widely used. As a toxic metal, cadmium

Fig. 3 Number of publications based on countries and type of nanomaterials



is used in many human resources such as batteries, nuclear reactors, pigments and paints [41, 42]. In recent years, it has been noticed due to serious health problems. As shown in Fig. 6, no study has been done on some heavy metals such as titanium and lanthanum.

Size of nanomaterials

The shape, size, and structure of nanomaterials play a major role in the performance of heavy metals from water and wastewater. Determining the size of nanoparticles is essential to understand the properties of the particles, how they interact with other compounds and how to use them in different applications .Nanomaterials are divided into the following categories by chemical composition: (a) carbon based nanomaterial which includes a variety of carbon structures, (b) metal based nanomaterials which comprises the major part of nanomaterials, (c) metal oxide nanomaterials which they are composed of metal and oxygen, (d) dendrimers, which are nano-sized with high molecular weight (e) multiphase materials nanocomposites with at least one phase in the nano dimension. These materials are subdivided into more calssifications [43, 44]. Figure 7 shows the average size of different types of nanoparticles (nm) used in the studies. Although the size of nanoparticles used has a wide range of sizes from e few hundreds of nanometers to

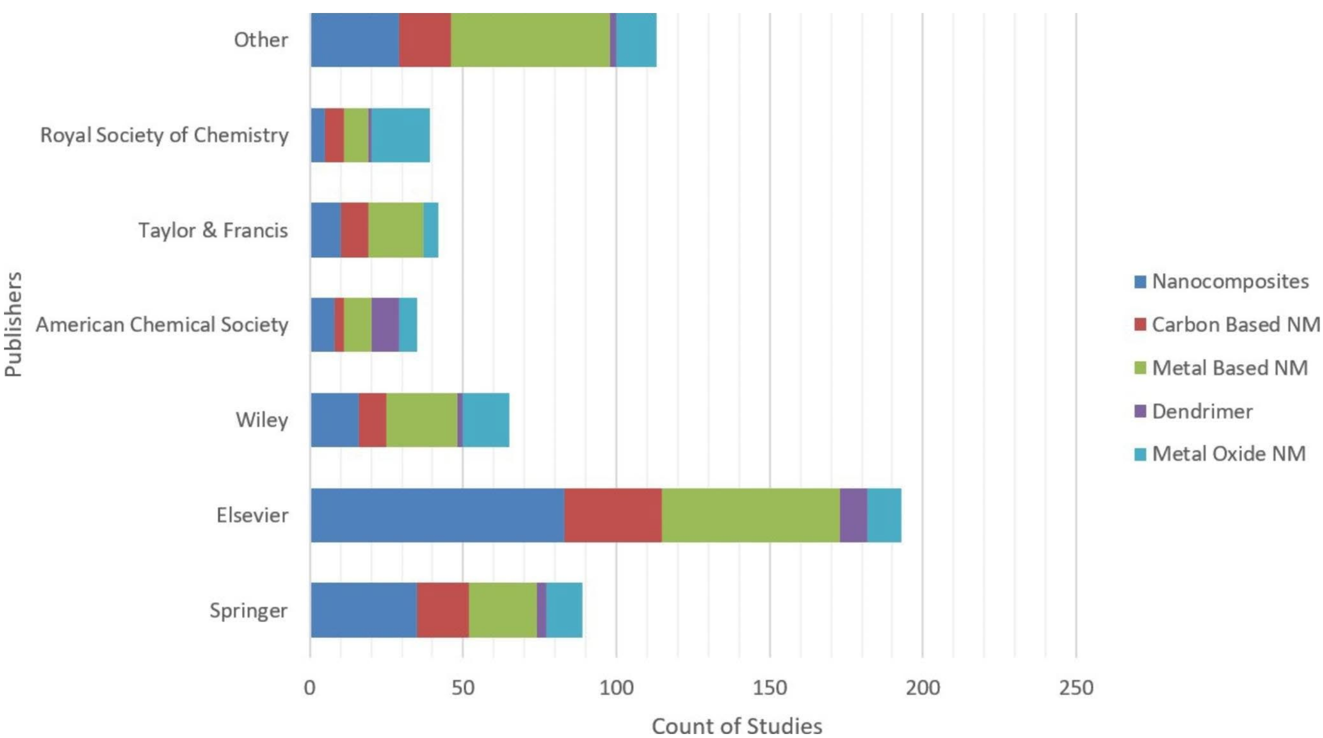


Fig. 4 Number of studies based on publishers and type of nanomaterials

particles less than 10 nanometers. The average size of dendrimers had the smallest size and were slightly more than 20 nm.

Figure 8 shows the statistical expansion of the types of adsorbents used and their optimal amount (mg L-1) for the removal of different metals. The optimum amount of metal-based nanomaterial adsorbent for removing heavy metals such as lead (II), cadmium (II), nickel (II), copper (II) and chromium (VI) were 250 mg/L. However, with the increase of the initial concentration of metals in aqueous solutions, the optimal amount of adsorbent also increases.

