Algerian Journal of Chemical Engineering

Journal homepage: http://www.ajce.periodikos.com.br/

Review Article

Magnetic nanoparticles for the Removal of Heavy Metals from industrial wastewater: Review

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ARTICLE INFO

Article history:
Received 04 September 2020
Revised 11 November 2020
Accepted 22 January 2021

Keywords:
Heavy metals;
Wastewater;
Magnetic particles;
nanoparticles;
iron oxide;

ABSTRACT

Heavy metals contamination of industrial wastewater has a severe problem; it has significant adverse effects on human health due to their toxic nature. Nanotechnologies are opening up perspectives in different fields, medicine, environment, and electronics. Nanomaterials have developed to remove heavy metals from polluted water. In this work, we study the technique's synthesis for magnetic iron oxide particles, their applications for the removal of heavy metal, their efficiency and advantages. The perspective of nanomaterials in heavy metal water treatment and the suggestion for future research direction are discussed.

1. Introduction

Water is one of the most important natural resources in the world; the demand for water increasing rapidly in the meantime by 1% annually [1] and water scarcity problem has become an important constraint for economic development [2]. Which is vital for the survival of all living beings and the development of humans. The population increases with the acceleration of industrialization and urbanization corresponds to the economic growth and development leading.

The use of chemical compounds, industrial compounds, and agricultural wastes pose the risk of polluting the existing water sources [3]. The contamination by heavy metals from industrial wastewater to the water resources effect life significantly because heavy metal ions can accumulate biologically in the environment and into food chains [4]. For example, heavy metals could cause damage to the kidneys, mental and central nervous functions, lungs, and other organs [2, 5].

Moreover, heavy metals can also exert adverse effects on the environment and other ecological receptors, as microorganisms cannot degrade into the environment. Heavy metals are highly toxic [6], most of which are even reported to be carcinogenic [7].Therefore, the process of heavy metal removal is very important, and, has drawn tremendous attention. Up until now, numerous technologies have been developed to solve this problem. Many researchers around the world tried various technologies to remove the toxic heavy metals from aqueous solutions.

The techniques includes adsorption, biological methods, electro coagulation, electro dialysis and various membrane [8]. Ion exchange [9], adsorption [10], filtration membrane [11], electrochemical treatment [12], and so on. Besides, it is often the case that different techniques are combined for a better removal result [13, 14].

However, most of these methods have several disadvantages such as high operating costs, low selectivity, incomplete removal, and production of large quantities of wastes. Therefore, physicochemical approach by using adsorption method be proposed to solve this problem [6].

Nanoscience is one of the most important research and development in modern science, the use of nanoscale materials offers several advantages, due to their unique sizes and physical properties. In a context where technologies current allow us to manipulate matter and synthesize products at scale atomic, nanoparticles

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http://dx.doi.org/ 10.5281/zenodo.4458444

demonstrate completely different properties from same composition [15].

For several years, a new treatment method using magnetic composite materials has been under study. Much research is being done on these materials, which combine both a good adsorption capacity for pollutants and strong magnetic properties [16]. With these two characteristics, it is possible to capture a pollutant present in the water and extract it, via the magnetic material by magnetic separation.

Magnetic nanoparticles make it possible to separate fine particles, colloids and organic pollutants, which are difficult to separate by conventional methods. For this technique, the magnetic particles used (fine particle of iron oxide) are generally smaller size than the non-magnetic material. Magnetic iron oxide particles are also used to capture several heavy metals at the same time such as cadmium, lead, copper or chromium [16.17].

In recent times, the demand for magnetic absorbent materials for removing pollutants has received considerable attention due to their easy separation after using [18, 19]. The function of the surface of nanoparticles can give materials with combined characteristics for example (magnetic effect of nanoparticles with the adsorption capacity of bentonite), which will allow the attachment of a large number of ions or charged molecules [20]. A studied way making it possible to obtain a magnetic material effective in adsorption is the insertion of magnetic nanoparticles in an adsorbent.

Likewise, it is possible to introduce magnetic nanoparticles within an activated carbon particle, without modifying the specific surface area, and therefore the adsorption capacity of this material increases [21].

The purpose of this review is to present an overview of the types of iron magnetic nanoparticles used for heavy metals remediation in wastewaters. We start by describing the iron oxide nanoparticles and their condition's synthesis. Thereafter, we review the recent developments in the application of magnetic nanosorbents.

2. Nanoparticles for removing heavy metals

2.1. Definition of a nanoparticle

Recent work by the British Standards Institute (BSI, 2005), the American Society for Testing and Materials (ASTM, 2006), defined nanoparticles as synthetic particles having one or more dimensions of less than 100 nm [22]. A remark is associated with this definition: "Unique properties which differentiate the nanoparticles from the original materials, typically developed at a critical scale of 100 nm". The new properties mentioned are, therefore, entirely dependent on the fact that at the scale of nanoparticles. The physicochemical properties are different from the properties of the macroscopic solid. It due to their high (surface / volume) ratio as well as quantum size effects [23].

