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Chapter 15

Removal of Heavy Metals Using Adsorption Processes Subject to an External Magnetic Field

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Abstract

Adsorption is a broadly used process for the removal of heavy metals and the world trend is directed to the application of new technologies to intensify existing processes. The properties of the magnetic field (intensity and arrangement) and the intrinsic magnetic properties of the adsorbent and the adsorbate are decisive for satisfactory results. The intensity of the magnetic field is important, because this implies that the greater number of spins present will align with the magnetic field according to the magnetic nature present, allowing the mobility of the adsorbate and generating heterogeneity on the surface of the adsorbent. Similarly, the arrangement of the magnetic field will determine the direction of the magnetic field lines. The application of a magnetic field as an alternative for the intensification of the adsorption process based on the consideration that the magnetic field is safe, environmentally friendly and economic.

Keywords: heavy metals, adsorption, magnetic field

1. Introduction

The importance of removing dissolved heavy metals from water is a primary concern for society because heavy metals represent a risk to both public and environmental health. Heavy metals are toxic and carcinogenic, and they can easily enter the food chain [1, 2].

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According to the Environmental Protection Agency (EPA), heavy metals are considered priority pollutants and must be eliminated or reduced from any water body that may or may not come into contact with the environment [3, 4]. There are a variety of conventional techniques capable of removing heavy metals from water, which have already been tested successfully, such as precipitation, ion-exchange, reverse osmosis, membrane separation, adsorption, and so on [5, 6]. Specifically, adsorption is one of the most promising and frequently used methods due to easy operation, high efficiency and economic benefits [7, 8]. The static magnetization coupled to an adsorption process has attracted special attention because it is an easy-to-implement, low-cost and environmentally friendly method. Recent studies have presented an improvement in the adsorption process when a magnetic field is applied [9], which is attributed to different factors and possible modifications that can change properties of the solution and material under magnetic field effect [10]. However, there are several parameters and operating conditions that are determinant within the adsorption process subjected to a static magnetic field such as the physicochemical and magnetic nature of the adsorbate and the adsorbent and the configuration and intensity of the magnetic field. The process by which the magnetic field affects the adsorbate-adsorbent system is complex, and parameters such as zeta potential, magnetic susceptibility, ionic radius and hydration of the ion have allowed to elucidate the possible mechanism.

2. Mineral adsorbents

Table 1 shows mineral adsorbents used in the removal of heavy metals from water such as Cd(II), Zn(II), Pb(II), Cu(II), Cr(VI) and Ni(II). Particularly, the Fe_3O_4/PANI/MnO_2 core-shell hybrid was the most efficient in the removal of Cd(II), showing an adsorption capacity of 154 mg/g [11]; followed by hydrotalcite-like compounds (ZnAl-diethylenetriaminepentaacetic acid and ZnAl-meso-2,3-dimercaptosuccinic acid) with values of 112.4 and 49.5 mg/g, respectively [12]. On the other hand, the metakaolin-based geopolymer showed the higher adsorption capacity of Zn(II) (74.5 mg/g) [13], in comparison with natural bentonit and bentonite (52.9 and 39.5 mg/g, respectively) and other adsorbents [11, 13, 14]. Also, for the removal of Cr(VI), the processed diatomite/MCM-41 composite was the most efficient with adsorption capacity of 70.9 mg/g [15]; followed by diatomite-supported magnetite nanoparticles with a value of 69.2 mg/g [16]. As well, different adsorbents were used to study the removal of Cu(II) and the better was the hydrotalcite-like compound ZnAl-meso-2,3-dimercaptosuccinic acid with an adsorption capacity of 69.9 mg/g [12]; this material was also used to remove Cd(II) with significant results. The Zn(II), Cr(VI), Cu(II) and Ni(II) are the heavy metals more studied with inorganic or mineral adsorbents. Respect to removal of Ni(II), natural bentonite obtained the higher adsorption capacity (50.0 mg/g) [13]. Finally, other heavy metal studied was the Pb(II) with the best value of 404.0 mg/g again using the hydrotalcite-like compound ZnAl-meso-2,3-dimercaptosuccinic acid [12].

Adsorbent	Removed species	Adsorption capacity (mg/g)	Ref
Fe_3O_4/PANI/MnO_2 core-shell hybrids	Cd(II)	154	[11]
	Zn(II)	[a]88.0 (17.6 mg/g)	
	Pb(II)	[a]99.1 (19.8 mg/g)	
	Cu(II)	[a]99.1 (19.8 mg/g)	
Processed diatomite/MCM-41 composite	Cr(VI)	70.9	[15]
Diatomite-supported magnetite nanoparticles	Cr(VI)	69.2	[16]
Tannin-immobilized activated clay	Cr(VI)	24.1	[17]
Modified natural zeolites	Cr(VI)	3.5–8.8	[18]
Modified rectorite	Cr(VI)	21	[19]
geopolymer/alginate hybrid spheres	Cu(II)	60.8	[20]
Aminopropyltriethoxysilane/sepiolite	Cu(II)	8.9	[21]
Multiwalled carbon nanotubes	Cu(II)	50.3	[22]
Zeolite	Cu(II)	9.0	[23]
Ca-alginate	Cu(II)	42.7	[24]
Bentonite/chitosan	Cu(II)	9.8	[25]
Inorganic-organic clays	Cr(VI)	16.6–17.9	[26]
Hydrotalcite-like compound ZnAl-NO_3			[12]
ZnAl-dtpa (diethylenetriaminepentaacetic acid)	Cu(II)	48.3	
	Pb(II)	145.0	
	Cd(II)	49.5	
ZnAl-dmsa (meso-2,3-dimercaptosuccinic acid)	Cu(II)	69.9	
	Pb(II)	404.0	
	Cd(II)	112.4	
Na-bentonite	Zn(II)	23.1	[13]
Polyphosphate-modified kaolinite clay	Zn(II)	27.8	[13]
Natural zeolite	Zn(II)	13.4	[13]
Kaolinite	Zn(II)	7.2	[13]
Metakaolinite	Zn(II)	12.4	[13]
Vermiculite	Zn(II)	31.7	[13]
Bentonite	Zn(II)	39.5	[13]
Zeolite	Zn(II)	18.7	[13]
Natural bentonite	Zn(II)	52.9	[13]

