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REVIEW ARTICLE

An overview of carbon nanotubes role in heavy metals removal from wastewater

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Abstract The scarcity of water, mainly in arid and semi-arid areas of the world is exerting exceptional pressure on sources and necessitates offering satisfactory water for human and different uses. Water recycle/reuse has confirmed to be successful and promising in reliable water delivery. For that reason, attention is being paid to the effective treatment of alternative resources of water (other than fresh water) which includes seawater, storm water, wastewater (e.g., dealt with sewage water), and industrial wastewater. Carbon nanotubes (CNTs) are called the technology of 21st century. Nowadays CNTs have been widely used for adsorption of heavy metals from water/wastewater due to their unique physical and chemical properties. This paper reviews some recent progress (from 2013 to 2018) in the application of CNTs for the adsorption of heavy metals in order to remove toxic pollutants from contaminated water. CNTs are expected to be a promising adsorbent in the future because of its high adsorption potential in comparison to many traditional adsorbents.

Keywords carbon nanotubes, heavy metals removal, water treatment

1 Introduction

Today, the rapid increase in the world population, unplanned urbanization, industrialization, agricultural activities as well as the immoderate use of chemical compounds has extremely caused environmental pollution and the release of heavy metals into the environment. The heavy metal ions come from various sources, such as current chemical industries along with pigment manufacturing, photographic materials, fertilizer manufacturing, tannery, products of petrochemical industries, leather tanning, printing, battery production industries, metallurgical, mining, etc. Such industries pose a sizeable danger to people as well as the surrounding communities. Nowadays, because of the chemical compounds and heavy metals discharge into the environment, the pollution of water and soil has been causing global concern. When found in discharge, polluted materials are launched into the environment which can be easily absorbed in fauna and plant life. Aquatic systems are mainly sensitive to pollution in all likelihood because of the shape of their food chain. Heavy metals enter the food chain, and they accumulate in fish and other safe to eat organisms. Because of biomagnifications, human beings get hold of the most impact, for the reason that they are at the top of the food chain. Subsequently, heavy metal contamination has been a crucial trouble [1–3].

The most common toxic ions in aqueous solutions which are accountable for particular issues include heavy metallic ions, dyes, oil, organic pesticides, and radionuclides, etc. [4–9]. Heavy metals cannot be decomposed, so they accumulate in living organisms. Toxic heavy metals include chromium (Cr), lead (Pb), zinc (Zn), arsenic (As), mercury (Hg), nickel (Ni), copper (Cu), and cadmium (Cd). These metal ions have excessive toxicity, non-biodegradable sites, and they have serious health problems in animals and humans. The impacts of heavy metals toxicity on human health and their consequences have been reported substantially, that these consists of obstructive lung disease, osteoporosis, inhibition of enzymes, renal dysfunction, high blood pressure, damage to the gastrointestinal tract, neurological disorder, speech problems, aggressive behavior, sleep disabilities, fatigue, irritability, mood swings, melancholy, vascular occlusion, memory loss, increased allergic reactions and autoimmune diseases [10–13]. A number of metals such as lead, mercury, chromium, arsenic, and cadmium can be dangerous to the human body.

As a matter of fact, human body desires small dose consisting of Zn(II), an excess of it could cause various

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diseases and health problems including lethargy, dryness in the throat, anemia, neurological symptoms, and raised thirst. Exposure to such poisonous ions can cause health problems along with kidney or liver damage, insomnia, Wilson ailment, diarrhea, most cancers, nausea, dermatitis, vomiting, persistent asthma, headache, and coughing [14–16].

It's far widely recognized that continual cadmium toxicity is the cause of Itai-Itai disease in Japan. The damaging consequences of cadmium also consist of renal disease, lung disease, and emphysema, testicular atrophy and malformations of the fetal skeleton [17,18]. Chronic lead exposure causes encephalopathy with the subsequent signs like insomnia, vertigo, migraine, seizures, and coma. The current global production of Pb(II) is still rising. Thus, the effective elimination of these toxic ions from wastewater is essential for health and environmental issues and is being widely studied. Adsorption found to be effective methods among the all available heavy metal removal methods. In the recent decade, carbon nanotubes (CNTs) have found a notable capability utility for the elimination of heavy metal ions, pollutants, and dyes [19–22]. This review outlines the metal ions removal from aqueous solutions using CNTs.

2 Possible strategies to deal with heavy metals in water

Over the last years, some of the technologies have been developed to tackle toxic heavy metal ions in water. Of course, the appropriate treatment method choice mainly hinges on the degree of preliminary heavy metals concentration and the economical methods. Several conventional methods and strategies such as precipitation [23–25], adsorption on manganese dioxide [26], ion exchange [27–29], coagulation-flocculation [30], solvent extraction [31,24,32–34], Solid Phase extraction [35,36], electrochemical treatment techniques [30], Magnetic solid phase extraction [37], reverse osmosis [38], membrane filtration [30,39] lime softening, oxidation, etc., have been used to eliminate the heavy metal ions [40–43]. These techniques have advantages and disadvantages. Coagulation and flocculation produce too much sludge, and flotation process is also costly. Although in ion exchange the elimination potential is high, reproduction of the resins leads to secondary contamination. Although the low-price and simple operation are the advantages of the chemical precipitation method, this method produces dangerous waste, and the removal of these residues needs an extraordinary operational cost. Electrodialysis consumes high energy, and its electrical usage makes it expensive [44,45].

Among the methods mentioned above, the adsorption is the inexpensive and powerful technique for the adsorption of some of metals and pollutants from wastewater [46–48]. This method changed into the maximum appropriate approach because of its high performance and economic consideration [49,50]. Additionally, the applied adsorbents can be reused via appropriate desorption method. Various adsorbents like activated carbon [51–58], biomaterials, zeolites [59], polymers, nanoparticles [60], functionalized chitosan [61,62], anaerobic granular biomass [63], peanut hulls pellets [64], resins [65], kaolinite [66], manganese oxides [67], biomaterials [68–70], peat [71], olive stone waste [72], chemically modified sugarcane bagasse [73], crab shell [74], fluorine and nitrogen co-doped magnetic carbons [75], activated sludge derived magnetic carbon [76], a triple layered core-shell structured zero valence iron@carbon@polyaniline (ZVI@C@PANI) nanocomposite [77], monolayer poly(acrylic acid)/γ-alumina nanocomposites (PAA/alumina) [78], synthesized fibrillar and particulate magnetic carbons [79], poly(acrylic acid)/alumina [80], porous Lignin based poly (acrylic acid)/organo-montmorillonite nanocomposites [81], etc., have been significantly used for adsorption of the heavy metal ions [49,59,82–88]. However, some of these absorbents have low adsorption capacity and removal efficiency removal efficiency for heavy metals. Therefore, researchers are trying to find and to synthesize more efficient and new promising absorbents. Adsorption with new adsorbents has low-cost, high adsorption capacities, suitable for removal a variety of pollutants, ease of the separation process, usable over a wide pH range, ability to remove of some metals simultaneously, easy operating conditions and good recyclable [45].

In current years, carbon nanotubes have been applied remarkably as a novel adsorbent to deal with the heavy metal ions and various contaminants in water. The researchers showed that carbon nanotubes are splendid adsorbent for adsorption of organic chemical compounds, heavy metals, radionuclides and pollutant molecules. Regardless of high costs, CNTs may be beneficial because of their high adsorption capacities that offset their high price, and the cost can be decreased because of their regeneration and frequent accessibility [89]. A number of studies have demonstrated that by entering CNTs into the surroundings, the metal adsorption happens rapidly and heavy metal ions toxicity would be increased. So the depletion of CNTs into the environment should be confined. However, the toxicity of CNTs sole is not drastic [90–93].

3 Use of nanomaterials for elimination of heavy metals

Despite the fact that conventional sorbents could eliminate heavy metal ions from wastewater, the low efficiencies and sorption capacities restrict their application. Nowadays, nanomaterials are employed as the new adsorbents for adsorption of heavy metal ions from wastewater to overcome the problems of conventional adsorbents. Nanomaterials are usually known as the materials with the size range from 1 to 100 nm. These are extensively being investigated due to the latest developments in nanotechnology and nanosciences [18]. Nano-structured materials as adsorbent have displayed a lot better performance in water treatment than traditional materials. Nanomaterials as heavy metal adsorbent should have the following properties: (a) The nature of sorbents should not be toxic, (b) the nanosorbents must have high absorption capacity and great power selectivity in absorbing contaminants, (c) the desorption of heavy metals could be easily, (d) the adsorbents could be able to be recycled several times [5].