2.2 Advantages of nanoparticles

The decrease in the size of nnoparticles leads to a large surface area to volume ratio. Which plays a predominant role in the properties involving exchanges at the interface between the considered object and its environment. This property makes the materials more reactive from a chemical point of view, making it the object for heterogeneous catalysis applications [24]. More than the small size nanomaterials often show some properties, such as a surface effect and quantum effect. These properties contribute to their strange adsorption capacity, which are favorable for the removal of heavy metal ions.

So far, tremendous studies on nanomaterials have carried out to investigate their applications on heavy metal water treatment. They have exhibited great potential as a promising alternative to adsorbing heavy metals from wastewater [2].

Nanoadsorbents, got interest because of having nano-size, which can hold excellent rate of adsorption with short time. Furthermore, nanoadsosorbents that can use as a separation medium in water decontamination to eliminate the organic, inorganic-based pollutants [25].

Literature review shows that many efforts already done for wastewater treatment and used nanomaterials as adsorbent for efficient results. Some challenges are required addressed entirely for commercialization purpose of the nano-size adsorbents for water decontamination as well as excellent adsorption measurements, selectivity, stability of material, operational duration of material, etc. However, there is an enormous need for an active approach to treat the wastewater and fabricate some new nanoadsorbents, which applied to control toxic ions and compounds from wastewater [26].

Due to their excellent absorption, capacities to heavy metals and the simplicity of their separation from wastewater by taking advantage of both magnetite core and organic or inorganic shells. Modified magnetite nanomaterials with the core structure- envelopes have shown great potential to remove heavy metals [25]

2.3. Magnetic particles

Nanotechnologies are opening up perspectives in different fields, medicine, environment, and electronics [27]. An illustrative example of these Applications is magnetic ferrofluids. They called (colloidal suspensions), which consist of magnetic nanoparticles dispersed in a non-magnetic, aqueous or organic solvent. The magnetic nanoparticles considered are most often mixed oxides of iron (III) and another divalent transition metal M (Fe, Co, Ni, Mn, Zn) in the general form Fe_2MO_4 [28].

2. 4. Chemical composition

Two constituents go into the composition of a ferrofluid: solid magnetic particles and a carrier liquid [29-32].

a. Solid particles

Magnetic oxides, mainly ferrite particles, constitute a large proportion of particles used in ferrofluids. To obtain them, either grinding or alkalinization of an aqueous mixture carried out. The particles can also be of the metallic type, for example nickel, cobalt, iron, etc. The advantage of these particles is their strong magnetization. On the other hand, their rapid oxidation leads to the reduction or loss of this magnetization.

b. The carrier liquid

There are two types:

Organic solvents: mainly used in commercial applications, they must have high temperature stability. As examples, mention may be made of: aliphatic hydrocarbon, carboxylic diester, silicone oil, polyphenyl ether, etc. in this case the solution is called " ferrofluid surfactatants "

> Polar solvents: mainly used in medical applications. Water and alcohols are the prime examples. This is the case with "ionic ferrofluid"

2.5. Examples of some ferrofluids

The most common material is iron oxide with different stoichiometry: Fe_2O_3 or Fe_3O_4. It synthesized from Fe^{2+} or Fe^{3+} ions in a basic medium. This material is one of the cheapest available and gives rise to a whole family of materials. Indeed, in Fe_3O_4 ferrites, the Fe^{2+} cations can replace by other divalent cations such as Co^{2+}, Zn^{2+}, Mn^{2+} or Mg^{2+} as well as their alloys. We can this adjust the magnetic moment of the ferrite according to the desired properties [27]

2. 6. Iron oxides for removing heavy metals.

2. 6. 1. Hematite ($\alpha - \mathrm{Fe_2O_3}$)

Among the iron oxides, hematite ($\alpha - Fe_2O_3$) is thermodynamic stable phase for large grains (α > 20 nm).

Hematite is also formed by the enlargement of iron oxide grains (Fe_3O_4 or γ-Fe_2O_3) in heat treatments (around 400-450 °C, at atmospheric pressure) or at high pressures [33]. Hematite nanoparticles proven to effective adsorbents towards heavy metals [34–35].