The high reducing potential and huge surface area of metal-based nanomaterials improve their performance for heavy metal removal from aqueous solution. It is worth noting that many metal-based nanomaterials contain zero-valent metals, so their removal mechanism for different heavy metals can change according to the standard reduction potential (E^0) of the heavy metals [45–47]. As shown in Fig. 8, carbon-based nanomaterials with low concentrations (5 mg L^{-1}) have excellent adsorption effects for Pb(II), Cd(II), Ni(II), Cu(II), and Fe(II). The adsorption active sites of carbon-based nanomaterials are mainly comprised of outer surface, interstitial channels, inner sites and outer groove sites [48].

Effect of initial solution pH on the adsorbent performance

One of the most significant parameters in the process of metal absorption is pH, because the properties of the metal in solution (for example solubility of heavy metal) and the charge of active sites on the surface can change depending on this parameter [49–51]. Most of the absorbents surfaces have a variable charges, so the pH value of the solution and the point of zero charge can affect the phenomenon of electrostatic interaction [52, 53].

In Fig. 9 the average optimum of pH for the removal of metals with various nanostructures are presented. As it can be observed from the Fig. 9 in most of the studies with various types of nanoparticles, pH $\leq$ 7 was more suitable for removing most of the metals. In the studies that used metal-based nanoparticles, the values of pH $\leq$ 6 were optimal for the removal of most metals, except for mercury (II) and chromium (III).

Order of reaction and adsorption isotherm models

The reaction order is the relationship between the concentrations of species and the rate of a reaction. Numerous kinetic models have described the reaction order of adsorption systems based on solution concentration. These include first-order, second-order, pseudo-first-order, and pseudo-second-order [54]. Figure 10 shows the results of the