Different types of nanomaterials such as nanotubes [94], nanometal or metal oxides, dendrimers, the carbon-based materials, graphene composites are acknowledged to show excessive adsorption capacities. Carbon-based nanomaterials [95] containing fullerenes, carbon nanotubes, graphene oxide [94] and nanodiamonds [96] are broadly used for elimination of heavy metals due to its high sorption capacities and non-toxic nature. However, in due course of time, with the improvement of nanotechnology carbon nanotubes, graphenes and fullerenes were synthesized and were used as nanosorbents.

4 Carbon nanotubes in water remedy

CNTs which were observed by Iijima in 1991 [97,98], are also used broadly to eliminate heavy metals in wastewaters. CNTs are allotropes of sp^2 hybridized carbon with a cylindrical nanostructure including six-membered carbon rings. There are two major varieties of CNTs: single-walled (SWCNTs) and multi-walled (MWCNTs) (as shown in Fig. 1) [99]. The SWCNTs contain one cylindrical graphene sheet, and analogously the MWCNTs consist several tubes in concentric cylinders. The lengths and diameters range of SWCNTs are usually greater than 1 μm, and 1–2 nm, respectively. Also, These parameters for MWCNTs are well over 1 μm, and 1–50 nm, respectively [100].

CNTs display specific structural, semiconductor, optoelectronic, electronic [101–104], mechanical, chemical and physical properties [102,105–109]. Lin et al. synthesized nano-$TiNb_2O_7$/carbon nanotubes composite and evaluated their electrical conduction. The results showed that the good contact between $TiNb_2O_7$ and CNTs particles reduces particle size and increases the electrical conductivity [110]. In order to increase the interstitial properties of epoxy resin composite, carbon nanotubes were bonded to carbon fibers by layer-by-layer grafting method. Zhao et al. indicated that carbon nanotubes improves the mechanical and interfacial properties of these composites [111]. CNTs seem to be ideal adsorbents among carbon nanomaterials due to their light mass density, high specific surface area, high reactivity, excessive porous and hollow structure, thermal resistance, alkaline and acid stability, the strong interaction between CNTs and the adsorbate molecules (e.g., heavy metal ions, dyes, organic pesticides, pollutant molecules) and unique chemical and physical properties [112–123].

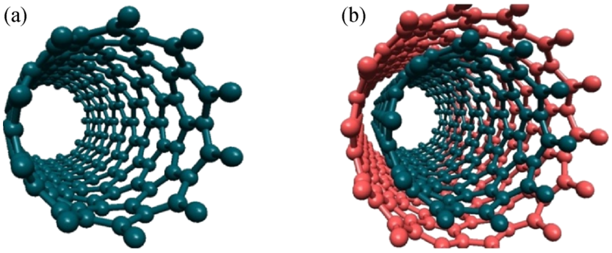

Fig. 1 Types of carbon nanotubes: (a) a SWCNT and (b) a MWCNT

There are several methods and techniques to produce CNTs. After production, carbon nanotubes have some impurities including carbonaceous and metallic impurities. Usually, by decreasing the diameter of the carbon nanotubes, the amount of impurities raises. Carbon impurities commonly include carbon nanoparticles (CNPs), fullerenes, and amorphous carbon. The metallic impurities commonly include residual of the metals particles that used in the production of CNTs as a catalyst, such as Fe, Ni, Co, etc. [124]. These impurities limit some applications of CNTs. Therefore, to improve the performance of CNTs, purification is an essential subject to be investigated. Fullerenes, by dissolving in specific organic solvents can be easily removed. The removal of amorphous carbon is moderately easy because of the high density of the defects in their structure, which allows their oxidation to be carried out under mild conditions. The basic problem in the purification of the CNTs is the removal of graphitic particles and polyhedral carbons.

Because of the strong van der Waals interactions of the raw carbon nanotubes and formation of big bundles held strongly together, CNTs usually cohere to each other and tend to aggregate in the aqueous solution. Therefore they are very hard to disperse homogeneously in most organic and aqueous solutions as well as poorly chemically [125]. This aggregation limits their potential applications, especially the ability of the CNTs to remove the pollutants and heavy metal ions due to a decrease in available adsorption sites. Although raw carbon nanotubes can be dispersed in solutions via ultrasonication, after finishing of sonication, dispersion of CNTs decrease with time and rapid aggregation of CNTS occurs [125–127]. To enhance the sorption capacities, CNT’s have been changed via oxidation combining with other metal ions or metal oxides and coupled with organic compounds. Surface modification and selective functionalization of CNTs improves dispersion of CNTs in aqueous solution [128–132]. Plasma method also found to be the new technique for improving the adsorption capacity and the dispersion of the CNTs in the solution due to introduce oxygen-containing groups onto the surface of CNTs [133–137].

The adsorption behavior of carbon nanotubes depends on their site density, purity, surface area, porosity, functional groups and kind of CNTs. There are several adsorption sites on CNT bundles to remove various contaminants, including inner sites, grooves, interstitial channels, and outside surface. A lot of research has been performed to investigate the adsorption process on absorbent sites [138–142]. It has been reported that the external sites are occupied earlier than the internal sites under the equal conditions. Because external sites are accessible for the adsorbing material [143]. The number of unblocked carbon nanotubes is effective in the adsorption process. Some researchers reported that opening the caps of CNTs enhances the number of sorption sites and adsorption capacity [144–146].

MWCNTs were functionalized with 8-hydroxyquinoline, which can be used to elimination of Pb(II), Cu(II), Zn (II) and Cd(II) [147]. Graphene is another form of carbon structure with a unique two-dimensional shape and superior mechanical and thermal properties, which is used as a nanosorbent. Hwang et al. [148] synthesized magnetic graphene adsorbents with a particle size of about 10 nm which provide a high binding ability for As(III) and As(V) and the consequences indicate that the excessive binding ability is because of the multiplied adsorption site in the graphenes composite. Nanoparticles fashioned with the aid of metal or metal oxides are some other inorganic nanomaterials, which might be used extensively to remove heavy metal ions in wastewater. Moreover, metal oxides have low solubility, minimal environmental effect and no secondary pollutants. Saeidi et al. fabricated graphene/activated carbon composite (GAC) from graphene oxide and glucose for adsorption of Pb(II) from wastewater. The GAC possessed high surface area (2012 m^2/g), large pore volume (1.61 cm^3/g) and mesopore structure. Regarding the Langmuir equation, maximum adsorption capacity for Pb(II) with GAC was 217.6 mg/g [149].