Adegoke et al. investigated the effect of morphology's hematite nanoparticles on the removal of Cr (VI) [36]. Shipley et al. investigated the adsorption capacities of nano-hematite towards the cation of Pb (II), Cd (II), Cu (II), and Zn (II) [37]. The affinity between the heavy metals and the adsorbent obeyed the following order: Pb (II) > Zn (II) > Cd (II) > Cu (II). The adsorption data fitted the pseudo-second-order rate, which indicates that the adsorption rate of the adsorbent. The thermodynamic data demonstrated that the adsorption for Pb (II), Cd (II), and Cu (II) was endothermic while that for Zn (II) was exothermic.

Given that nano-hematite possesses many merits such as non-toxicity, high stability, and an excellent metal adsorption capacity, it is a promising adsorbent to treat wastewater containing heavy metals.

2. 6. 2. Magnetite (Fe_3O_4) and maghemite (γ-Fe_2O_3)

Magnetite crystallizes in a cubic system. It constructed from the faces-centered cubic arrangement of oxygen O^{2-} anions. The magnetite has also-called spinel AB_2O_4 structure (Figure 01) for (A), the atoms in tetrahedral sites surrounded by four O^{2-} ions, and (B) the atoms in octahedral sites surrounded by six O^{2-} ions [15].

The ferric ions distributed in the tetrahedral and octahedral sites when the ferrous ions are only in the octahedral sites: this called an inverse spinel structure. Its mesh parameter is a = 8.396 Å. Maghemite (γ-Fe_2O_3) nanoparticles have reported extensively to treat heavy metals, in wastewater [38–41].

The advantages of maghemite nanoparticles for treating heavy metals lie in many aspects. Akhbarizadeh et al. synthesized maghemite nanoparticles by using a single-step method and employed them to treat wastewater containing Cu (II), Ni (II), Mn (II), Cd (II), and Cr (VI) by batch method [42].

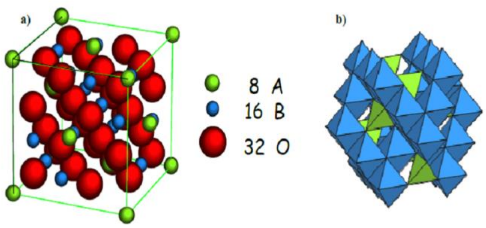

Fig 1. Representation of the crystallographic structure of an AB_2O_4 spinel structure with an origin taken from a tetrahedral site [15].

Similar to maghemite, magnetite nanomaterials easily separated from the aqueous solution after treatment by adding a magnetic field (Figure 02).

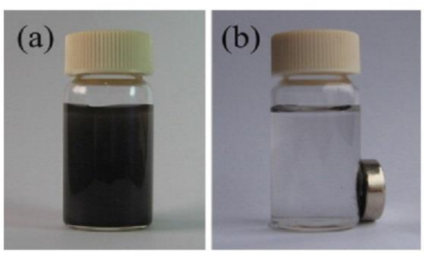

Fig 02. (**a**) Digital photograph of the Pb2+ solution with dispersed Fe_3O_4-SiO2 composite microspheres and (**b**) digital photograph of the Pb^{2+} solution after magnetic separation using an external magnetic field. Reproduced with permission from [2].

Giraldo et al. synthesized magnetite nanoparticles by using a co-precipitation method and the obtained nanoparticles were used to treat Pb (II), Cu (II), Zn (II), and Mn (II) in a batch mode [43, 44].

2.7. Conditions for the synthesis of iron oxide nanoparticles

Magnetic nanoparticles require particularly draconian conditions of synthesis and storage (inert atmosphere, anhydrous medium) excluding all immediate practical applications. We chose to study the structural and dynamic properties of suspensions of particles whose synthesis was the best controlled: maghemite.

The conditions of the five most selected synthesis methods summarized briefly in Table 01.

Table 01: Synthesis techniques for magnetic iron oxide particles [45-46]

Method Synthesis	**Co-precipitation**	**Thermal Decomposition**	**Microemulsion**	**Polyol Medium**	**Hydrothermal**
synthesis conditions	Very simple	Complicated inert atmosphere	Complicated	Very easy at solvent T °C Boiling	Simple High pressure
Temperature T(°C)	20 - 90	100 - 320	20 - 50	> 180	220
reaction Time	Minutes	Hours and Days	Hours	Hours	Hours and Days
solvent	Water	Organic	Organic	Organic	Water or Water /Ethanol
size (nm)	< 20	≤ 20	≤ 50	< 10	≤ 1000
Size Distribution	Relatively narrow	Very narrow	Relatively narrow	Very narrow	Very narrow

Control of morphology	Fair	Very good	Good	Very good	Very good
Yield	High	High	Fair	Average	Average

3. Magnetic nanoadsorbents

The application of magnetic nanoadsorbents to solve environmental problems has attracted great interest in recent years from the research.

Iron oxides are present in natural state as important buffer in soils and exert a significant control on the concentration of metal's trace in soil solution [47].