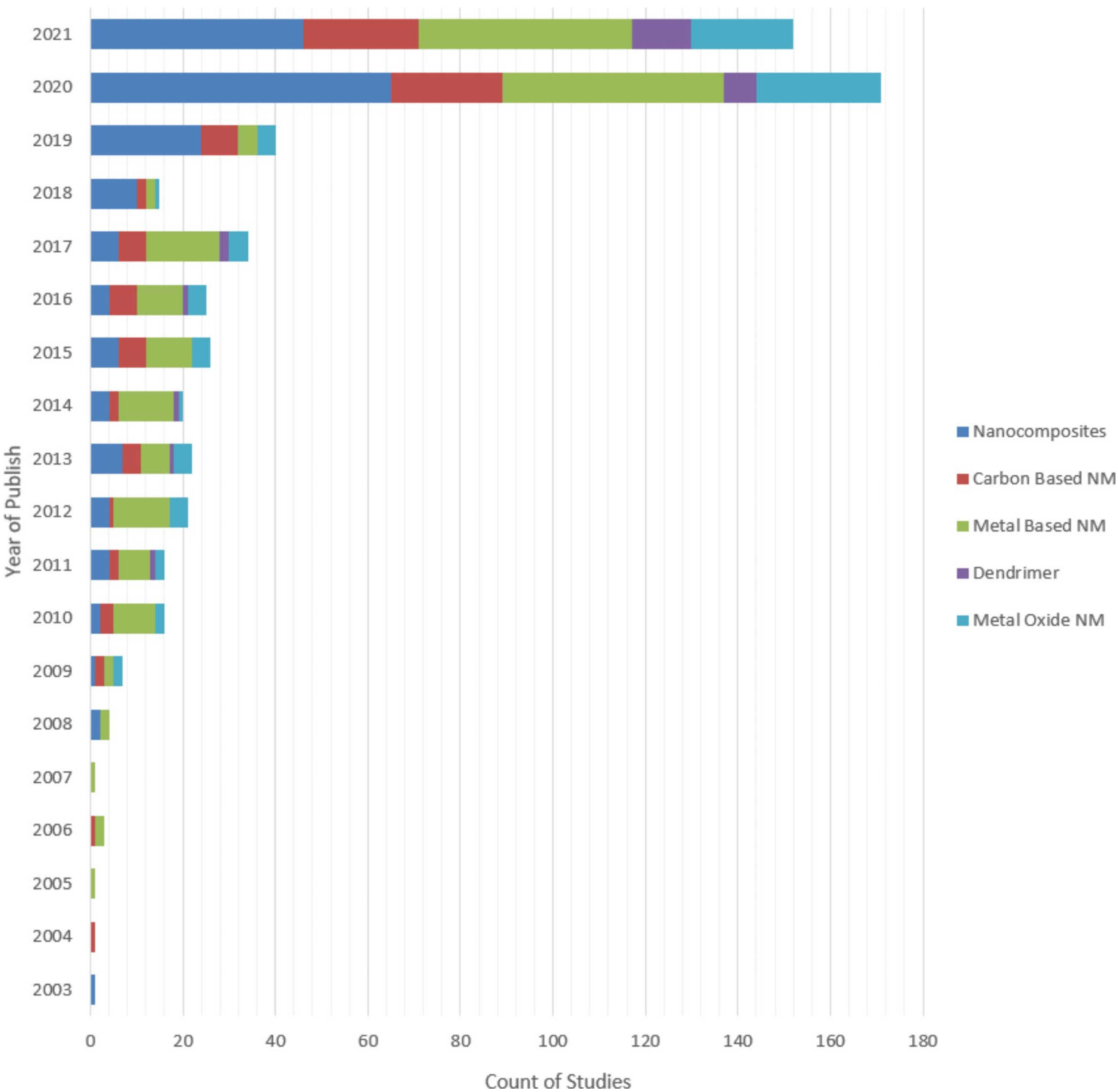


Fig. 5 Number of the studies based on the application of all type of nanomaterial since 1980 to Jan 2021

reaction rate in the reviewed articles. As shown in Fig. 10 in most of the studies conducted, the reaction rates have been consistent with pseudo-second-order reactions. However, in the case of some metals, such as Mn (II), Cr (III), Zn (II), Pb (II), Cu (II) and Ni (II) other degrees of reaction have also been reported.

In the pseudo-second-order kinetic model, it is assumed that the rate-limiting step is chemical adsorption and predicts the behaviour in the entire adsorption range. In this situation, the absorption rate depends on the capacity of the adsorbent material. [20, 55].