Anion selective para toluene sulfonic acid (pTSA) doped polyaniline@graphene oxide-multiwalled carbon nanotube composites (pTSA-Pani@GO-CNT) were applied for the removal of Cr(VI) and Congo red (CR) dye [150]. Hayati et al. reported the adsorption of Cu(II) and Pb(II) from aqueous solution using CNT modified with four generations of poly-amidoamine dendrimer (PAMAM/CNT). The results showed that PAMAM/CNT nanocomposites were a super-adsorbent. The high adsorption capacities for Cu(II) and Pb(II) ions were obtained as 3333 and 4870 mg/g respectively [151]. Ahmadzadeh Tofighy et al. investigated the removal of Cu(II) from aqueous solutions using acid–chitosan-functionalized CNTs sheets [152]. Functionalized MWCNT wrapped polypyrrole nanocomposite (PPy/o-MWCNT) was used in the adsorption of Pb(II), Ni(II) and Cd(II) ions from aqueous medium [153]. Srinivasa et al. prepared chitosan/MWCNTs (CS/MWCNTs) composite beads and used as a biosorbing agent to remove copper ions from an aqueous solution. Based on the Langmuir isotherm modael, maximum adsorption capacity for Cu(II) with CS and CS/MWCNTs composite was found to be 178.57 and 454.55 mg/g [129]. In another study, Lasheen et al. investigated the adsorption capabilities for the removal of Cu(II), Pb(II), Cd(II), Ni(II), and Cr(VI) on as-produced MWCNT and oxidized MWCNT [154]. Jiang et al. synthesized novel-modified magnetic multi-walled carbon nanotubes (Fe_3O_4/O-MWCNTs) via evaporating acid purification and co-precipitation method [155]. A new functionalized MWCNTs by 3-aminopyrazole (MWCNTs-f) was synthesized and used for adsorption of Cd(II) from aqueous solutions. At the optimum conditions, the removal efficiencies of Cd(II) using MWCNTs-f, MWCNTs-COOH, and MWCNTs were 83.7%, 65.9% and 35.7%, respectively [156]. Mubarak et al. investigated the adsorption capacity of functionalized carbon nanotubes and magnetic biochar for the removal of Zn(II). Zinc ions removal efficiencies were 99% and 75% for functionalized CNT and magnetic biochar, respectively [157]. Isocyanate functionalized multiwalled carbon nanotubes were used for adsorption of Pb(II) from aqueous solutions and selective separation of Tl-201 from Pb-201. The adsorption efficiency of Pb-201 was found to be 99% [126]. Feasible water flow filter filled with Fe_3O_4-functionalized non-oxidative graphene/CNT (M-G/C) composites were applied for arsenic removal [158]. CNTs are one-dimensional (1D) nanostructures, and graphene-based materials are two-dimensional (2D) structure nanomaterial [158–160]. Three-dimensional (3D) structures with large specific surface areas obtain from the mixture of the graphene-based materials and CNTs. These composites were Functionalized with metal oxides for removal of arsenic from the water. Yoon et al. in their study on adsorpon of As ions, used the Couette-Taylor flow technique to prepare Fe_3O_4-non-oxidative graphene (M-G), Fe_3O_4-CNT (M-C), and Fe_3O_4-non-oxidative graphene/CNT (M-G/C) composites. They compared the Couette-Taylor flow method with hydrothermal synthesis method. The results showed that the functionalization of Fe_3O_4 on the non-oxidative graphene and CNTs via the Couette-Taylor flow reactor is more uniform and quick, but in hydrothermal synthesis the uniform distribution and functionalization of the materials is not possible. The adsorption of As(III) and As(V) onto the Fe_3O_4-functionalized carbon-based nanomaterials was performed and compared using a continuous flow filter and the batch method. In flow filter, the M-G/C filter demonstrated effective and high removal of As(III) and As(V) from the water. The M-G/C composite has 3D structure, with high specific surface area, thereby priovid a longer pathway for water flow. These results display that the filter makes adsorption sites more available for arsenic adsorption on materials produced [158]. Saadat et al.

investigated the adsorption capacity of SWCNTs, MWCNTs, SWCNTs/Fe and carbon nanofibers for lead removal. The results showed that the SWCNTs have the great potential for adsorption of Pb(II) ions. They prepared single-walled carbon nanotubes-doped walnut shell composite (SWCNTs/WSh) and evaluated their potential for the removal of Pb(II) ions. SWCNTs/WSh were found to be the most suitable compared to unmodified CNTs in terms of the adsorption capacity [161]. Zerovalent iron doped MWCNT (ZCNT) was synthesized and evaluated its adsorption ability of As(III) and As(V). The Ethylenediaminetetraacetic acid (EDTA) has been widely used in this composite as the chelating agent, and it has retained zerovalent state of iron. In addition to zerovalent iron (ZVI), this adsorbent has several functional groups such as carboxyl, hydroxyl and amine groups [162]. Oxidized MWCNT were used in the removal of Cu(II) ions from aqueous solutions [163]. Functionalization of MWCNTs surface with Phosphonuim based deep eutectic solvents (DES) were prepared for removal of arsenic from water. AlOmar et al. prepared two DESs systems using two phosphonium based salts, methyl triphenyl phosphonium bromide (MTPB) and benzyl triphenyl phosphonium chloride (BTPC), in conjugation with glycerol, which acts as a hydrogen bond donor. Oxidized-CNT were synthesized using $KMnO_4$ via sonication for two hours to introduce K-CNT. K-CNT was added to MTPB-based DES (KM-CNT) and BTPC-based DES (KB-CNT) separately. These functionalized CNTs including $KMnO_4$, MTPB and BTPC based DES were used to adsorb As(III) from water [164]. The synthesis and adsorption capability of Ni(II), Zn (II), As(III) and Co(II) metals by PAMAM/CNT nanocomposite was studied. The PAMAM dendrimers have low toxicity, availability and cheapness. The results showed that PAMAM/CNT can be used as a super-adsorbent for the adsorption of heavy metal ions from wastewater [165]. Stannic molybdophosphate–MWCNTs hybrid (MWCNT/SMP) were synthesized and used to investigate and compare strontium adsorption from aqueous solution by hybrid and its pristine material, SMP and ox-MWCNTs. MWCNTs-SMP exhibited a 4.9-fold higher adsorption capacity for Sr(II) ions compared to raw SWCNTs, and a 1.35-fold higher adsorption capacity compared to ox-MWCNTs [166]. polyamine/CNT composites were produced through a one-pot polycondensation reaction of melamine, paraformaldehyde, 1,6-diaminohexane and chlorinated CNTs under optimal conditions in the presence of *N*,*N*-dimethyl formamide as a solvent. The pure melamine-formaldehyde-diaminohexane (MFDH) polymer and the functionalized composites (MFDH-1, MFDH-2, MFDH-3 and MFDH-4) containing 0.01%, 0.02%, 0.05% and 0.1% weight of chlorinated carbon nanotubes as the starting precursors were used to remove lead ions from aqueous solutions. The results showed that MFDH-4 have a greater potential for adsorption of Pb(II) [167]. Yang et al. prepared diamine functionalized mesoporous silica on MWCNTs (NN-$mSiO_2$@MWCNTs) by adding [amino-ethylamino] propyltrimethoxysilane to mesoporous SiO_2 coated MWCNTs ($mSiO_2$@MWCNTs). These materials were applied to remove Pb(II), Cu(II), Ni (II), and Zn(II) from aqueous solution. In order to investigate the adsorption ability of the these materials, the experiments were carried out by stirring 20 mg of adsorbents in 20 mL of 100 mg/L aqueous metal solutions at pH 6.2, and temperature 25°C. The results indicated that the adsorption capacity order of these metals by CNTs is NN-$mSiO_2$@MWCNTs > $mSiO_2$@MWCNTs > O-MWCNTs > MWCNTs, and the adsorption percentage for mentioned metals is in the order of Pb(II) > Cu(II) > Zn (II) > Ni(II) [168]. Voelcker et al. studied Hg(II) ions adsorption using Thiol-derivatized SWCNTs (SWCNT-SH). These adsorbents were prepared via reacting oxidized SWCNTs with decarboxycysteine hydrochloride. The adsorption behavior study of mercury on these adsorbents demonstrate that the adsorption capacity of the SWCNT-SH is 3.0 times higher than that of the raw MWCNTs and 4.0 times higher than that of the activated carbon [169]. In another article, Sankararamakrishnan et al. discussed the adsorption of Cr(VI) and Cd(II) by nano floral clusters (NCs). Composite nano floral clusters were synthesized by chemical vapor deposition using activated alumina as the substrate. Ni(II) nitrate and Fe(III) nitrate were used as catalysts for the growth of CNTs. Novel NCs showed great potential as adsorbents due to the presence of CNT, activated alumina, amorphous carbon and several functional groups such as hydroxyl, carboxyl, and carbonyl [170]. Table 1 shows the brief summary of the adsorption of heavy metals using non-modified, functionalized CNTs, and the interaction mechanisms between the CNTs and heavy metal ions.

The mechanism of metal ions adsorption onto CNTs depends on the properties of the absorbed functional groups onto CNTs, and metal ions. Various adsorption mechanisms have been reported such as the electrostatic interaction, physisorption, surface complexation, van der Waals interactions, chemisorption, soft acid-soft base interaction, ion exchange, hydrophobic interactions, external diffusion, intraparticle diffusion, cation-π interaction, anion-π interaction between the adsorbent and the metal ions. Among these, chemical interaction and electrostatic interactions between the surface functional groups of CNTs and the heavy metal ions is the major adsorption mechanism.