Adsorbent	Removed species	Adsorption capacity (mg/g)	Ref
Metakaolin-based geopolymer	Zn(II)	74.5	[13]
Vermiculite	Ni(II)	25.3	[13]
Natural bentonite	Ni(II)	50.0	[13]
Zeolite	Ni(II)	1.98	[13]
Na-bentonite	Ni(II)	24.2	[13]
Ca-bentonite	Ni(II)	6.3	[13]
Natural clinoptilolite	Ni(II)	8.7	[13]
Montmorillonite	Ni(II)	21.1	[13]
Metakaolin-based geopolymer	Ni(II)	42.6	[13]
Mixed silica-alumina oxide	Cu(II)	0.1	[14]
	Ni(II)	0.2	
	Zn(II)	4.2	

[a]Removal percentage.

Table 1. Mineral adsorbents used in the adsorption of heavy metals from water.

3. Mineral carbons

The mineral carbons used to study adsorption of heavy metals from water are peat, lignite, anthracite, graphite and principally the bituminous carbon, which is the most used. The bituminous carbon has been physically activated with steam and modified by impregnation with calcium, phosphoric acid, sulfur and nitric acid, and subsequently, a thermal treatment has been applied [27–32].

Table 2 concentrates the mineral carbons used in the adsorption of heavy metals from water reported in the literature. There are few reports about the use of this kind of adsorbents and

Mineral carbon	Modification method used	Modifying agent	Removed species	Adsorption capacity (mg/g)	Ref
Bituminous carbon	Impregnation-thermal treatment	Calcium-H_3PO_4	Zn(II)	3.4	[27]
			Cd(II)	8.0	
			Ni(II)	5.0	
Bituminous carbon	—	—	Cd(II)	5.1	[28]
			Cu(II)	13.3	
			Cr(VI)	10.5	
Bituminous coal (AC-109)	Impregnation-thermal treatment	Sulfur	Hg^0	1.6	[29]
			Hg(II)	0.7	

Mineral carbon	Modification method used	Modifying agent	Removed species	Adsorption capacity (mg/g)	Ref
Bituminous coal (AC-107)	Impregnation-thermal treatment	Sulfur	Hg^0	1.9	[29]
			Hg(II)	0.7	
Bituminous coal (AC-C2)	Impregnation-thermal treatment	HNO_3	Hg^0	1.8	[29]
			Hg(II)	0.7	
Bituminous charcoal	Physical activation	Steam	Cu(II)	[a]0.1	[30]
			Cr(VI)	[a]0.3	
			Zn(II)	[a]0.3	
Lignite charcoal	Physical activation	Steam	Cu(II)	[a]0.1	[30]
			Cr(VI)	[a]0.3	
			Zn(II)	[a]0.3	
Bituminous coal	Physical activation	Water vapor	Cd(II)	1.7	[31]
			Zn(II)	2.0	
Bituminous carbon	Impregnation-thermal treatment	Calcium-H_3PO_4	Zn(II)	1.6	[32]
			Cd(II)	2.7	
			Ni(II)	2.3	

[a]Industrial effluent.

Table 2. Mineral carbons used in the adsorption of heavy metals from water.

the bituminous carbon is the most studied. Specifically, the better adsorbent to remove Zn(II), Cd(II) and Ni(II) is the bituminous carbon modified with calcium and H_3PO_4 (3.4, 8.0 and 5.0 mg/g, respectively) [27]. On the other hand, the unmodified bituminous carbon shows the higher adsorption capacity of Cu(II) and Cr(VI) with values of 13.3 and 10.5 mg/g, respectively [28]. Additionally, it is important to mention that bituminous charcoal and lignite charcoal were used in adsorption studies of a metallurgic effluent containing Cu(II), Cr(VI) and Zn(II), and the adsorption capacities were low [30]. Finally, Hg^0 and Hg(II) were studied employing bituminous coal modified with sulfur and HNO_3 and adsorption capacities of 1.9 and 0.7 mg/g, respectively, were obtained [29].

4. Magnetic adsorbents

The term "***magnetic adsorbent***" is commonly used to describe the materials derived from iron (particles of γ-Fe_2O_3, Fe_3O_4 and $CoFe_2O_3$) or composites prepared from different types of matrix modified with iron compounds, which are used in the adsorption of water contaminants [33]. Particularly, for the adsorption of heavy metals from water, the principal magnetic adsorbents used are the composites, which are prepared with different methodologies using iron compounds and a specific matrix such as activated carbon, biochar, carbon nanotubes, α-cellulose, chitosan, cotton, resins, silica, starch, polymers, different type of biomass, and so on (see **Table 3**) [34–55]. The methods used in the preparation of magnetic composites can be