Figure 11 shows the results of the most isotherm models used of metal adsorption by nanoparticles. Adsorption isotherms are mathematical equations describing the adsorption capacity for an absorbent at a constant temperature and as an important parameter for the design of the absorption process. So far, based on different assumptions, various isotherm equations have been proposed such as: Langmuir, Freundlich, Temkin and Dubinin–Radushkevich [56–58]. According to Fig. 11, almost all the results of the studies had been in agreement with Langmuir and Freundlich isotherms, although the correspondence with the Langmuir model had

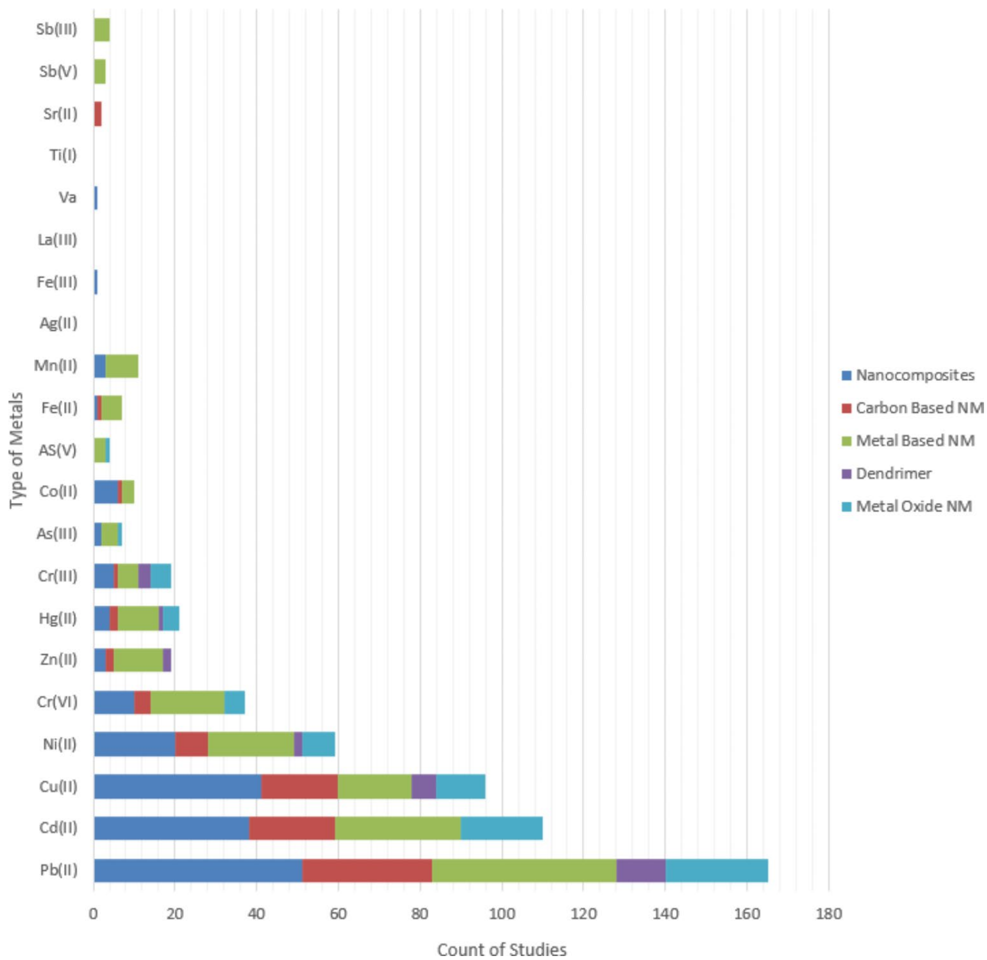


Fig. 6 Comparison of the number of studies based on the type of heavy metals and nanomaterials used

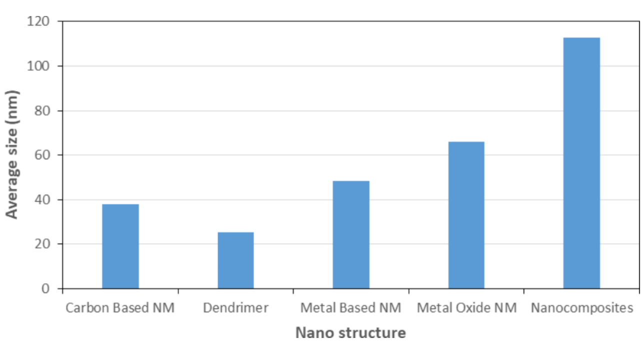


Fig. 7 Average size of different nanoparticles used in studies

been higher with a significant difference. In the case of metals such as Cr (III) and Ag (II), the matching results with Langmuir and Freundlich models had been almost equal.