The equilibrium adsorption isotherm is usually used to describe the adsorption mechanism, interaction between adsorbent and adsorbate, and type of coverage. They could also provide essential information about the maximum adsorption capacity of adsorbent. The adsorption data can be fitted by pseudo second order rate equation, Langmuir, Freundlich, Temkin, Dubinin-Radushkevich (D-R) isotherm models, and intraparticle diffusion model to describe the equilibrium adsorption and adsorption mechanism. The most common isotherm models used in the metal ions sorption by CNTs are the Langmuir and Freundlich models. In most instances, one of the isotherms can best explain the mechanism of heavy metals adsorption. In Langmuir isotherm, a monolayer adsorption of heavy metal ion on the adsorbent surface occurs, and This model assumes that the adsorbent surface is homogeneous; while the Freundlich isotherm is based on multilayer adsorption and in high concentrations of the adsorbates, the adsorption capacity does not remain constant. In this model surface of the adsorbent is heterogeneous [171–173]. The results of the interaction mechanisms between CNTs and heavy metal ions for the different adsorbents are listed in Table 1.

Table 1 Adsorption of heavy metals using non-modified and functionalized CNTs, and their maximum adsorption capacities*

Adsorbent	Adsorbate	Q_m /(mg·g^{-1})	Experimental conditions	Interaction mechanism	Best fitted isotherm model	Ref.
Functionalized MWCNTs	Zn(II)	1.05	pH 10, initial concentration = 1.1 mg/L, adsorbent mass = 0.09 mg, contact time = 120 min, T = 50°C, agitation speed = 120 r/min	Electrostatic interaction	Langmuir, Freundlich	[158]
NN-mSiO$_2$@MWCNTs	Cu(II)	66.577	pH 6.2, initial concentration = 100 mg/L, adsorbent mass = 20 mg, contact time = 30 min, T = 25°C	Chemisorption	Langmuir	[168]
SWCNT	Hg(II)	40.16	pH 5, initial concentration = 30 mg/L, adsorbent mass (m/v) = 0.25 mg/mL, contact time = 1 h, T = 25°C	Chemisorption	Langmuir, Freundlich	[169]
Thiol-derivatized SWNTs	Hg(II)	131.58	pH 5, initial concentration = 30 mg/L, adsorbent mass (m/v) = 0.25 mg/mL, contact time = 1 h, T = 25°C	Strong chemisorption, soft acid-soft base interactions	Langmuir	[169]
Raw MWCNTs	As(V)	2.995	pH 4, initial concentration = 94 μg/dm^3, adsorbent mass = 100 mg/dm^3, contact time = 45 min, T = 25°C	Electrostaticinteraction, chemisorption	Langmuir–Freundlich (Sips)	[174]
Oxidized MWCNTs	As(V)	3.613	pH 4, initial concentration = 94 μg/dm^3, adsorbent mass = 100 mg/dm^3, contact time = 45 min, T = 25°C	Chemisorption, electrostaticinteraction, specific adsorption	Langmuir–Freundlich (Sips)	[174]
e-MWCNTs	As(V)	12.175	pH 4, initial concentration = 94 μg/dm^3, adsorbent mass = 100 mg/dm^3, contact time = 45 min, T = 25°C	Chemisorption, electrostatic interaction, specific adsorption	Freundlich	[174]
MWCNTs	Cu(II)	3.19×10^{-5} mol/g	pH 5, initial concentration = 2.36×10^{-4} mol/L, T = 30°C	Electrostatic interaction, surface complexation, surface precipitation	Langmuir	[175]
SWNTs	Cr(VI)	20.3	pH 4, initial concentration = 500 μg/L, adsorbent mass = 100 mg/L, contact time = 720 min	Chemisorption, physisorption, electrostatic interaction	Langmuir	[176]
MWNTs	Cr(VI)	2.48	pH 4, initial concentration = 500 μg/L, adsorbent mass = 100 mg/L, contact time = 720 min	Chemisorption, physisorption, electrostatic interaction	Langmuir	[176]
MWCNT–HAP	Co(II)	16.26	pH 6, initial concentration = 1.69×10^{-4} mol/L, adsorbent mass = 0.6 g/L, T = 20°C	Surface complexation	Langmuir	[177]
Sulphur containing MWCNTs (S-MWCNTs)	Hg(II)	72.8 μg/g	pH 12.15, initial concentration = 100 ppb, contact time = 60 min	Chemisorption, soft acid-soft base interactions, electrostatic interaction	Freundlich	[178]
Ag-MWCNTs	Cu(II)	53.29	pH 6, initial concentration = 50 mg/L, adsorbent mass = 0.05 g, contact time = 100 min, T = 20–40°C, agitation speed = 180 r/min	Chemisorption, ion exchange	Langmuir	[179]

(*Continued*)

Adsorbent	Adsorbate	Q_m /(mg·g^{-1})	Experimental conditions	Interaction mechanism	Best fitted isotherm model	Ref.
Ag-MWCNTs	Cd(II)	51.97	pH 7, initial concentration = 50 mg/L, adsorbent mass = 0.05 g, contact time = 100 min, T = 20–40°C, agitation speed = 180 r/min	Chemisorption, ion exchange	Langmuir	[179]
CF-CNTs-A	As(V)	16.84	pH 4, initial concentration = 5 mg/L, adsorbent mass = 0.2 g/L, contact time = 360 min, T = 25°C, agitation speed = 150 r/min	Electrostatic interaction, surface complexation	Freundlich	[180]
CF-CNTs-A	As(III)	16.21	pH 7.5, initial concentration = 5 mg/L, adsorbent mass = 0.2 g/L, contact time = 300 min, T = 25°C, agitation speed = 150 r/min	Surface complexation	Langmuir	[180]
CF-CNTs	As(V)	30.96	pH 4, initial concentration = 5 mg/L, adsorbent mass = 0.2 g/L, contact time = 360 min, T = 25°C, agitation speed = 150 r/min	Electrostatic interaction, surface complexation	Freundlich	[180]
CF-CNTs	As(III)	28.74	pH 7.5, initial concentration = 5 mg/L, adsorbent mass = 0.2 g/L, contact time = 300 min, T = 25°C, agitation speed = 150 r/min	Surface complexation	Langmuir	[180]
CS/MWCNTs	Cu(II)	454.55	pH 5.5, initial concentration = 100 mg/L, adsorbent mass = 0.05 g, contact time = 90 min	Surfacecomplexation, van der Waals interactions	Langmuir	[129]
SWCNTs/WSh	Pb(II)	185.18	pH 5, initial concentration = 50 mg/L, adsorbent mass = 1 g/L, contact time = 30 min, T = 25°C, agitation speed = 65 r/min	Electrostaticinteractions, chemisorption, precipitation	Langmuir	[161]
CNT	As(III) As(V)	0.124 0.373	pH 7, initial concentration = 500 μg/L, adsorbent mass = 2.5 g/L, contact time = 240 min	Electrostatic interactions	Langmuir	[162]
ZCNT	As(III) As(V)	111.1 1666.7	pH 7, initial concentration = 500 μg/L, adsorbent mass = 2.5 g/L, contact time = 240 min	Complexation, chemisorption, diffusion processes	Langmuir	[162]
NCs	Cr(VI)	264.5	pH 2, initial concentration = 100 mg/L, adsorbent mass = 2.5 g/L, contact time = 240 min, T = 30°C, agitation speed = 100 r/min	Electrostaticinteractions, chemisorption, intraparticle diffusion, external diffusion	Langmuir	[170]
NCs	Cd(II)	229.9	pH 7.5, initial concentration = 100 mg/L, adsorbent mass = 2.5 g/L, contact time = 240 min, T = 30°C, agitation speed = 100 r/min	Electrostaticinteractions, complexation, chemisorption, intraparticle diffusion, external diffusion	Langmuir	[170]
MWCNTs	Hg(II)	5.479	pH 6, initial concentration = 100 mg/L, adsorbent mass = 0.02 g, contact time = 90 min, agitation speed = 110 r/min	Ion exchange, physisorption, chemisorption, external diffusion	Langmuir	[181]
MWCNT-OX	Hg(II)	27.32	pH 6, initial concentration = 100 mg/L, adsorbent mass = 0.02 g, contact time = 90 min, agitation speed = 110 r/min	Ion exchange, physisorption, chemisorption, complexation, external diffusion	Langmuir	[181]
CNT-I	Hg(II)	123.45	pH 6, initial concentration = 100 mg/L, adsorbent mass = 0.02 g, contact time = 105 min, agitation speed = 110 r/min	Ion exchange, physisorption, chemisorption, complexation, external diffusion, soft-soft interaction	Langmuir	[181]