Different iron oxide nanoparticles have proved their effectiveness for the treatments of heavy metals and organic pollutants from wastewater effluents.

Metal uptake of some iron oxides nanoadsorbents are collect in table 02.

Table 02. Maximum adsorption capacities of various iron oxides sorbents for heavy metals removed [47].

Adsorbent	Heavy metal ions	Maximum adsorption capacity qmax(mg/g)	pH
Fe_3O_4	Cu(II)	11.89	5.3
Mixed magnetite-hematite	Pb(II)	617.3	
	Cr(III)	277.0	7.0
	Cd(II)	223.7	
maghmite	As(V)	50	3.0
γ-Fe_2O_3	Cr(VI)	15.6	
	As(III)	12.28	
	Cd(II)	55.19	2.5
	Cu(II)	33.09	7.0
	Ni(II)	21.51	
	Zn(II)	20.72	
Fe_3O_4	As(III)	23.8	7.0
	Pb(II)	36	5.5

Z. Mokadem et al. are investigate the ability of magnetic nanoparticles for removal of heavy metal cations. The adsorption of prepared magnetic nanoparticles toward Cu^{2+}, Pb^{2+} and Zn^{2+} ions examined as a function of various physical- chemistry conditions. The equilibrium kinetics found to be second order model of investigated system. Langmuir model have the best corroborating obtained results. Maximum adsorption capacities at pH 5.5 and 25°C was found 87.87, 167.78 and 51.20 mg.g^{-1} for Cu^{2+}, Pb^{2+} and Zn^{2+}respectively [48].

Muibat Diekola Yahya et al are developed and characterized the cobalt ferrite-supported activated carbon (CF-AC) for the removal of Cr and Pb(II) ions from tannery wastewater. The experimental conditions were estimated and reported as removal efficiency of 98.2% for Cr and 96.4% for Pb (II) ions at the optimal conditions of 5, 0.8 g, 80 min, and 333 K for pH, adsorbent dose, contact time, and temperature, respectively. It found that the Freundlich isotherm model fit better. The maximum adsorption capacities (Q_m) of Pb (II) and Cr adsorbed onto CF-AC were determined to be 6.27 and 23.6 mg/g, respectively. The adsorption process conformed well to pseudo-second order kinetics for both metals. The thermodynamic parameters showed that adsorption of Cr and Pb (II) ions onto CF-AC was spontaneous, and endothermic [49].

Zhuo-nan Huang et al. were prepared magnetic multi-wall carbon nanotubes. They were used it as adsorbents for the removal of Cr (VI) in aqueous solutions. The effects of adsorbent dosage, the concentration of Cr (VI) in aqueous solution, temperature, and pH value on the removal efficiency were studied. Results showed that the adsorption capacity of the magnetic multi-wall carbon nanotubes increased with the initial Cr (VI) concentration, but decreased with the increase of adsorbent dosage. The adsorption amount increased with contact time. The adsorption kinetics were best represented by the pseudo second-order kinetic model, and the adsorption isotherms indicated that the Langmuir model better reflected the adsorption process. The obtained calculation results for the Gibbs free energy revealed that the adsorption was a spontaneous and endothermic process. The enthalpy deviation was 3.835 kJ/mol [50]

4. Conclusion

Nanomaterials have extensively exploited to remove heavy metals, in water owing to their exceptional properties. In this work, magnetic nanoparticles were discussed. All types of the iron oxide have high efficacy, so the nano-hematite possesses such as non-toxicity, high stability, and an excellent metal adsorption capacity, it is a promising adsorbent to treat wastewater containing heavy metals.

The magnetite easily separated from the aqueous solution after treatment by adding a magnetic field. According to the various techniques of synthesis of iron oxides, it can be saw that co-precipitation is the simplest method. In the other, hand the hydrothermal method.

It showed that adsorbent developed from metal ferrite-supported activated carbon could be efficiently used as an environmentally friendly alternative adsorbent, for removal of heavy metals ions in wastewater.

The magnetic multi-wall carbon nanotubes showed significant potential for application in adsorption of heavy metal ions.

Further experimental and theoretical research are required to understand

(a) The behavior of engineered nanoparticles in real environmental conditions according to the nature, origin and compositions of treated water.
(b) The optimum regenerating systems for successive treatment cycles without losing the sorption efficiency and stability of magnetic particles.
(c) The fate of heavy metal ions after desorption under real conditions since the adsorption approach allows only the immobilization of contaminants, but not degradation

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Recommended Citation

Z. AKCHICHE, A. B. ABBA and S. SAGGAI. (2021). Magnetic nanoparticles for the Removal of Heavy Metals from industrial wastewater: Review. Algerian Journal of Chemical Engineering, 01,08-15. http://dx.doi.org/ 10.5281/zenodo.4458444