It should be explained that non-compliance with other isotherms does not necessarily mean that non-compliance of the results with other isotherms. Rather, it is possible that only these two isotherms have been examined in many studies, or the degree of agreement with other isotherms has been lower compared to these two isotherms.

Review of the literature shows that, similar to the absorption of heavy metals, Langmuir and Freundlich isotherms were used in most studies to absorb various organic and inorganic impurities from water and wastewater. The most important feature in the Langmuir isotherm is that monolayer adsorption takes place in homogeneous sites, while in the Freundlich isotherm, multilayer adsorption is applied in heterogeneous sites [59, 60].

As shown in Fig. 11 the compliance with the Temkin model was reported only in two studies.

Conclusion

In this work, studies on the removal of heavy metals from aqueous solution by nanomaterials were reviewed and statistically analysed.

The results showed that the studies of the absorption of heavy metals from aqueous solutions have been very attractive and the development of nanotechnology has played an essential role in accelerating the number of studies and increasing the publication of articles in this field.

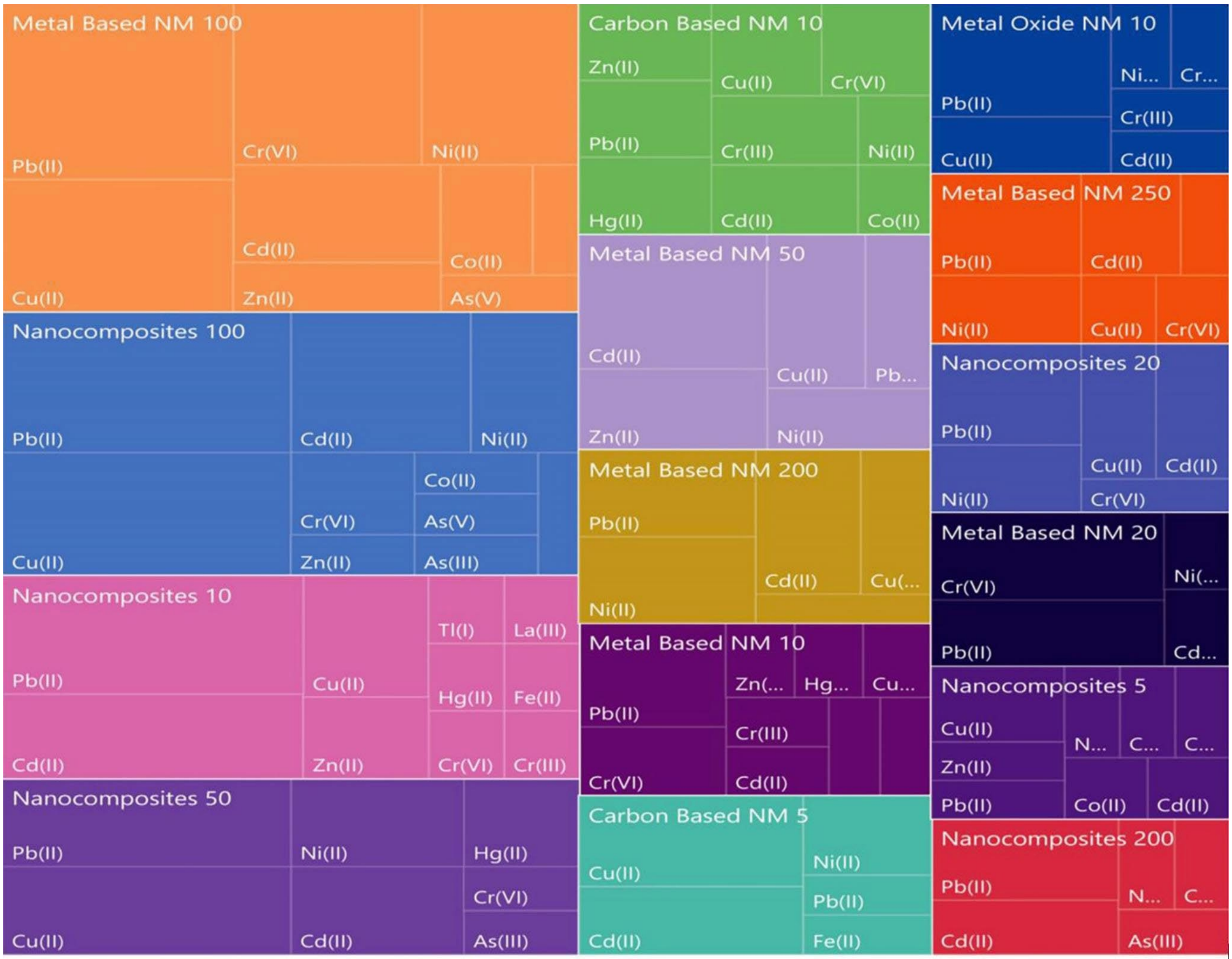