Matrix	Modified method	Iron compounds	Removed species	Adsorption capacity (mg/g)	Ref
Activated carbon	Co-precipitation	$FeSO_4·7H_2O$, $FeCl_3·6H_2O$	Cu(II)	[a]99.8	[34]
Activated carbon	Co-precipitation	$Fe(NO_3)_3·9H_2O$	As(III)	38.8	[35]
			As(V)	51.3	
Activated carbon	Co-precipitation	$FeCl_2$	As(V)	1.95	[36]
Activated carbon	Reduction reaction	FeO_4S	As(III)	18.19	[37]
			As(V)	12.02	
Carbon nanotubes	Co-precipitation	$FeSO_4·4H_2O$, $FeCl_3·6H_2O$	Ni(II)	51.62	[38]
Biochar from *Astragalus mongholicus*	Co-precipitation	$FeSO_4·4H_2O$, $FeCl_3·6H_2O$	Cr(VI)	44.74	[39]
α-Cellulose	Extrusion dropping technology	Fe_3O_4	Cu(II)	47.573	[40]
			Pb(II)	37.994	
			Zn(II)	20.800	
Chitosan	Co-precipitation	$FeCl_3·6H_2O$, $FeSO_4$	Cu(II)	188	[41]
			Cd(II)	159	
			Zn(II)	72	
			Cr(VI)	46	
Chitosan	Hydrothermal	$FeCl_3·6H_2O$	Pb(II)	97.97	[42]
			Cu(II)	83.65	
Chitosan	Crosslinking method	Fe_3O_4	Cu(II)	143.27	[43]
Cotton	Impregnation-thermal treatment	$Fe(NO_3)_3·9H_2O$	Cr(VI)	3.74	[44]
Cyclosorus interruptus	Co-precipitation	$FeSO_4·7H_2O$, $FeCl_3·6H_2O$	Pb(II)	90.1	[33]
Dowex 50 WX4 resin	Co-precipitation	$FeCl_3·6H_2O$, $FeCl_2·4H_2O$	Cr(VI)	400	[45]
			Ni(II)	384	
			Cu(II)	416	
			Cd(II)	398	
			Pb(II)	380	
Graphene oxide	Co-precipitation	$FeSO_4·7H_2O$, $FeCl_3·6H_2O$	Hg(II)	400	[46]
Macroalgal biomass (Kelp)	Impregnation-thermal treatment	$FeCl_3·6H_2O$	Cd(II)	34.89	[47]
			Cu(II)	46.74	
			Zn(II)	95.49	

Matrix	Modified method	Iron compounds	Removed species	Adsorption capacity (mg/g)	Ref
Mesoporous silica	Thermal decomposition	$FeCl_3 \cdot 6H_2O$	Fe(III)	[a]79.9	[48]
Palygorskite	Co-precipitation	$FeCl_3 \cdot 6H_2O$, $FeCl_2 \cdot 4H_2O$	Pb(II)	26.7	[49]
Pine bark	Impregnation-thermal treatment	$Fe(NO_3)_3 \cdot 9H_2O$	Pb(II)	25.294	[50]
			Cd(II)	14.960	
Poly p-phenylenediamine	Co-precipitation	$FeCl_3 \cdot 6H_2O$, $FeSO_4 \cdot 7H_2O$	As(V)	35.14	[51]
Poly(vinylidene fluoride)	Impregnation-thermal treatment	$Fe(NO_3)_3 \cdot 9H_2O$	Cr(VI)	1423.4	[52]
Silica	Co-precipitation	$FeCl_2 \cdot 4H_2O$, $FeCl_3 \cdot 6H_2O$	Pb(II)	270	[53]
			Hg(II)	303	
			Pb(II)	256.4	
Starch	Crosslinking method	Fe_3O_4	Pb(II)	[a]83.1	[54]
			Cu(II)	[a]66.5	
Waste orange peel	Co-precipitation	$FeSO_4 \cdot 7H_2O$, $FeCl_3 \cdot 6H_2O$	As(III)	11.12	[55]

[a]Removal percentage.

Table 3. Magnetic composites used in the adsorption of heavy metals and metalloids from water.

grouped in the following categories according to the data reported in the literature [56]: (1) chemical co-precipitation, (2) thermal decomposition, (3) hydrothermal, (4) polyol process, (5) sol-gel and (6) chemical reduction. In this context, it is relevant to mention that the chemical co-precipitation is the most used method for the preparation of magnetic composites and this can be carried out in different routes, which include the following: (1) mixture of iron solutions ($FeSO_4 \cdot 7H_2O$ and $FeCl_3 \cdot 6H_2O$ or $FeCl_3 \cdot 6H_2O$ and $FeCl_2 \cdot 4H_2O$ or $Fe(NO_3)_3 \cdot 9H_2O$) with the matrix, and then, the addition of ammonium or sodium hydroxide or $NH_3 \cdot H_2O$ to precipitate the iron nanoparticles on the matrix surface [33, 34, 36, 38, 39, 41, 45, 49, 55]. (2) The preparation of nanoparticles of iron (Fe_3O_4) by precipitation using $FeCl_3 \cdot 6H_2O$ and $FeCl_2 \cdot 4H_2O$ with additives such as $CaCl_2 \cdot 2H_2O$, $Na_3PO_4 \cdot 12H_2O$, EDTA [57, 58]. (3) First, the precipitation of nanoparticles of iron (Fe_3O_4), second, the mixture of Fe_3O_4 with materials such as *Cyclosorus interruptus*, silica and p-phenylenediamine-thiourea-formaldehyde polymer; and finally, the addition of 3-aminopropyltriethoxysilane or tetraethyl orthosilicate or potassium persulfate, depending on the final application of composite [33, 51, 53].

The second most popular method used in the preparation of magnetic composites includes the impregnation of the matrix with iron compounds such as $Fe(NO_3)_3 \cdot 9H_2O$ and $FeCl_3 \cdot 6H_2O$ and a thermal treatment of impregned materials using drying on a regular oven overnight [44], pyrolysis in conventional systems [47, 50, 52, 59] or pyrolysis in microwave system [60].

Table 3 summarizes the different types of magnetic composites used in the removal of some heavy metals and metalloids from water such as Pb(II), Cu(II), Ni(II), Cd(II), Zn(II), Cr(VI), Fe(III), Hg(II), As(II) and As(V). In general, the composites prepared with the resin Dowex 50 WX4 were the most efficient in the removal of Ni(II), Cu(II), Cd(II) and Pb(II), showing an adsorption capacity of 384, 416, 398 and 380 mg/g, respectively [45]. These values are higher than the data registered for other composites obtained from carbon nanotubes [34], α-cellulose [40], chitosan [41–43], silica [53], starch [54] and different types of biomass such as waste orange peel, pine bark and *Cyclosorus interruptus* (see **Table 3**) [33, 50, 55]. Additionally, it is relevant to mention that for the removal of Cr(VI), the composite obtained with poly(vinylidene fluoride) shows the higher adsorption capacity (1423.4 mg/g) in comparison with the samples prepared from chitosan (46 mg/g), cotton (3.74 mg/g) and Dowex 50 WX4 resin (400 mg/g) [41, 44, 45]. In this context, the adsorption of As(III) and As(V) has also been studied employing magnetic composites prepared with activated carbon principally [35–37]. For example, the composite prepared with bituminous coal carbon is showing a high adsorption capacity of As(III) and As(V) (38.8 and 51.3 mg/g, respectively), in comparison with the materials prepared with an inorganic matrix as the poly p-phenylenediamine (34.15 mg of As(V)/g) [35, 51]. Finally, it is important to mention that one of the most toxic heavy metals, Hg(II), was studied employing magnetic composites obtained with silica and with graphene oxide, highlighting the latter material with an adsorption capacity of 400 mg/g [46].

