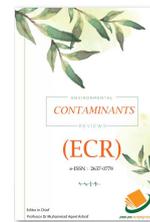


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REVIEW ARTICLE

HEAVY METAL UPTAKE FROM CONTAMINATED WATER USING CARBON NANOTUBES: A REVIEWHumaira Gul¹, Saima Nasreen^{2*}¹Department of Environmental Sciences, International Islamic University, Islamabad, 44000, Pakistan²Department of Environmental Sciences, The Women University Multan, Multan, 60000, Pakistan*Corresponding Author Email: saima_ma@yahoo.com

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ABSTRACT

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Now a day's Heavy metal pollution has become one of the most serious environmental problems. Removal of heavy metals from contaminated water is of special concern due to their recalcitrance and persistence in the environment. Methods like chemical precipitation, ion-exchange, adsorption, membrane filtration, flotation and electrochemical for heavy metal removal have been extensively studied. But these technologies have limited efficiency, extended treatment time and operation. Nanotechnology is highly developed technologies that can be applied for the complete removal of heavy metals from water. This study presents CNT that have been used to treat heavy metal from wastewater. Adsorption is most versatile and widely used method. CNTs as a novel type of adsorbent because of their unique properties like chemical stability, mechanical and thermal stability, and the high surface area. In order to reduce environmental problems, Carbon nano tubes are capable of adsorption of heavy metals. CNT are promising adsorbent material because of their hollow and layered nano sized structures, high mechanical strength and remarkable electrical conductivities.

KEYWORDS

Heavy metals, Carbon nanotubes, Wastewater, Adsorption

1. INTRODUCTION

Water Quality has become of paramount importance worldwide. Fresh water is a scarce natural resource and it is not equally distributed over a globe. Fresh water shortage is a worldwide serious concern [1]. Appropriate quality of water is not available for industrial and domestic use. Due to ongoing water shortage the continuous monitoring and Water treatment is a strategic choice. Water resources are highly polluted because of rapid industrialization. Water resources pollution is due to the indiscriminate disposal of metal ions. Metals pollution has been causing worldwide concern. The natural and anthropogenic sources introduce a certain amount of heavy metals to the environment and increase its concentration and distribution [2].

Heavy metals are defined as metallic element that has a high atomic weight and a density than that of water. It has atomic number above 20. Although it is a natural occurring element and found throughout the earth crust. Heavy metals Sources in the environment include industrial, agricultural, pharmaceutical, domestic effluents. Anthropogenic activities affect the natural geological and biological redistribution of heavy metals through air, water and soil pollution. Primary sources of heavy metals are point sources such as coal-burning power plants, mines, foundries, smelters as well as diffuse sources such as combustion by-products and vehicle emissions [3].

Amongst pollution Heavy metal is considered to be an important environmental problem due to its toxic effects and accumulation throughout the food chain and hence in the human body. The two main sources of heavy metals in wastewater are natural and human [4]. The natural sources of pollutant in waste water are soil erosion, volcanic activities, urban run offs and aerosols particulate. Human factors like metal finishing and electroplating processes, mining extraction operations, textile industries and nuclear power. Volcanic eruptions have hazardous impacts to the environment, climate and health of exposed individuals. Carbon dioxide, Sulphur dioxide, carbon monoxide, hydrogen

sulphide, organic compounds and heavy metals, such as mercury, lead and gold are also released are released during eruptions [5].

Heavy metals are not biodegradable and tend to accumulate in living organisms. The persistence of heavy metals is because of their non-biodegradable and toxicity nature. The main source of these metals in wastewater are tanneries, metal plating facilities, mining operations, fertilizer industries, batteries, paper industries and pesticides etc. Environmental contaminations occur as a result of metal corrosion, soil erosion and atmospheric deposition, of metal ions results into leaching of heavy metals from water resources to soil and ground water. Weathering and volcanic eruptions are natural processes and also significantly contribute to heavy metal pollution. Industrial sources like metal processing in refineries, coal burning in power plants, petroleum combustion, nuclear power stations and plastics, textiles. They discharge toxic heavy metals directly and indirectly into the environment [6].