(*Continued*)

Adsorbent	Adsorbate	Q_m /(mg·g^{-1})	Experimental conditions	Interaction mechanism	Best fitted isotherm model	Ref.
Sulfur incorporated MWCNT (CNT-S)	Hg(II)	151.51	pH 6, initial concentration = 100 mg/L, adsorbent mass = 0.02 g, contact time = 105 min, agitation speed = 110 r/min	Ion exchange, physisorption, chemisorption, complexation, external diffusion, soft acid-soft base interaction	Langmuir	[181]
Amino and thiolated functionalized MWCNTs	Hg(II)	84.66	pH 6, initial concentration = 10 mg/L, adsorbent mass = 400 mg/L, contact time = 60 min, T = 25°C, agitation speed = 200 r/min	Physisorption, soft acid-soft base interaction	Langmuir	[182]
p-MWCNTs	Cu(II)	NA 36.82 based on D-R model	pH 5, initial concentration = 20 mg/L	Physical adsorption	Freundlich	[183]
Sulfonated MWCNTs (s-MWCNTs)	Cu(II)	NA 43.16 based on D-R model	pH 5, initial concentration = 20 mg/L	Electrostaticinteractions, physisorption, innerspheresurface complexation	Freundlich	[183]
Raw MWCNTs	Hg(II)	32.4	pH 4.3, initial concentration = 4.0 mg/L, agitation speed = 200 r/min	Electrostatic interactions, complexation, chemisorption, intraparticle diffusion	Langmuir	[184]
OH-MWCNTs (with a phenolic hydroxyl functional group)	Hg(II)	120.1	pH 4.3, initial concentration = 4.0 mg/L, agitation speed = 200 r/min	Electrostatic interactions, complexation, chemisorption, external diffusion, intraparticle diffusion	Langmuir	[184]
COOH-MWCNTs	Hg(II)	127.6	pH 4.3, initial concentration = 4.0 mg/L, agitation speed = 200 r/min	Electrostatic interactions, complexation, chemisorption, external diffusion, intraparticle diffusion	Langmuir	[184]
As-produced MWCNT	Pb(II) Cu(II) Cr(VI) Cd(II) Ni(II)	48.0 45.0 43.0 39.0 35.0	pH 5.5, initial concentration = 20 mg/L, adsorbent mass = 1 g/L, contact time = 120 min, agitation speed = 200 r/min	Chemisorption, electrostaticinteractions	Langmuir, Freundlich	[154]
Oxidized MWCNT	Pb(II) Cu(II) Cr(VI) Cd(II) Ni(II)	75.0 70.4 67.0 66.0 59.2	pH 5.5, initial concentration = 20 mg/L, adsorbent mass = 1 g/L, contact time = 120 min, agitation speed = 200 r/min	Chemisorption, electrostaticinteractions	Langmuir, Freundlich	[154]
Oxidized MWCNT	Cu(II)	200.0	pH 6, initial concentration = 50 mg/L, adsorbent mass = 0.03 g, contact time = 75 min, T = 25°C	Chemisorption, electrostatic interaction	Langmuir	[163]
Raw MWCNTs	Sr(II)	3.13	pH 6, initial concentration = 30 mg/L, contact time = 24 h, T = 25°C, agitation speed = 150 r/min	NA	Langmuir	[166]
Oxidized MWCNTs	Sr(II)	10.87	pH 6, initial concentration = 30 mg/L, contact time = 24 h, T = 25°C, agitation speed = 150 r/min	Chemisorption	Langmuir	[166]

(*Continued*)

Adsorbent	Adsorbate	Q_{m} /(mg·g^{-1})	Experimental conditions	Interaction mechanism	Best fitted isotherm model	Ref.
MWCNT/SMP (Hybrid)	Sr(II)	14.92	pH 6, initial concentration = 30 mg/L, contact time = 24 h, T = 25°C, agitation speed = 150 r/min	Chemisorption	Langmuir	[166]
MnO$_2$-CNTs	Hg(II)	58.82	pH 5–7, initial concentration = 10 mg/L, adsorbent mass = 0.02 g/20 mL, contact time = 80 min, T = 25°C, agitation speed = 180 r/min	Electrostaticinteractions, physisorption	Freundlich	[172]
Al$_2$O$_3$/MWCNTs	Cd (II)	27.21	pH 7, initial concentration = 1 mg/L, adsorbent mass = 1 g/L, contact time = 240 min, T = 25°C, agitation speed = 150 r/min	Electrostatic interactions, physisorption, surface complexation, surface precipitation, van der Waals interactions	Langmuir	[185]
MWCNTs	Cr(VI)	13.2	NA	Electrostatic interactions	Langmuir	[186]
IL-oxi-MWCNTs	Cr(VI)	85.83	pH 2.5–4.0, initial concentration = 20 mg/L, adsorbent mass = 0.15 g, contact time = 40 min, agitation speed = 150 r/min	Electrostatic interactions, cation-π interaction,anion-π interaction	Freundlich	[186]
Raw CNTs	Cd(II)	1.66	pH 7, initial concentration = 1 mg/L, adsorbent mass = 50 mg, contact time = 120 min, agitation speed = 150 r/min	Electrostatic interactions	Langmuir	[187]
O-MWCNTs	Pb(II) Cd(II) Cu(II) Zn(II)	76.7 32.2 15.3 13.6	pH 5, initial concentration = 40 mg/L adsorbent mass = 5 mg, contact time = 30 min, , T = 25°C	Electrostatic interaction	Langmuir	[188]
Acid modified CNTs	Cd(II)	4.35	pH 7, initial concentration = 1 mg/L adsorbent mass = 50 mg, contact time = 120 min, agitation speed = 150 r/min	Electrostatic interactions, chemisorption, ion exchange	Langmuir	[189]
MWCNTs	Hg(II)	25.641	pH 7, initial concentration = 0.1 mg/L, adsorbent mass = 0.5 g/L, contact time = 120 min, T = 25°C, agitation speed = 150 r/min	Chemisorption	Freundlich, Langmuir	[190]
MWCNT-TA	Cu(II)	30.85	pH 6, initial concentration = 50 mg/L, adsorbent mass = 0.01 g, contact time = 240 min, T = 25°C, agitation speed = 220 r/min	Charge interaction	Langmuir	[191]
Raw CNTs	Cr(VI)	3.115	pH 4, initial concentration = 1 mg/L, adsorbent mass = 75 mg, contact time = 240 min, agitation speed = 200 r/min	Electrostatic interactions	Langmuir	[52]
Acid modified CNTs	Cr (VI)	1.314	pH 3, initial concentration = 1 mg/L, adsorbent mass = 75 mg, contact time = 240 min, agitation speed = 200 r/min	Electrostatic interactions	Langmuir	[52]
MWCNTs-NCO	Pb(II) Pb-201	196.10	pH 6, initial concentration = 10 mg/L, adsorbent mass = 5 mg, contact time = 30 min, T = 25°C	Electrostatic interactions	Langmuir	[126]
As-synthesized CNTs	Cu(II)	55.25	pH 7, initial concentration = 800 mg/L, adsorbent mass = 50 mg, contact time = 300 min, T = 25°C	Chemisorption, diffusion-based process	Langmuir, Freundlich	[152]
Acid-treated CNTs	Cu(II)	82.64	pH 7, initial concentration = 800 mg/L, adsorbent mass = 50 mg, contact time = 300 min, T = 25°C	Chemisorption, diffusion-based process	Langmuir, Freundlich	[152]
Chitosan-treated CNTs	Cu(II)	120.48	pH 7, initial concentration = 800 mg/L, adsorbent mass = 50 mg, contact time = 300 min, T = 25°C	Chemisorption, diffusion-based process	Langmuir, Freundlich	[152]