Fig. 8 Statistical expansion of the types of adsorbents used and their optimal amount (mg L-1)

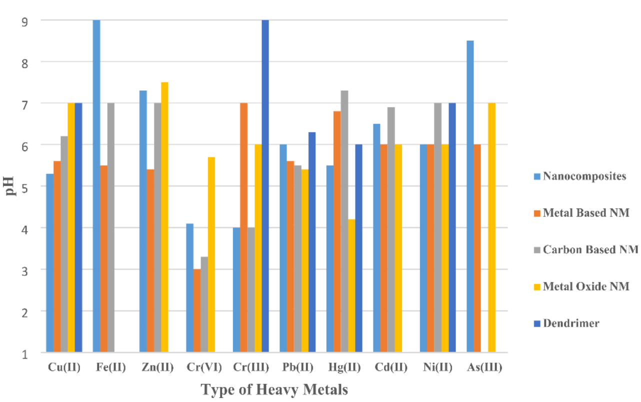


Fig. 9 The average optimum of pH value for the removal of metals with various nanostructures

The publication of articles resulting from these studies began in early 2000 and has continued with a significant upward trend since 2010. Most of the studies (90.8%) were conducted on a laboratory scale and used synthetic solutions. Compared to other countries, authors with affiliation from China have published more articles.

Compared to other metals, most studies have been done on Pb, Cd and Cu, respectively.

Different types of nanomaterials have been used in these studies. The ranking of the use of nanomaterials included: nanocomposites > metal oxide nanomaterials > metal-based nanomaterials > carbon-based nanomaterials > dendrimers, with the wide range of sizes from less than 10 nm to several hundreds of nanometers.

The application of the amount of nanoparticles in different studies has been different depending on the type of nanoparticle and the type and concentration of the metal in the aqueous solution. However, studies have shown that the amount of use of carbon-based nanoparticles is lower than other nanoparticles.

Although studies have shown that depending on the type of metal and nanoparticles, the optimum pH for absorption