In summary, the adsorption of heavy metals has been studied using magnetic composites prepared principally by co-precipitation method, however, a homogeneous comparison is not possible because all the adsorption conditions and physicochemical properties of magnetic adsorbents are different (porosity, chemical functionally, etc.). But, the investigation in this area is increasing in past years and the data reported in this chapter will serve as a basis for future publications.

5. Adsorbate properties and adsorption conditions

The adsorption of heavy metals on different adsorbents occurs via ion-exchange, complexation, coordination, chelation, microprecipitation, electrostatic interaction and/or combination of these mechanisms [61]. Consequently, these adsorption mechanisms are defined by the physicochemical characteristics of adsorbents (texture and chemical functionality), the adsorption conditions (pH, temperature, mass of adsorbent, treated volume, type of adsorption system, etc.) and the properties of adsorbates. Particularly, the principal properties of heavy metals considered in the adsorption mechanism are as follows: (1) ionic radius, (2) electronegativity, (3) atomic weight, (4) hydration radius, (5) oxidation number and (6) electrostatic potential.

According to the data reported in the literature, the ionic radius and the electronegativity are the two principal properties used in the interpretation of heavy metal adsorption mechanisms. For example, **Figure 1** shows the adsorption results of Pb(II), Cd(II), Cu(II), Ni(II) and Zn(II) as function of ionic radius. The adsorbents used were the red marine alga *Kappaphycus*

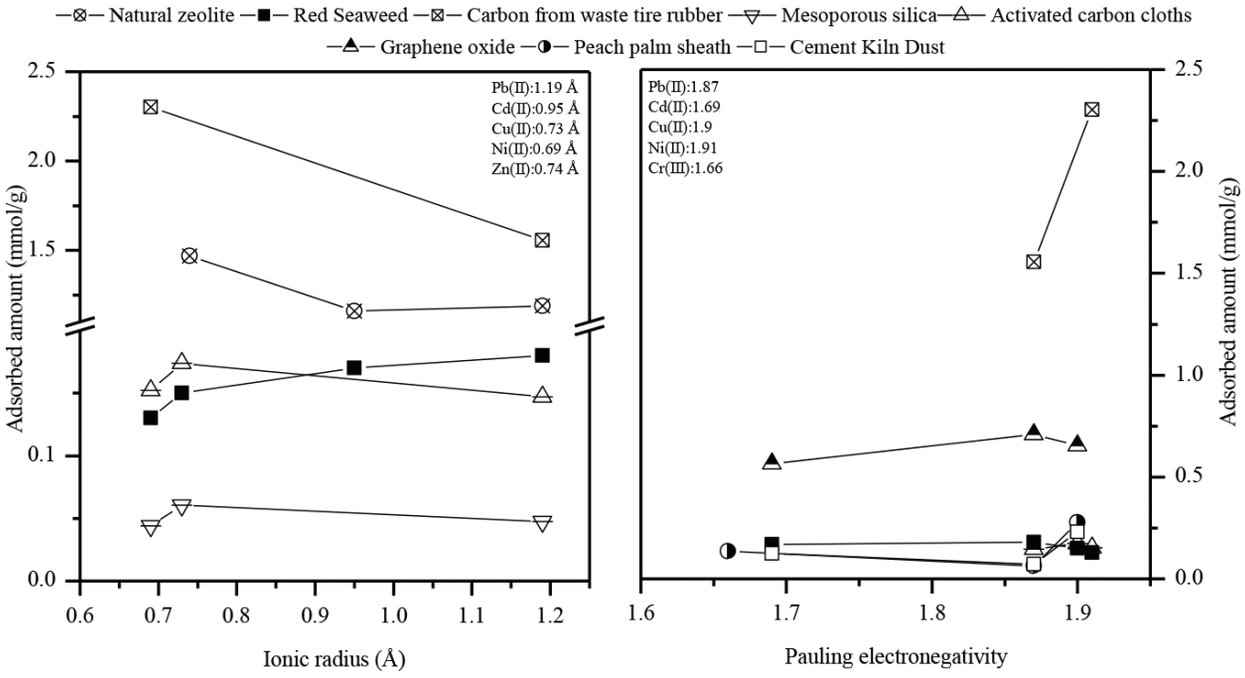


Figure 1. Adsorption results of heavy metals from water of different adsorbents in function of ionic radius and Pauling electronegativity.

alvarezii, carbon from waste tire rubber, mesoporous silica, natural zeolite and activated carbon cloths [61–65]. In general, a clear behavior is not defined and only in the adsorption of Pb(II), Cd(II), Cu(II) and Ni(II) on red marine alga, a clear correlation was obtained between the adsorbed amount of metal ions and the ionic radius. In this case, the adsorbed amount was higher for the heavy metal with greater ionic radius (Pb:1.19 Å), showing a maximum adsorption capacity of 0.51 mmol/g by the mechanism of electrostatic interaction [61]. The opposite behavior was observed for the adsorption of Pb(II), Cd(II), Ni(II) and Zn(II), using natural zeolite and carbon prepared from waste tire rubber as an adsorbent, where the adsorbed amount decreased as the ionic radius increased [62, 68]. Also, the hydration radius has been used to understate the adsorption of heavy metals and the results also do not follow a specific trend [66].