Heavy metals toxicity factors include dose, route of exposure, and chemical species, as well as the age, gender, genetics, and nutritional status of exposed individuals. Metals are classified according to their degree of toxicity like arsenic, cadmium, chromium, lead, and mercury that are of public health significance [7]. Removal of these metal ions is important from the wastewaters before releasing into the environment. Toxic metal ions into food chain through waste discharges into water bodies. Heavy metals are toxic and carcinogenic. Toxic Heavy metals are identified as harmful to environment and human health. Heavy metals cause serious health problems including the respiratory, cardiovascular, hematologic, immune and reproductive systems. Toxic heavy metals are Arsenic, zinc, copper, nickel, mercury, cadmium, lead and chromium. Removal of these metals is one of the most essential processes in wastewater treatment [8]. Among them arsenic is considered as a high priority one. It is naturally present in rocks air, plants, water, soil and animals. Forest fires, volcanic activity, Erosion of rocks and minerals also release arsenic into the environment. Anthropogenic activities also release Arsenic into the environment [2]. Other Activities like pesticides, fertilizers use, fossil fuel

combustion, mining, smelting, land filling adds to releases of arsenic. Arsenic is well known toxic element and enters in water by pollutants discharge from industries, mining. Natural leaching of rocks containing arsenic, processing of mineral deposits [9].

Zinc is one of the most common heavy metals present in waste water. It is essential trace element for human health. It is a shiny bluish-white metal. Zinc is most commonly used in photocopier paper, plastics, cosmetics, and printing inks [10]. It is used in various industries like paint, batteries, smelting, fertilizers and pesticides and fossil fuel combustion. The wastewater from these industries contain large quantities of zinc. It is important for regulation of many biochemical processes and also play role in physiological functions of living tissue. High amount of zinc can cause stomach cramps, nausea, anemia skin irritations, and vomiting [8].

Nickel ions are non-biodegradable toxic metal. It is a carcinogen element and it cause serious lung, kidney problems and also pulmonary fibrosis and skin dermatitis. The major sources of nickel in waste water comes from industrial process like electroplating, batteries manufacturing, mine, metal finishing and forging [11].

Cadmium has been classified as a probable human carcinogen by U.S. Environmental Protection Agency. It is a soft, bluish-white metal. It can be easily cut with knife. It is similar respects to zinc. Cadmium and its compounds are highly toxic [10]. Cadmium exposes human health to severe risks. Chronic exposure of cadmium results in kidney dysfunction and high levels of exposure will result in death. Excess Cd is stored in the liver and kidneys. Seafood, organ meats contain highest levels among food. Metal smelters, Coal burning, Incineration of Cd-containing waste are source of Environmental contamination [12].

Lead is a bluish-gray metal present in small amounts in the earth's crust. Lead is neurotoxin element. Printing, dying discharge of battery manufacturing, and other industries are major sources of lead in waste water [13]. Other lead sources in the environment include, household dust, food container and lead-based paints. It damages to central nervous system, kidney, liver and reproductive system. The toxic symptoms include anemia, insomnia, headache, dizziness, irritability, weakness of muscles, and renal damages [14].

A reported that Chromium exists in two states: Cr (III) and Cr (VI).

Chromium (VI) is more toxic than Cr (III) [15]. It affects human physiology, accumulates in the food chain and causes severe health problems ranging from simple skin irritation to lung carcinoma. It is used in metal leather tanning, textile industries, and electroplating. Industrial emissions, leaching from hazardous waste sites and Un intended release during mining and smelting of gold, copper and other metals are sources of Environmental Contamination.

Day by day pollutants are increasing while the problems posed by traditional pollutants are not efficiently solved yet. Removal of toxic metals from water is one of the biggest challenges in ensuring safe water for all as well as protecting the environment [16]. Conventional available technologies for the removal of heavy metals from waste water include precipitation, co-precipitation, adsorption reverse osmosis, and bioremediation membrane filtration, ion exchange [17].

The shortcomings of these technologies include limited efficiency, extended treatment time and operation cost. However, only adsorption is the most versatile and widely used method. Other methods are high cost and low feasibility for small-scale industries. This show the way to the search new and highly developed technologies like nanotechnology, that can be applied for the complete removal of heavy metals from water [18]. Nanotechnology is one of these technologies that were deeply researched for its utilization in water treatment [19]. Nanotechnology has introduced different types of nonmaterials to water industry in recent year that can have promising outcomes. Nan sorbents like carbon Nano tubes (CNTs), polymeric materials, zeolites have exceptional adsorption properties and are applied for removal of heavy metals, organics, and biological impurities. It was investigated by many researchers in the last few years that carbon nanotubes (CNTs) are used in removing different types of water contaminants [20]. Carbon nanotubes are Single and Multi-walled carbon nano tubes (MWCNT) and used for removal of metal ions, such as lead, copper, cadmium, silver, nickel. CNTs are efficiently used worldwide due to their nano size, large surface area, high mechanical strength and remarkable electrical conductivities. CNT are promising adsorbent material because of their hollow and layered nano-sized structures [21].