(*Continued*)

Adsorbent	Adsorbate	Q_m /(mg·g^{-1})	Experimental conditions	Interaction mechanism	Best fitted isotherm model	Ref.
Acid–chitosan CNTs	Cu(II)	158.73	pH 7, initial concentration = 800 mg/L, adsorbent mass = 50 mg, contact time = 300 min, *T* = 25°C	Chemisorption, diffusion-based process	Langmuir, Freundlich	[152]
PPy/O-MWCNTs	Pb(II) Ni(II) Cd(II)	408.2 409.4 392.0	pH 6, initial concentration = 100 mg/L, adsorbent mass = 60 mg/L, contact time = 60 min, *T* = 30°C, agitation speed = 200 r/min	Chemisorption, physisorption	Langmuir	[153]
Fe_3O_4/O-MWCNTs	Pb(II)	67.25	pH 5, initial concentration = 200 mg/L, adsorbent mass = 1 g/L, contact time = 360 min, *T* = 25°C	Chemisorption, electrostatic interactions, complexation, chemical bonds' interactions	Langmuir	[155]
Fe_3O_4/O-MWCNTs	Zn(II)	3.759	pH 5, initial concentration = 30 mg/L, adsorbent mass = 1 g/L, contact time = 360 min, *T* = 25°C	Chemisorption, electrostatic interactions, complexation, chemical bonds' interactions	Langmuir	[155]
KM-CNTs	As(III)	23.40	pH 3, initial concentration = 1 mg/L, adsorbent mass = 20 mg, contact time = 55 min, agitation speed = 180 r/min	Chemisorption, complexation, ion exchange, electrostaticinteraction	Freundlich	[164]
KB-CNTs	As(III)	14.23	pH 6, initial concentration = 1 mg/L, adsorbent mass = 20 mg, contact time = 55 min, agitation speed = 180 r/min	Chemisorption, complexation, ion exchange, electrostatic interaction	Langmuir	[164]
PAMAM/CNTs	Ni(II) Co(II) Zn(II) As(III)	3900 3800 3650 3500	pH 7, initial concentration = 100 mg/L, adsorbent mass = 0.03 g/L, contact time = 15 min, *T* = 25°C, agitation speed = 14000 r/min	Chelating, chemisorption, electrostatic interactions, van der Waals, encapsulating interactions, complexation	Langmuir	[165]
NaAlg-CNTs	Co(II)	456.5	pH 6.8, initial concentration = 400 mg/L, adsorbent mass = 0.1 g, contact time = 540 min, *T* = 21°C	Electrostatic interactions, $\pi-\pi$ electron-donor/acceptor interactions	Freundlich	[192]
NaAlg-HAp-CNT nanocomposite beads	Co(II)	1111.1	pH 6.8, initial concentration = 400 mg/L, adsorbent mass = 0.1 g, contact time = 540 min, *T* = 21°C	Electrostatic interactions, $\pi-\pi$ electron-donor/acceptor interactions	Freundlich	[192]
N_2H_4-SH-Fe_3O_4/O-MWCNTs	Pb(II) Zn(II)	40.0 21.28	pH 6, initial concentration = 40 mg/L, adsorbent mass = 10 mg, contact time = 30 min	Chemisorption, physisorption	Freundlich	[193]
pTSA-Pani@GO-CNT	Cr(VI)	142.85	pH 2, initial concentration = 200 mg/L, adsorbent mass = 0.02 g, contact time = 500 min, *T* = 30°C	Electrostatic interactions, hydrophobic interactions, $\pi-\pi$ skating interactions	Langmuir	[150]

(*Continued*)

Adsorbent	Adsorbate	Q_m /(mg·g^{-1})	Experimental conditions	Interaction mechanism	Best fitted isotherm model	Ref.
PAMAM/CNTs	Cu(II) Pb(II)	3333 4870	pH 7, initial concentration = 100 mg/L, adsorbent mass = 0.03 g/L, T = 25°C	Chelating, electrostatic interactions, van der Waals, encapsulating interactions, complexation	Langmuir	[151]
MWCNTs	Cd(II)	43.103	pH 8–9, initial concentration = 50 mg/L, adsorbent mass = 20 mg, contact time = 60 min, T = 45°C, agitation speed = 250 r/min	Chemisorption, electrostaticinteraction,	Langmuir	[156]
MWCNTs-COOH	Cd(II)	212.766	pH 8–9, initial concentration = 50 mg/L, adsorbent mass = 20 mg, contact time = 60 min, T = 45°C, agitation speed = 250 r/min	Chemisorption, electrostaticinteraction,	Dubinin–Radushkevich	[156]
MWCNTs-f	Cd(II)	400	pH 8–9, initial concentration = 50 mg/L, adsorbent mass = 20 mg, contact time = 60 min, T = 45°C, agitation speed = 250 r/min	Chemisorption, electrostaticinteraction,	Temkin	[156]
NiO/CNT	Pb(II)	24.63	pH 7, initial concentration = 20 mg/L, adsorbent mass = 0.1 mg, contact time = 10 min	van der Waals interaction, electrostatic attraction, chemisorption	Freundlich	[194]
Acidified functionalized MWCNTs	Pb(II) Cu(II) Cd(II) Ni(II)	166 123 101 95	pH 9, initial concentration = 100 mg/L, adsorbent mass = 0.5 g, contact time = 10 h, agitation speed = 200 r/min	Chemisorption, complexation, ion exchange,	Langmuir, Freundlich	[195]
MWCNTs	Hg(II)	71.1	pH 7, initial concentration = 100 μg/L, adsorbent mass = 0.5 g/L, contact time = 90 min, T = 20°C, agitation speed = 245 r/min	Chemisorption, electrostaticinteraction	Freundlich, Dubinin–Radushkevich	[196]
MWCNTs-OH	Hg(II)	78.9	pH 7, initial concentration = 100 μg/L, adsorbent mass = 0.5 g/L, contact time = 90 min, T = 20°C, agitation speed = 245 r/min	Chemisorption, electrostaticinteraction	Freundlich, Dubinin–Radushkevich	[196]
MWCNTs-COOH	Hg(II)	134.0	pH 7, initial concentration = 100 μg/L, adsorbent mass = 0.5 g/L, contact time = 90 min, T = 20°C, agitation speed = 245 r/min	Chemisorption, electrostaticinteraction	Freundlich, Dubinin–Radushkevch	[196]
MWCNTs-NH_2	Hg(II)	205.0	pH 7, initial concentration = 100 μg/L, adsorbent mass = 0.5 g/L, contact time = 90 min, T = 20°C, agitation speed = 245 r/min	Chemisorption, electrostatic interaction	Freundlich, Dubinin–Radushkevich	[196]
Fe_3O_4/CNT	Pb(II)	21.55	pH 6, initial concentration = 20 mg/L, adsorbent mass = 0.10 g, contact time = 40 min, T = 25°C	Chemisorption, physisorption	Langmuir, Freundlich	[197]

* Q_m: the maximum adsorption capacity based on Langmuir isotherm model; NA: not available; T: temperature; e-MWCNTs: ethylenediamine-MWCNTs; MWCNT–HAP: MWCNT–hydroxyapatite; Ag-MWCNTs: silver nanoparticles deposited multiwalled CNTs; CF-CNTs-A: Ce–Fe mixed oxide decorated multiwalled carbon nanotubes in the absence of NaSDBS; CF-CNTs: Ce–Fe mixed oxide decorated multiwalled carbon nanotubes in the presence of NaSDBS; MWCNT-OX: oxidized MWCNTs; CNT-I: iodide incorporated MWCNT; p-MWCNTs: purified MWCNTs; Al_2O_3/MWCNTs: alumina decorated MWCNTs; IL-oxi-MWCNT: ionic liquid-functionalized oxi-MWCNTs; oxi-MWCNTs: oxidized multiwalled carbon nanotubes; ionic liquid; tetra *n*-heptylammonium bromide; O-MWCNTs: oxidized MWCNTs; MWCNT-TA: tartaric acid modified MWCNT; PAMAM/CNTs: poly-amidoamine dendrimer modified carbon nanotubes; NaAlg-HAp-CNT: amide group functionalized multi-walled carbon nanotube in the network of sodium alginate containing hydroxyapatite; NiO/CNT: nickel oxide/MWCNTs nanocomposites