On the other hand, the Pauling electronegativity of heavy metals has also been correlated with the adsorption of heavy metals. **Figure 1** shows the adsorption results of Pb(II), Cd(II), Cu(II), Ni(II) and Cr(III) employing different adsorbents in function of Pauling electronegativity [61, 62, 65, 67–69]. Particularly, it is interesting to note that a defined behavior was observed in the adsorption of Pb(II) and Ni(II) using the carbon from waste tire rubber as an adsorbent [62]. Specifically, this adsorbent has the higher adsorption capacity of Ni(II) and Pb(II), and the adsorption was higher for the more electronegative metal. This behavior can be associated with the chemical functionality of carbon because this material was activated with hydrogen peroxide and consequently, the presence of O─H group of phenols and C═O group of esters favored the adsorption of heavy metals [62]. Additionally, for the adsorption of Pb(II), Cd(II), Cu(II) and Cr(III) on cement kiln dust and peach palm sheath, the

electronegativity does not keep a relation with the absorbed quantity of metals [68, 69]. However, it is important to mention that the Cr has a different oxidation number (III) in comparison with the other heavy metals (II). In this context, there are few reports that have studied the effect of oxidation number of heavy metals in the adsorption process and particularly, the works related with the adsorption by complexation mechanism have considered this property. For example, in the removal of Ni^{2+}, Fe^{2+} and Fe^{3+} (with external hydroxyl functional groups) was demonstrated that high-valence species complex more quickly than the low-valence due to the ability to accept more of the ligand's electron donation, where the adsorption followed the trend $Ni^{2+} > Fe^{3+} > Fe^{2+}$ [70].

Finally, it is relevant to mention that there are many factors which affect heavy metal adsorption such as initial concentration, temperature, adsorbent dose, pH, contact time and stirring speed. However, the pH plays a vital role in deciding the maximum adsorption capacity of the adsorbent such as activated carbons, carbon nanotubes, graphene, biosorbents, low-cost adsorbents (vegetal and industrial wastes), silica, chitosan, zeolites, alumina, clay, algal biomass, red mud, magnetic composites, and so on. The pH affects the surface charge of the adsorbent, the degree of ionization and speciation of the surface functional groups and metal ions [71–74]. In general, the adsorption of metallic species increases with increasing pH in certain range because at low pH, there a competition between the metallic species and the H^+ ions of aqueous solution.

6. Magnetic ordering and zeta potential

The behavior of the materials when they are exposed to a magnetic field is different, depending on their physicochemical and magnetic nature, this behavior can be observed in **Table 4**. [75].

On the other hand, superparamagnetism is observed in very small particles of transition metals and their compounds, particularly their oxides. It can be used to characterize fine dispersions of metal, alloy and their oxides and has applications in several areas. Parameters such as the magnetization per cubic centimeter or magnetization per gram are better parameters than the susceptibility for describing superparamagnetism and magnetically ordered ferromagnetic and ferromagnetic materials. The applied magnetic field is expressed in oersteds (Oe). Furthermore, the magnetic moment in ferromagnetic and ferromagnetic materials refers to the saturation moment and not the effective moment. Ferromagnetic metals, such as Fe, Co, Ni and insulators such as γ-Fe_2O_3 and Fe_3O_4, show the well-known hysteresis curve; in their unmagnetized state, the unpaired electrons associated with each atom have a net magnetic moment or magnetization, which is the vector sum of all unpaired electrons in that domain. In this sense, it is important to explain that when ferromagnetic and ferromagnetic materials are heated above a critical Curie temperature, they change over to paramagnetic behavior; thus the hysteresis disappears. In contrast, ideal superparamagnetic systems, when sufficiently cooled below a critical blocking temperature will experience a very slow relaxation time. Their net magnetic moment will align parallel to the applied field and appear to behave as if they had an apparent ferromagnetic behavior. This aspect will result in hysteresis of

Magnetic behavior	Description
Diamagnetic	A magnetic field acting on any atom induces a magnetic dipole for the entire atom by influencing the magnetic moment caused by the orbiting electrons. These dipoles can oppose the magnetic field, causing the magnetization to be less than zero. A diamagnetic behavior gives a negative susceptibility. Materials such as copper, silver, gold and alumina are diamagnetic at room temperature
Paramagnetic	A net magnetic moment due to electronic spin is associated with each atom when materials have unpaired electrons. When a magnetic field is applied, the dipoles line up with the field, causing a positive magnetization. Because the dipoles do not interact, extremely large magnetic fields are required to align all of the dipoles; so, the effect is lost as soon as the magnetic field is removed. This behavior can be observed in metals such as aluminum and titanium
Ferromagnetic	This type of behavior is promoted by the unfilled energy levels in the *3d* level of iron, nickel and cobalt. In this kind of materials, the permanent unpaired dipoles easily line up with the imposed magnetic field due to the exchange interaction. Large magnetizations are obtained even for small magnetic fields
Antiferromagnetic	In this behavior, the magnetic moments produced in neighboring dipoles line up in opposition to one another in the magnetic field, even though the strength of each dipole is very high. Examples of these materials are manganese, chromium, MnO and NiO
Ferrimagnetic	Different ions have different magnetic moments, like in ceramic materials. In a magnetic field, the dipoles of ion *A* may line up with the field, while dipoles of ion *B* oppose the field. But because the strengths of the dipoles are not equal, a net magnetization results. Ceramics called ferrites perfectly represent this behavior

Table 4. Classification and magnetic behavior of materials.

"apparent" ferromagnetic behavior. Conversely, above the critical blocking temperature, the hysteresis will disappear and the clusters will show a unique curve with no hysteresis [76].

It is possible to classify the materials from the magnetic point of view as a function of the number of Bohr magnetons, whose parameter can be calculated from the Langevin function, first derived for the noninteracting paramagnetic spins and to the noninteracting:

$$\sigma/\sigma_s = coth(\mu_c H/kT) - (kT/\mu_c H) \quad (1)$$

where σ is the magnetization per gram; σ_s is the magnetization of saturation; μ_c is the magnetic moment of the cluster; H is the applied magnetic field; k is the Boltzmann constant; T is the temperature (K).