A group researchers found that CNTs show exceptional adsorption capability and high adsorption efficiency for heavy metal removal from water [13]. The adsorption of several heavy metals by carbon nano tubes is much heavier than any other method [22].

Table 1: Conventional Methods and their Draw backs

Conventional Methods	Drawbacks	References
Chemical precipitation	It is usually adapted to treat high concentration wastewater containing heavy metal ions and it is ineffective when metal ion is in low concentration. It is not economical and can produce large amount of sludge which is to be treated with great difficulties.	[23]
Ion exchange	It is expensive, when large amount of wastewater Containing heavy metal in low concentration, so they cannot be used at large scale.	[24]
Membrane filtration	This process has high cost, and process Complexity.	Landaburu-Aguirre et al., 2009
Coagulation e flocculation	This has increased sludge volume generation and chemical consumption.	[25]
Flotation	Only limitation of this method involves high initial capital cost, high maintenance and operation costs.	[26]
Electrochemical	High initial capital investment and the expensive electricity supply has its disadvantages.	[27]

2. ADSORPTION

Heavy metals removal from waste water by adsorption is now a day's recognized and as well effective and economic method. Design and operation of these methods offer flexibility. High quality treated effluents are produced by this method. Sometime Adsorption is reversible, adsorbents can be regenerated by suitable desorption process.

3. CARBON NANO TUBE ADSORBENT

Carbon nanotubes are a new member of the carbon family. CNTs have two main types with high structural perfection. Single-walled carbon nanotubes consist of a single graphite sheet flawlessly wrapped into a cylindrical tube and multiwalled carbon nanotubes contain an array of concentric cylinders as shown below [28].

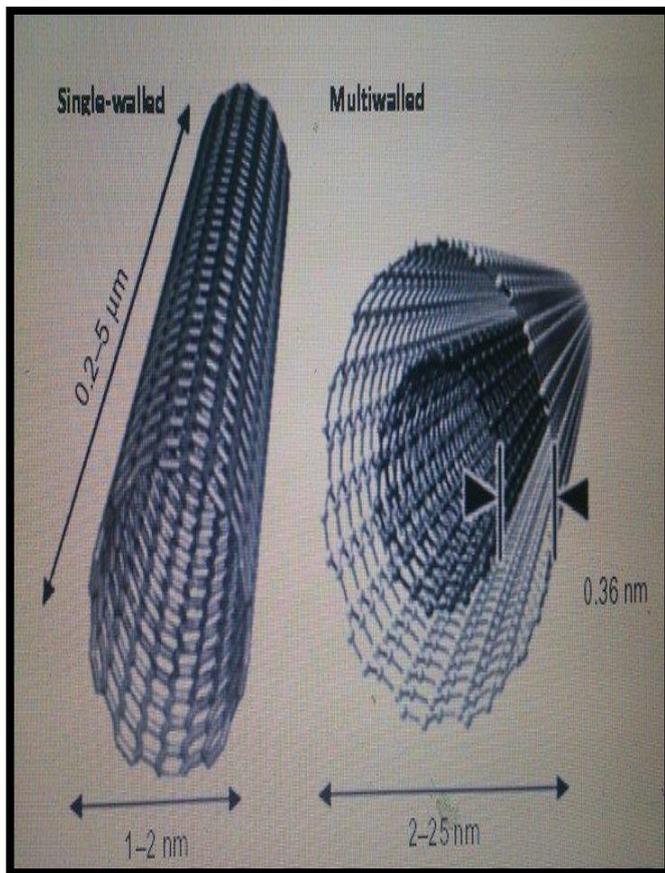


Figure 1: Carbon nanotubes Molecular structure and typical dimension of (left) single walled carbon nanotubes and (right) multiwalled carbon nanotubes [28].

They are the most promising nano structured materials. The vision of developing novel carbon-based nonmaterial has developed interest among worldwide researchers [29]. It has large adsorption capacity, which is because of their pore structure, surface area and the existence of a wide spectrum of surface functional groups [1]. For water treatment It have good anion and cation adsorption material. Their hollow and layered nanosized structures make them a good candidate as adsorbents. It has been widely studied for their excellent properties and applications. It is relatively new adsorbents CNTs [28].

4. HEAVY METAL REMOVAL FROM WASTE WATER BY USING CNT

It has been proven that CNT possess great potential for removing heavy metal such as lead, and nickel from wastewater [10,30]. These studies results showed that CNTs are capable for adsorption of heavy metals. It is very complicated mechanisms by which the metal ions are sorbed onto CNTs and appear attributable to electrostatic attraction, sorption e precipitation and chemical interaction between the metal ions and the surface functional groups of CNTs. Sorption capacities of metal ions significantly increase after oxidized by HNO_3 , NaClO and KMnO_4 solutions [1].