Many factors are effective in the adsorption of metal ions by carbon nanotubes, such as pH, adsorbent mass, contact time, temperature, the initial metal ion concentration, ionic strength, agitation speed, surface charge, competition among metal ions, and isoelectric point. These adsorption parameters should be studied and optimized to increase the absorption efficiency. The pH value of the solution is one of the significant, dominant and effective factors in the metal ions adsorption onto the adsorbents. That it can be modified the surface charge of the adsorbent and ionization degree and adsorbate specification [198–200]. There are different species of metal ions and the competing complexation reactions in aqueous solutions depending on the pH [126]. The effect of pH without the existence of the adsorbent is done in order to evaluate if there was no hydroxide precipitation throughout the experiments. The acid modification of CNTs and the major mechanism for adsorption of divalent metal ions on the modified CNTs surface are shown in Fig. 2. Generally, increasing the solution pH (higher than point of zero charge) has a direct effect on the cation ions adsorption. With the increment of pH, the surface charge of the modified CNTs becomes negative and an electrostatic attraction between the positively charged of the metal ions and negative surface charge of the CNTs occurs. Thereby, the higher adsorption of metal ions by modified CNTs happens. On the other hand, at lower pH values less cation adsorption is observed. In acidic solution, hydronium ions concentration is high, thus competition between H^+ and cation ions to interact with active sites on adsorbent could occur. Also, at low pH values the surface charge of the adsorbent becomes positive which leads to the high coulombic repulsion of metal ions. In acidic solution, sorption sites of adsorbent are protonated and inhibited from reaction with metal ions (Fig. 3). Sankararamakrishnan et al. in their study on adsorption on of Hg on the raw CNT, CNT-OX, CNT-I, CNT-S, found that the adsorption capacity of adsorbents was in the order of CNT-S > CNT-I > CNT-OX > CNT. Figure 4 represents interaction mechanism of Hg(II) with CNT-OX, CNT-I, CNT-S [181].

Chen et al. in their study, have been proposed that As(V) species are adsorbed on the CF-CNTs via surface complexation, including monodentate and bidentate complexation through the replacement of OH groups, and electrostatic interactions. The Ce(IV) of CF-CNTs causes a part of As(III) oxidizes to As(V), and adsorption process is followed by simultaneous adsorption of As(V), but the rest of the As(III) is possibly adsorbed on the Ce–Fe mixed oxide surface of CF-CNTs via the formation of monodentate complexes. Figure 5 displays the possible interaction mechanisms of arsenic ions adsorption by CF-CNTs [180]. The results indicated that the adsorption of heavy metals on CNTs depends mainly on the properties of CNTs and metal ions.

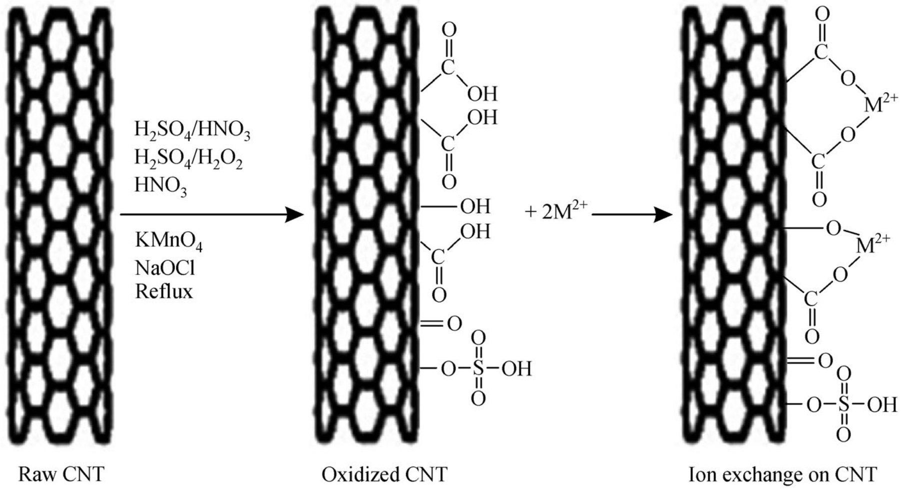

Fig. 2 Schematic diagram of the acid modification of CNTs by various methods and the major mechanism for adsorption of divalent metal ions on the modified CNTs surface

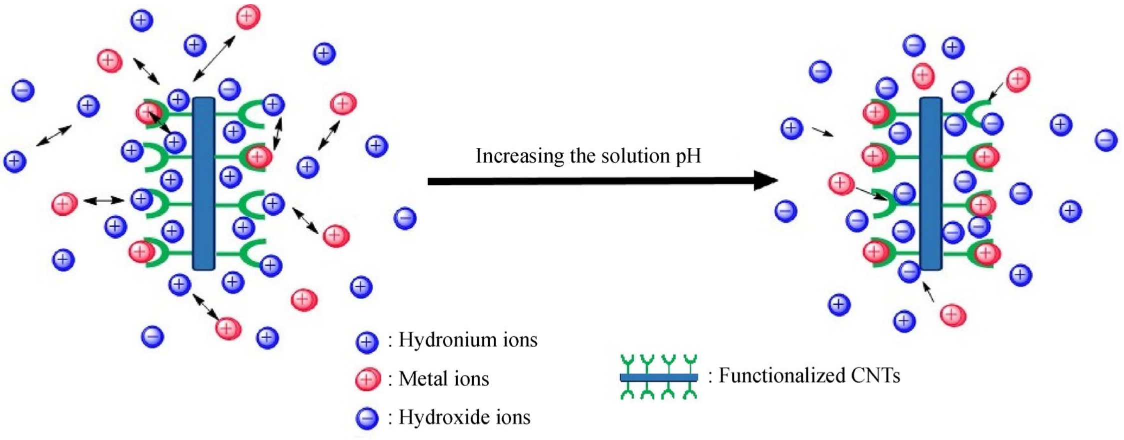

Fig. 3 Adsorption mechanism of metal ions by functionalized CNT with increasing the solution pH