A parameter used to corroborate the effect of the magnetic field on an adsorbate-adsorbent system is the zeta potential. Some studies have reported this parameter, such as Zhang et al. in 2004 where the zeta potential was measured in magnetized and non-magnetized Ca-rectorite suspensions observing that in the magnetized solutions the zeta potential was larger than in the nonmagnetized ones when the Cu concentration was zero. In contrast, the magnetic treatment reduced the zeta potential in Ca-rectorite suspensions containing Cu [77]. This same behavior was observed in a study made with Na-rectorite dispersions magnetized and not magnetized in the presence and absence of Zn [78].

In the literature, there is a little information about the mechanism of magnetic effects on adsorbate-adsorbent systems. However, one of the clearest suggests that magnetic exposure alters the arrangement of water molecules, ions and hydrated ions adsorbed on the surface of the particle in such a way that the effective adsorbed layer becomes thicker, then the slipping plane shifts outward from the particle surface so that the magnitude of zeta potential will be reduced, and the apparent size of the particles becomes larger so that their diffusivity will be reduced [79].

7. Importance of the physicochemical and magnetic properties of the adsorbates in the adsorption process assisted by an external magnetic field

The physicochemical properties of adsorbates play a very important role in the adsorption phenomenon in general. Properties such as ionic radius, electronegativity, valence, charge and hydration number are considered fundamental for the selection of the adsorbent material and the understanding of the adsorption mechanism, since these parameters help to determine forces and mobilities [80]. In particular, adsorbates such as heavy metals are present in a positive ionic state, this implies that there is an instability in charges; however, being dissolved in water, they remain stable in the solution because they balance their charge with the electronic cloud of oxygen present in the water molecule, which is highly negative, thus the ion is surrounded by water molecules quasi-stabilized; this phenomenon is called ion hydration. The hydration of the ion is fundamental in the adsorption process, since it establishes the adsorption force that each of the ions will have on the surface of an adsorbent material, in addition, it determines the advantage over other ions to occupy active sites, which explains the Gouy-Chapman theory.

The Gouy-Chapman theory states that ions of equal charge are adsorbed with equal force on the surface of the adsorbent; however, the hydration radius determines the adsorption strength, being those with lower hydrated radius more strongly retained; this implies an increase in the size of the ion, reducing its mobility [81]. Based on this, the hydration of each ion will be different, since it will be surrounded by a certain number of water molecules depending on the size of the ion and its charge, and this number of water molecules surrounding the ion is called the number of water molecules of hydration.

To know the hydration number of an ion, it is necessary to study the mobility of the ion within the solution, this mobility is mainly affected by two forces: the electric force Fe (Eq. (2)) and the viscous force Fv (Eq. (3)):

$$Fe = z_i e_0 x \tag{2}$$

where $z_i e_0$ is the electric charge; x is the electric field.

$$Fv = 6\pi r \eta v \tag{3}$$

where r is the ionic radius; η is the viscosity; v is the ion speed.

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The phenomenon of hydration is explained when the ion reaches a constant speed, which implies that the electric and viscous forces are equalized (Eq. (4)).

$$r = \frac{z_i e_0 x}{6\pi\eta v} \tag{4}$$

Bearing in mind that the electric mobility u is the result of the velocity ratio on the electric field (Eq. (5)), we find the hydrated radius r_h (Eq. (6)):

$$u = \frac{v}{x} \tag{5}$$

$$r_h = \frac{z_i e_0}{6\pi\eta u} \tag{6}$$

Finally, the hydration number n_h is found by differentiating the hydrated radius r_h with the crystallographic radius of the r_{Cris} ion divided by the molecular radius of the water r_{H_2O} (Eq. (7)):

$$n_h = \frac{r_h^3 - r_{Cris}^3}{r_{H_2O}^3} \tag{7}$$

Ionic hydration is mainly due to thermodynamic phenomena [82]; however, studies have been reported where changes in the hydration of the ions attributable to exposure to an external magnetic field were observed, including variation in ion displacement and complex formation [83]. Alterations in ionic hydration due to exposure to a magnetic field can have a greater or lesser effect depending on the thickness of the hydration layer of each ion and the thermodynamic conditions of hydration, suggesting that magnetic field treatment causes changes in the structure of the water that hydrates the ions [84]. It is theorized that the magnetic field favors the hydration of ionic species due to the creation of normal Lorentz forces that help to penetrate the boundary layer of natural hydration generating a molecular arrangement that allows greater hydration [85]. In addition, the transport of the adsorbates to the active sites is promoted due to the impulse generated by the Lorentz forces and the electrostatic forces of Van der Waals, achieving an advantage over viscous forces [86]. On the other hand, it has been shown that divalent cations, such as heavy metals, are more susceptible to the presence of a magnetic field, since they seem to promote their polarization, as well as showing a "magnetic memory" effect that it can break when the temperature increases [87, 88], and at the same time, it has been observed that the magnetic memory effect can disappear nonlinearly in time depending on the configuration of the applied magnetic field and the exposure time to the magnetic field [89]. The alteration in the polarity of the hydrated ions due to the exposure to a magnetic field also generates changes in the zeta potential (ζ) of the solutions, influencing the pH values and encouraging the adsorption of metallic species [77–79, 90]. **Figure 2** explains the improvement in ionic hydration under the influence of a magnetic field which is due to a better accommodation of the water molecules surrounding the ion, not only in the first hydration layer, but also in the second layer although with a less force than in the first.