Some researchers found the oxygen us functional groups on MWCNTs. It plays an important role in lead adsorption to form chemical complex adsorption, which accounts for 75.3% of all the Pb adsorption capacity. Pb^{2+} can be easily regenerated from the acidified MWCNTs by adjusting pH of the solution.

Another studied for lead removal of lead showed that Oxidized MWCNTs increases due to the formation of functional groups (COOH and $-\text{OH}$), which ionize the acidified MWCNTs. The adsorption of Pb^{2+} onto acidified MWCNTs is not uniform and mainly assembled on the cap and defective sides of MWCNTs.

Another study in 2009 investigated the capability of carbon nanotubes (to adsorb lead in aqueous solution [30]. The optimum pH was 5 in which gave 85% removal of Pb (II) took place from aqueous solution. Adsorptions

capacity of Pb^{2+} is 102 mg. The highest percentage removal of lead (96.03%) can be achieved at pH 5. The adsorption properties depend significantly on the pH value. The high efficiency for Pb^{2+} removals by CNTs suggests that CNTs can be good Pb^{2+} adsorbents and have great potential applications in environmental protection.

Some researchers discussed the adsorption of Cd^{2+} by CNTs. He used hydrogen peroxide to oxidize the CNTs to reach equilibrium state with a high adsorption capacity at lower Ph [12]. The adsorption capacity is highly pH-dependent. CNTs are able to adsorb Cd^{2+} with high adsorption capacity Pore size and specific surface do not affect adsorption of heavy metal. KMnO_4 is a better oxidizing solvent for adsorption of heavy metals. At the same pH value, the zeta potential for three kinds of oxidized CNTs follows the order of $\text{H}_2\text{O}_2 < \text{KMnO}_4 < \text{HNO}_3$.

A noted that sorption kinetics follows the pseudo-second-order rate law while the sorption thermodynamics indicates the endothermic and spontaneous nature [31]. Sorption affinity between Zn^{2+} and CNT surface is stronger than that between Ni^{2+} and CNT surface. Others researchers reported that Al_2O_3 on CNT have promising potential applications in removing soluble heavy metals [32]. Carbon nanotubes were grown on micron-sized Al_2O_3 particles in an atmosphere of methane and hydrogen at 700 °C under the catalysis of Fe-Ni. The adsorption of heavy metals are in the following order $\text{Pb}^{2+} > \text{Cu}^{2+} > \text{Cd}^{2+}$.

Some researchers discussed that Zn ion is highly toxic to aquatic organisms. It has a high potential to bio-accumulate [33]. Zn ion exhibits high toxicity to aquatic organisms and it may cause high chronic toxicity in some case It is important to remove Zn from wastewater during the treatment. In water, Zn ion react with neutral or ionic compounds to form inorganic salts, stable organic complexes, inorganic or organic colloids. High adsorption capacity of Zn^{2+} suggests that purified SWCNT and MWCNT have great application for removal of Zinc from water. Single and Multi wall CNTs are better adsorbents compared to powdered activated carbon. The quantity of Zn ion in is dependent upon the solubility of these forms in which it is present. This study explained adsorption increased when the pH is adjusted from 1-8, achieved maximum between pH 8-11 and decreased at pH 12. The time taken for the adsorption to reach the equilibrium is 1 hour, while for the powdered activated carbon it took 2 hours. A comparison between the CNTs and commercial PAC for the adsorption of Zinc was also carried out. The maximum adsorption capacities of Zinc calculated by the Langmuir model for the Single and Multi-wall carbon Nano tubes and PAC were 43.66, 32.68 and 13.04 mg g⁻¹, respectively.

A studies reported that MWCNTs have been produced by decomposing acetylene gas in the presence of Ferrocene as a catalyst [10]. The oxidation of the as-produced MWCNTs improved their hydrophilic properties and increased their cation exchange capacity. CNTs due to the formation of different functional groups on the CNTs surface. Langmuir parameters showed that the oxidized CNTs have greater adsorption capacity for nickel ion removal from water than that for the peanut shells or as-produced CNTs.