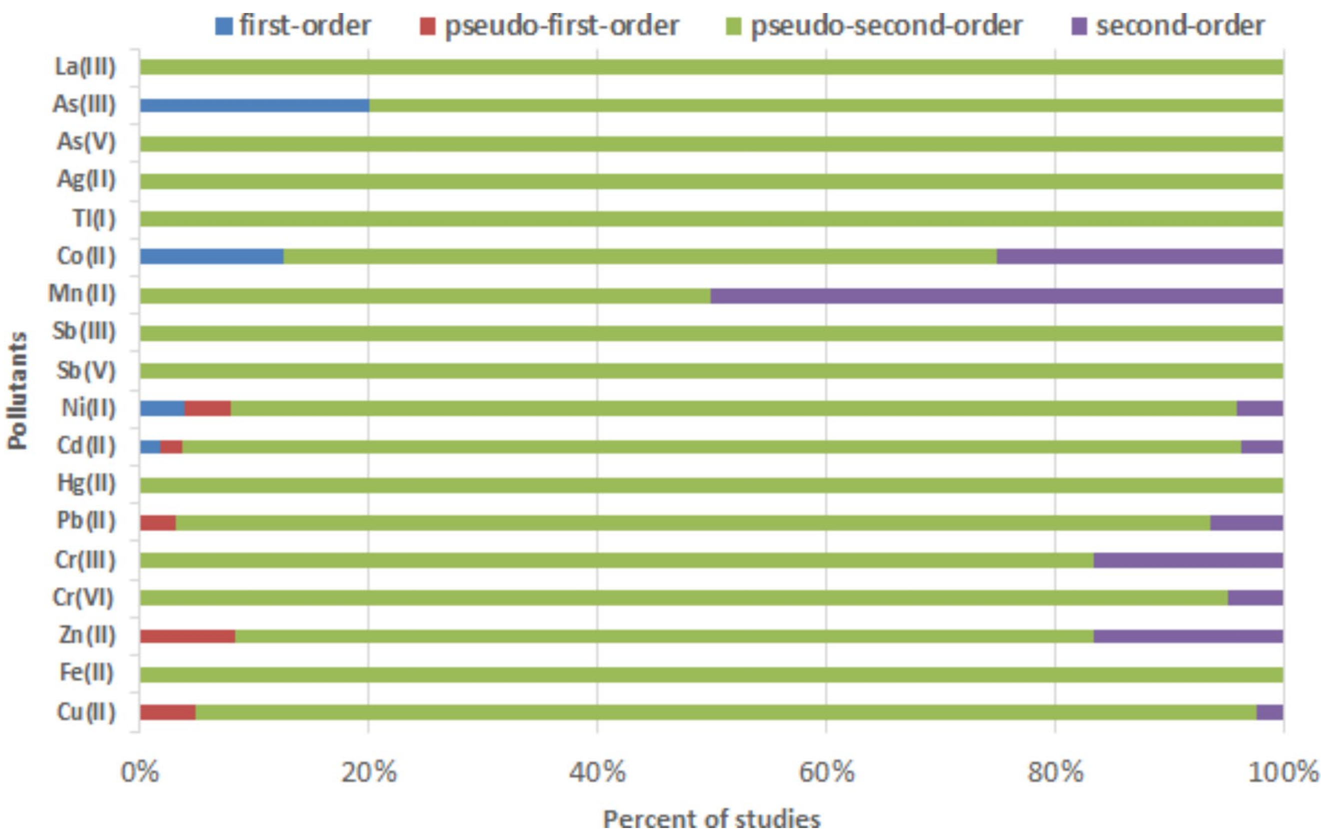


Fig. 10 Agreement of the results of studies with different degrees of reaction

is from less than 5 to 9, but in most studies, pH ≤ 7 has been reported as optimal.

Studies have shown that the absorption of heavy metals by nanoparticles mostly follows pseudo second-order and pseudo first-order reactions, respectively. Also, these studies have more agreement with Langmuir and Freundlich adsorption isotherms respectively.

The results of studies show that the synthesis and optimization of new nanomaterials with extraordinary properties can be considered as a new and competitive technology. However, more studies are needed to investigate the removal of heavy metals in real samples and to overcome some challenges in the full-scale application of nanoparticles.