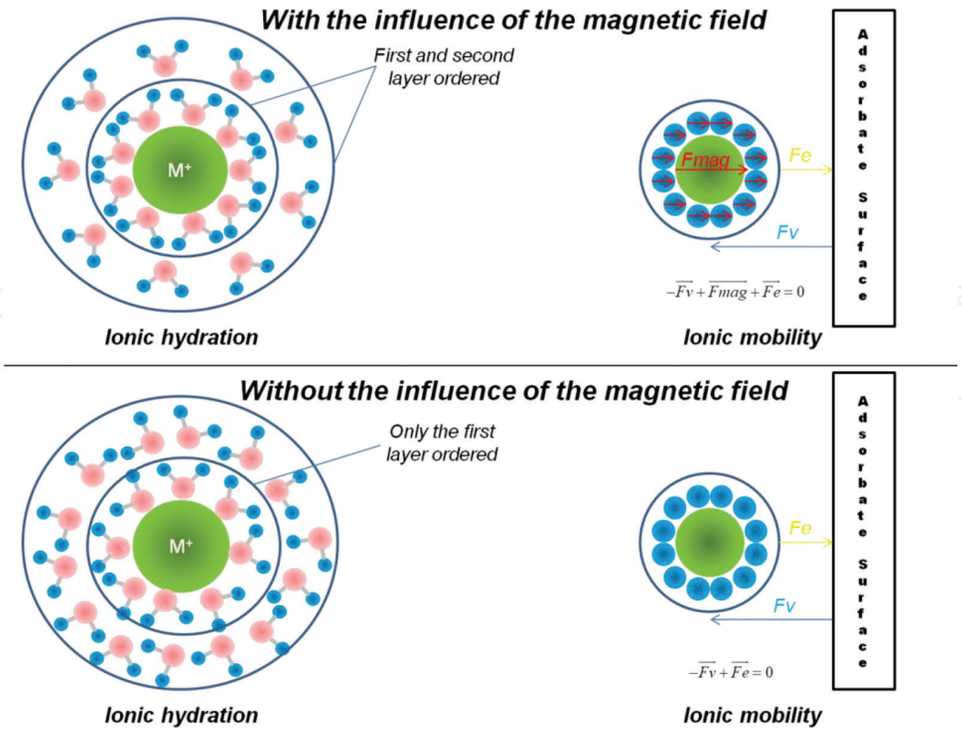


Figure 2. Mechanism of adsorption of heavy metals with and without the presence of a magnetic field.

The improvement in mobility of the hydrated ion under the influence of the magnetic field is due to the fact that the magnetic susceptibility of both the ion and the water molecules generates a component of magnetic force (*Fmag*) that added to the electrostatic force (*Fe*), which manage to overcome the viscous force (*Fv*), making the ion more likely to collide with the surface of the adsorbent.

The interactions that occur between the ions and the magnetic field can be explained by understanding the magnetic susceptibility; this is an intrinsic property that has all the matter. Magnetic susceptibility (χ_{Mag}) is a magnitude that represents the sensitivity to the magnetization of a material influenced by a magnetic field, determined by the sum of a diamagnetic component or diamagnetic susceptibility (χ_{Dia}) and a paramagnetic component or paramagnetic susceptibility (χ_{Para}) [91] (Eq. (8)), understanding that the diamagnetic matter opposes the flow of the magnetic field and the paramagnetic matter is oriented in the same direction of the magnetic field.

$$\chi_{Mag} = \chi_{Para} + \chi_{Dia} \tag{8}$$

Substances that are magnetized in the same direction of the magnetic or paramagnetic field have an odd electronic orbital and their magnetic susceptibility is positive, while those that are magnetized against the magnetic field or diamagnetic have their electronic orbital even and have negative magnetic susceptibility [92, 93], and each substance has a different value of magnetic susceptibility, inclusive, the same substance with different oxidation state can vary in its magnitude of magnetic susceptibility. In the case of ionic species such as heavy

metals, they have distinct values of magnetic susceptibility between them, coupled with this, the water molecules that surround a hydrated ion also have a magnetic susceptibility, so as a whole, generates a new magnitude of magnetic susceptibility, being the sum of the susceptibility of the solitary ion and that of each of the water molecules that surround it in the main layer. Based on this, the net susceptibility of the hydrated ion will depend on the magnetic susceptibility value of the ion χ_{Mag} and the multiplication of the magnetic susceptibility of water χ_{H_2O} by the hydration number n_h (Eq. (9)).

$$\chi_h = \chi_{mag\ ion} + \left[(n_h)(\chi_{H_2O})\right] \tag{9}$$

As an example, the susceptibilities of three heavy metals are shown in **Table 5**: Pb, Cd and Zn, where the Pb has a magnetic susceptibility lower than that of the Cd and Zn; contrary to the values of ionic radio and electronegativity; however, we found that the hydration index n_h of Pb is lower than that of Cd and Zn, which is consistent with the Gouy-Chapman theory. On the other hand, the value of the magnetic susceptibility of water, which, like that of heavy metals is negative, which implies its diamagnetism and with which it will be possible to determine the hydrated magnetic susceptibility, resulting to be now larger than the hydrated magnetic susceptibilities of both Cd and Zn, compared to that of Pb.

As can be seen in the specific case of Pb, Cd and Zn are diamagnetic species despite being heavy metals; however, many transition metal complexes exhibit both diamagnetic and paramagnetic complexes. For example, the compounds of the complex ion $[Co(NH_3)6]^{3+}$ do not have unpaired electrons, but the compounds of $[CoF_6]^{3-}$ ion have four per metal ion. Both complexes contain Co(III) with $3d^6$ electronic configuration [94], although both Zn and Cd are elements of electronic configuration *d*, they exhibit diamagnetic behavior because they are at the limit of groups IIB and tend to behave similar to group IIIA.

On the other hand, a simple way to know the magnetic ordering of ionic species in aqueous solution may be the coloration of the salt from which they start, since in heavy metal compounds, orbitals often are divided into two sets of separate orbitals by the division energies, which corresponds to wavelengths of light in the visible region. The absorption of visible light causes electronic transitions between orbitals in these sets, which imply a correlation between the paramagnetism and the salt coloration, while the diamagnetism of some metallic nitrates is colorless [94].

	χ_{mag} (1 × 10^{-6} emu/mol)	r_{ion} (pm)	n_h (min–max) [2]	χ_h (1 × 10^{-6} emu/mol)
Pb	−23	1.2	4–7.5	−120.275
Cd	−19.7	0.97	10–12.5	−181.825
Zn	−9.5	0.74	10–12.5	−171.625
H_2O	−12.97	—	—	—

Table 5. Physicochemical and magnetic parameters of different ions and hydrated ions.