A studies reported that the negative value of ΔG^0 suggested that the adsorption of Cu^{2+} onto as-produced and modified CNTs were spontaneous [7]. Positive ΔH^0 values revealed that the adsorption of Cu^{2+} onto CNTs was endothermic, which was supported by the increase in the adsorption of Cu^{2+} with temperature. This study investigated the equilibrium adsorption of Cu^{2+} onto as-produced and modified CNTs at various pH values, ionic strengths, and temperatures. This study suggests that HNO_3 and NaOCl modification not only increased the area of active adsorption sites of CNTs but also increased the proportion of available adsorption sites; additionally, NaOCl modified the surface of as-produced CNTs more effectively than HNO_3 .

Another study for removal of Ni ion showed that the adsorptions of Ni on the magnetic composites are strongly dependent on pH and ionic strength [27]. The adsorption capacity of the magnetic composites is much higher than that of MWCNTs and iron oxides. The magnetic composites can be prepared with a high adsorption capacity MWCNTs.

Other group researchers reported that environmentally friendly adsorbents CNTs immobilized by calcium alginate were designed for copper adsorption [13]. Results showed that copper removal efficiency of the CNTs/CA is high and reaches 69.9% even at a low Ph. The CNTs/ CA copper adsorption capacity can attain 67.9 mg/gatthe copper equilibrium concentration of 5 mg/.

Table 2: Brief Summary of Removal of Heavy Metals Using Non-Modified CNT

Heavy Metals	CNTS	Initial Concentration mg/L	CONDITIONS	Adsorption capacity mg/L	AUTHORS
Pb ⁺²	MWCNT	10_80	pH 5, dosage 0.05g, Temperature 280–321K	1	[12]
Pb ⁺²	MWCNT	10_60	pH 5, Temp 298–323K Dosage 1 g	4 18	Wang et al., 2007a
Zn ⁺²	SWCNTs MWCNTs PAC	10_80	pH 8-11, Contact time 60 mintues	43.66 32.68 13.04	[33]
Ni ²⁺	As produced CNTS	1_200	pH 2-8, Adsorption time 20-50 minutes, CNTs dosage 20 mg/l	18.00	[10]
Ni ²⁺	MWCNTs/iron oxide	6.0	pH 6.5, Temperature = 25 ± 2 °C	9.30	[27]
Pb ⁺²	CNT	40	contact time 80 min , agitation speed 50 r/min	102	[30]
Pb ⁺² Cu ⁺² Cd ⁺²	CNT	5_60	pH 5, dosage 0.05g, Temperature 298K	62.50 27.03	[32]
Cu ⁺²	CNT	43.1	pH 2_9, Ionic strength 0.01M, Temperature 280–320K, CNT dosage 0.5 g	8.25	[7]

Table 3: Brief Summary of Removal of Heavy Metals using Functionalized CNT

Heavy Metals	CNTS	Initial Concentration mg/L	CONDITIONS	Adsorption capacity mg/L	AUTHORS
Ni ²⁺	MWCNTs/oxidized CN	0.2	CNTs dosage 1g, pH 1	10	[10]
Pb ⁺²	Oxidized MWCNTs Pristine Annealed	10-100	Temperature 298K Oxidation 1–6 hours, CNTs dosage 0.025g	91 7.2 21	[7]
Cd ⁺²	Oxidized CNTs	1	CNT dosage 10 mg, Contact time 120 minutes, Agitation Speed 150 rpm, Temperature 298K, pH 7	34.36	[30]
Cd ⁺²	Oxidized CNTs HNO ₃ KMnO ₄ H ₂ O ₂	1.1–9.50	pH 12, CNTs dosage 0.05–0.20 g, Contact time 4 hours	5.1–11.8 11–19 2.6–8.4	[13]
Ni ²⁺	Oxidized SWCNTs MWCNTs	10–80	Ph 1_8, CNTs dosage 0.05 g, Agitation speed 180	47.85 38.46	[31-35]

5. CONCLUSIONS

The heavy metal pollution is a major concern for environment conservativity and human health. Carbon nano tube adsorbent is a recognized method for the removal of heavy metals from waste water at low concentration. Up till now, removal process has not reached the

optimum conditions. CNTs are new members of the carbon family and have special characteristics of shape and novel properties. Many researchers reported that removal of heavy metals using CNTs from water treatment. The order of heavy metal ions removed from waste water by CNT above depend on properties such as ionic strength, pH, foreign ions, CNT mass, contact time, initial metal ion concentration, and temperature.

Synthesis conditions have influences on the nature of the CNTs product formed. Conditions like temperature, gas composition and the nature and composition of metallic catalysts leading to CNT formation, which, in turn, affect the properties of the final product. These processes are still at experimental stages only, but we consider them to be very much promising at least at the pilot plant. Wastewater treatment in large scale to remove heavy metals and separation in conjunction with carbon nanotubes is expected to create a major breakthrough in the coming future.

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