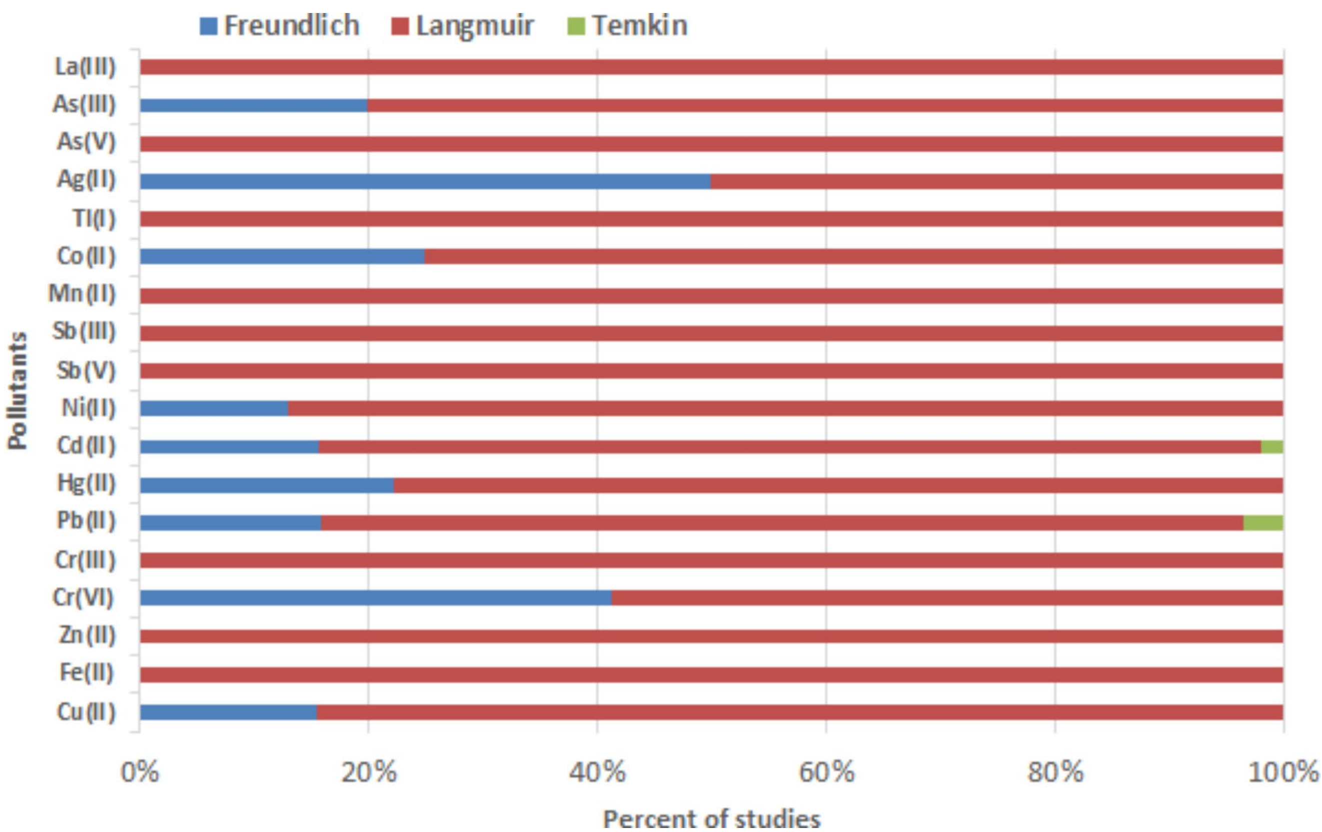


Fig. 11 Compatibility of studies results with different adsorption isotherms

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Authors' contributions Ahmadreza Yazdanbakhsh: methodology – writing – original draft and final editing, Shervin Adabi: conceptualization – search & methodology, Abbas Shahsavani: data analysis, Amir Sheikhmohammadi: review & editing and Mahdi Hadi: data Analysis- formal analysis.

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Data Availability All the necessary data have been mentioned in the paper. If other researchers need additional data, they can contact with the corresponding author.

Declarations

Ethics approval The authors of this article have covered all the ethical points, including non-plagiarism, duplicate publishing, data distortion, and data creation in this article. This project has been registered in Shahid Beheshti University of Medical Sciences with the code of ethics of IR.SBMU.PHNS. 1399.082.

Conflict of interest The authors of this article declare that they have no conflict of interests.

Consent to participate All participants signed a written consent for participation in this study.

Consent for publication Not applicable.

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