Overall, a strong interdependence of the physicochemical and magnetic properties of ionic adsorbates can be observed and how they interact with exposure to a magnetic field.

8. Generation and configuration of a magnetic field applied to an adsorption process

Knowing the characteristics of the magnetic field that will interact with both the adsorbate and the adsorbent is overriding, three design parameters can be mentioned in the application of a magnetic field for assistance in the adsorption process: (1) type of source, (2) arrangement or trajectory of field lines and (3) magnetic field strength.

The source of the magnetic field can be classified by its generation in three different ways: alternating current (AC) electromagnets, direct current (DC) electromagnets and permanent magnets.

In general, the electromagnets consist of a solenoid made from cable made of an electrically conductive material, these solenoids generate a magnetic field in their cavity to the passage of electric current. The design of the solenoid goes from linear, annular and cylindrical forms according to the application, where the magnetic field strength that could generate depends on the number of turns, intensity and current potential [95].

The AC electromagnets are distinguished by passing electric current in which the magnitude and direction of the current vary in cyclical periods. This type of electromagnets have been used to generate magnetic fields applied in multiple ways ranging from heating by induction micellar particles to remove dyes from water [96], the acceleration in the germination of seeds under the application of a magnetic pulse [97], to be used as an incentive for the adsorption of volatile organic compounds [98]. They present some advantages in the generation of temperatures by induction and work with magnetic pulses of high frequency; however, it has some other disadvantages such as the creation of a heterogeneous and discontinuous magnetic field.

DC electromagnets are usually the most used for assistance in processes to eliminate water pollutants, and they are completely similar to AC electromagnets, with the difference that the field they generate is homogeneous and continuous due to the nature of the electric current, although they can also generate pulsations [99–101]. Studies have been carried out on the application of an electromagnet in adsorption processes, where it has been found that the field generated by them decreases the equilibrium time [101], increases the efficiency of dye removal by orienting the molecules and creating micromagnets from magnetic adsorbent materials [102]. It was also found that there is an effect of the pulsed magnetic field on the morphology of the surface of the adsorbents and the intensification of the adsorption activity [103]. However, the heating of the solenoid generates a disadvantage because the temperature control in adsorption studies is fundamental, so it would be necessary to use a cooling system for temperature control [9], raising the equipment costs.

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Within the electromagnets, there is a new modality that is also used for water purification processes; it is the superconducting magnets that generate the high-gradient magnetic field. The superconducting magnets are capable of generating a magnetic field from 2 to 10 T [95, 104].

Finally, permanent magnets, which are manufactured from ceramic and nonceramic ferromagnetic materials that have iron, nickel, cobalt, rare earth and even silica as the main elements [105–108].

The magnetic force that permanent magnets could present depends on two factors: the coercive field that presents the material and the field strength with which it is loaded in its manufacture where it is possible to find a wide variety of intensities that can be higher than 2 T per unit of length. In addition to this, they have the advantage of offering a reduction in the operational cost related to the consumption of electric power for the generation of magnetic field [95]. In addition, both the polarity of the magnet and the versatility of its shape can be an advantage in the design of devices for assisting the adsorption process. On the other hand, its fragility, oxidation in environments with humidity and limitation in size are rejected for industrial application. Like electromagnets, permanent magnets have been used for their application in adsorption processes, showing changes in kinetics [108]; modification in the magnetic ordering of materials [109], modification in the magnetic ordering in addition to the removal of metals from groundwater without modifying components of Ca and Mg materials [110].

Like the source, the configuration of the magnetic field is significant in the design of an adsorption system assisted by magnetic fields, since the influence, gradient of intensity and direction will determine the type of effect in some of the adsorption phases. The configuration of the magnetic field will influence in various ways ranging from orienting, hydrating and redirecting the adsorbate, to magnetize, activate and prevent the degeneration of the adsorbent [111–115].

In the case of adsorption in continuous systems, the application of the magnetic field could be based on the orientation of the magnetic field, which could be parallel or perpendicular to the directions of the fluid flow [95]. Particularly in the use of permanent magnets, they have been used in the treatment of electrolytes where they were arranged around the treatment circuit, so that the applied field was orthogonal to the direction of fluid flow [116].

For adsorption studies in batch systems, there are multiple configurations. Experiments have been carried out in which a pair of permanent magnets is arranged in parallel, where the space between them is the adsorption system showing favorable results [117]. This could be explained thanks to the sum of magnetic intensities, which is a phenomenon that occurs when it has two or more magnets in parallel and their polarities are attracted, and this is only effective at short distances and this distance is a function of the intensities particular of each permanent magnet involved. On the other hand, the variation of adsorption results was investigated by applying different magnetic dispositions taking as an output variable the removal of the adsorbate, where three different configurations in which permanent magnets were disposed and evaluated, particularly the behavior of the fluidized adsorbent particles was evaluated. In the contaminated solution, obtaining different degrees of removal of a dye,

this study explains that the effect of the Lorentz forces modify the trajectories of the adsorbent particles inside the container, opposing the turbulent forces created by the mechanical agitation [112]. On the other hand, it has been found that there is a relationship between the magnetic configuration and the gradient of magnetic intensities, in addition to the arrangement of magnetic fields that can influence the solution, the adsorbent material and both [113].

The evaluation of different magnetic systems can be designed from computer simulations which can quantify and visually inspect the trajectories of the field lines, the gradient of magnetic intensities and the spatial distribution profiles of the magnetic flux density [84, 118–120], in such a way that it is possible to be certain that the adsorption system will be affected by the magnetic field.

Considering all the information presented in this chapter, it is possible to affirm that the magnetic field is a viable tool within the adsorption processes for the removal of heavy metals in an efficient and environmentally friendly way; however, there are different parameters that are essential to take into account to achieve the expected results. The mechanism under which the magnetic field acts on the adsorbate-adsorbent system is quite complex and depends on several variables within the system.

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