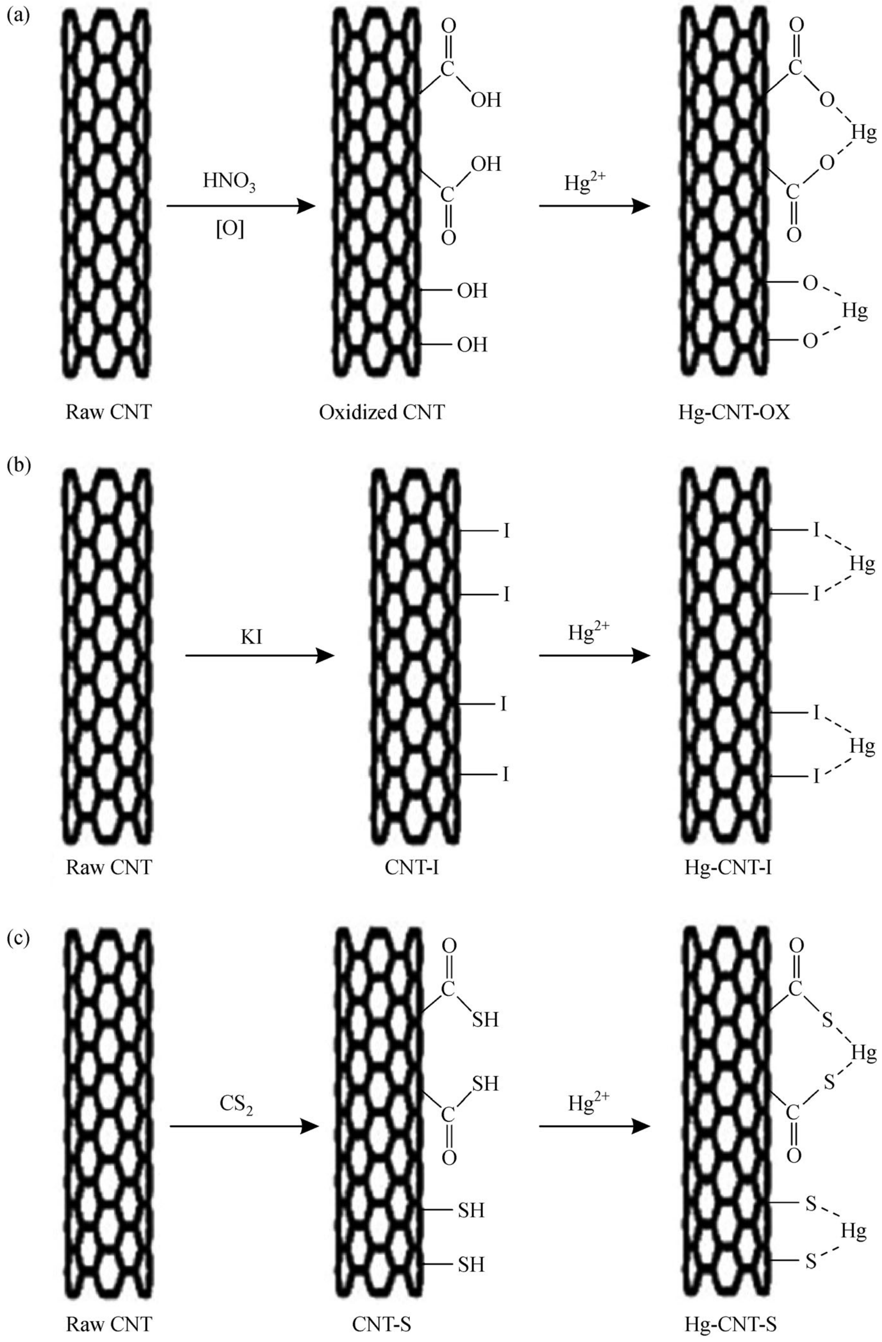

Fig. 4 Adsorption mechanism of Hg(II) with the (a) CNT-OX, (b) CNT-I, (c) CNT-S

5 Conclusions

The presence of hazardous heavy metal in wastewater has become a global concern. In latest years, CNTs have been applied to the removal of organic contaminants and heavy metals from wastewater via adsorption techniques. This review highlighted recent developments in the elimination of heavy metals by CNTs and their composites from water/ wastewater. Raw CNTs, acid modified and functionalized CNTs with other materials have been used for heavy metal ions adsorption from aqueous solution. Although, raw CNTs have shown suitable adsorption capability for a number of the metal ions, almost in all studies the adsorption capacity of the acid modified CNTs is higher than that of raw CNTs. Acid treatment is expected to open the caps of tube and decrease the length of CNTs. The oxidation of the carbon surface not only eliminates impurities and makes the surface more hydrophilic but also introduces oxygen-containing groups such as carboxyl, hydroxyl and carbonyl groups (mainly carboxylic)

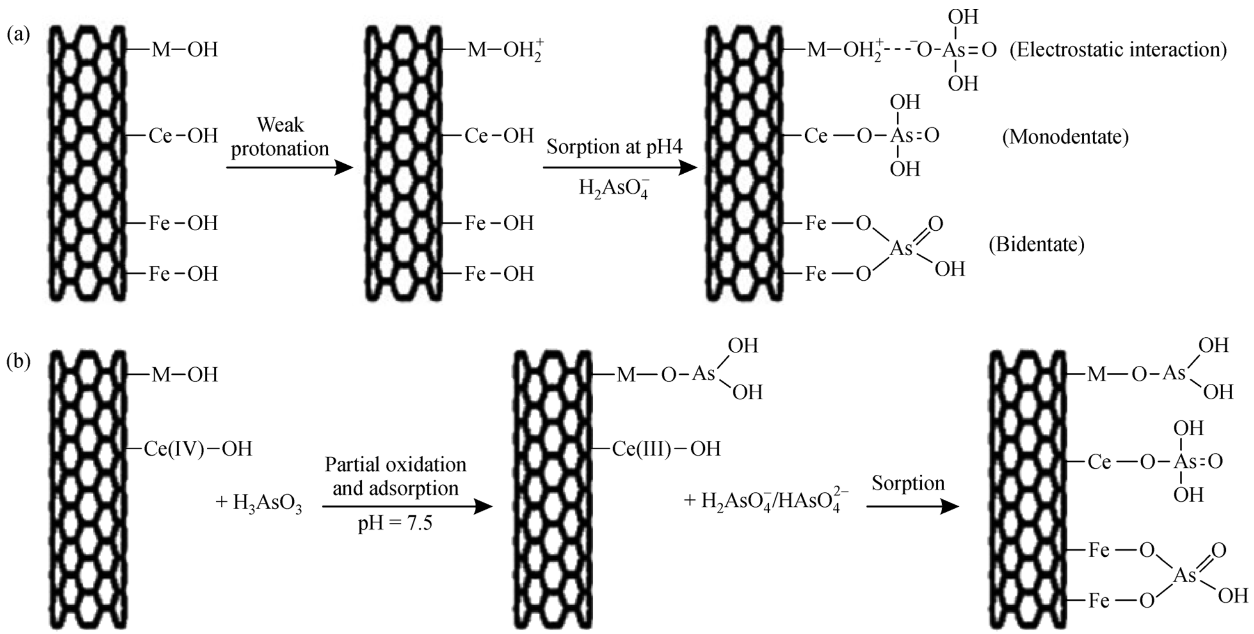

Fig. 5 Schematic diagram of the proposed mechanism for adsorption of (a) As(V) and (b) As(III) on the CF-CNTs

on their surface. A great number of adsorption sites are produced by these functional groups and thereby the CNTs absorption capacity is raised. This might be due to electrostatic interaction between the divalent heavy metal ions and the negative charge on the surface of CNTs after acid treatment. Lots of research have shown that functionalized CNTs have excellent adsorption efficiency for the heavy metal ions. Functionalization of CNTs not only increase the adsorption sites, but also makes them well dispersed in the solution. Adsorption of heavy metal ions by CNTs depends on various parameters such as pH, adsorbent mass, contact time, temperature, agitation speed, ionic strength. Process of adsorption also depends on the unblocked and the number of open nanotubes. The results of these researches demonstrate that in most of the studies, the experimental data match well with Langmuir model, and the maximum adsorption capacity is calculated by the Langmuir isotherm model. Although the price of CNTs is approximately high at the present time, the excellent regeneration capacity through desorption of metal ions offers CNT composites can be reused as an efficient adsorbent for the heavy metal ions removal from aqueous solutions.

SWCNTs have very large specific surface area, thereby they can show better adsorption properties in removal of heavy metals compared with MWCNTs. However, the Limitation of using SWCNTs is their high cost. The low price of MWCNTs makes them good candidates for adsorption, but they have low adsorption capacity compared with SWCNTs. The functionalization of MWCNTs improves their adsorption capacity. Recent researches have indicated that CNTs are promising candidates for heavy metal elimination.

6 Need for further research

Some of the CNTs production techniques are on the lab scale, as reported in the literature. Now the researchers are focusing at the economic large scale production of carbon nanotubes [201]. Improvement of the treatment techniques for the regeneration/reuse of carbon nanotubes can also decrease the price of CNTs. These techniques and finding a cost-effective way of the CNT production need to be further studied. In order to improve properties of CNTs, many researchers have been attempting to modify the CNTs surface. However, in some cases, modification of CNTs needs great quantities of chemicals, which causes environmental pollution. It is expected that due to presence of metal catalyst in raw CNTs, the toxicity of the raw CNTs is more than functionalized CNTs [202]. It is also important to study the toxicity of carbon nanotubes, their effects and finding new and safe methods for the surface modification of CNTs.

The nanotubes are too small to be easily filtered. Some strategies such as membrane filtration and ultra-centrifugation separation used to separate CNTs from the aqueous solutions, but they have the main limitations of the membrane blockage, the amount of time and high energy requirement, respectively. It seems that magnetic separation as alternative method can help to overcome these limitations.

These studies confirmed that CNTs has good potential in water treatment applications. In order to remove of heavy metals and to increase the adsorption properties of CNTs, CNTs were functionalized with different functional groups. Therefore, CNTs with different functional groups would be potential adsorbents for water treatment in future.

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