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ORIGINAL ARTICLE

The Evaluation and Comparison of Single- and Multi-Walled Carbon Nanotubes in the Removal of Heavy Metals from Water

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KEYWORDS

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ABSTRACT: The water scarcity crisis and water pollution are among the key issues both in the current century and in the future. Several different techniques have been introduced for the decontamination of water pollution, among which using the nanotubes is one of the most influential. In this project, the single-walled and multi-walled carbon nanotubes were employed to test the possibility of removing heavy metal ions, such as Fe^{2+}, Cd^{2+}, Pb^{2+} and Ni^{2+}. The effects of three factors including carbon nanotubes, ions' initial concentration, and contact time were tested. According to the difference rate of nanotubes' adsorption diagrams, the maximum differences in the performance of single- and multi-walled are obtained as 30 percent for the initial solution concentration and around 5 percent for Nano adsorbent concentration. The results showed that in the different initial concentrations of metal ions, multi-walled nanotube had a better performance, while in case of changing the amount of adsorbent; the single-walled nanotube had a higher adsorption. And if the time changes, multi-walled carbon nanotube can have a higher adsorption.

INTRODUCTION

Water health and sanitation being in danger is one of the fundamental and substantial issues dealing with the so-called unidentified water pollution situations in the different uses of Nano technology. The presence of probable and potential pollutant concentrations definitely necessitates proposing the basic strategies for controlling the distribution of pollutants in aquatic systems and the possibility to change the time patterns of these variables is among the important issues, requiring some examination and study based on professional knowledge, judgment, and experience [1]. The general issue is concerned with wastewater, water pollution, or in other words, wastewater production. When the wastewater is kept in the tanks for a long time, the natural procedure of wastewater treatment starts. First, the net volume of clean running water dilutes a limited amount of wastewater, but the bacteria present in water should typically have access to enough oxygen so that it can perform its task of wastewater decomposition [1].

The heavy metals are generally defined as a group of metals with concentrations above 6000 Kg/m^3. The most important elements in this group are Lead, Cadmium, Chromium, Copper, Mercury, Nickel, Zink, Vanadium, and other metals, which are associated with toxic pollution. It should be noted that elements such as Arsenic and Selenium are also taken into consideration because of their

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specific toxic characteristics. Most of these metals (except MO) are extremely soluble at low PH and can enter the vital components of living creature [2].

The presence of metallic ions in wastewater from oil refineries, gas and petrochemical complexes, etc. has caused many environmental problems such as the soil contamination near these units, the pollution of the rivers and the death of various aquatic organisms and different diseases for the residents of these areas. These ions enter the wastewater of these complexes due to the transportation of petroleum products and the effluent resulting from catalytic processes of oil refinement. The presence of heavy metals in the industrial wastewaters is considered a major problem in discharging them into the surface waters. Some heavy metals, such as mercury, lead, copper, cadmium, chromium, and nickel, are poisonous even in very small amounts. Heavy metals are elements with harmful effects on the human health due to their stability and biological accumulation properties, which have been listed as important contaminants by the United States Environmental Protection Agency. The accumulation of heavy metals in the food chain and their sustainability in nature, as well as their discharge by many industrial activities is a known phenomenon. The increased industrial activity has always been the main cause of most environmental pollution problems and ecosystem degradation, which leads to the accumulation of toxic metals pollutants such as chromium, copper, lead, cadmium, zinc, nickel, etc. in water [1].

Nano technology is concerned with the world of invisible microscale particles, which are led through physical and chemical forces and cannot be used in macro or real scales. These particles are called Nano particles, according to some scientists. These particles have unusual, unique characteristics, which cannot be usually observed in other cases [1]. Carbon nanotubes include circular structures, consisting of carbon atoms that can be arranged into the form of one or more walls and have metallic or quasi-conductor properties as well. Extensive research and developments are conducted and achieved around the world for removing water heavy metals through using electrochemistry to identify the usages of carbon nanotubes for industrial purposes. And recently, the possibility of using the tubes was proposed for environmental purposes. Carbon nanotubes can also be used to track the pollution and conduct the possible evaluation of water pollutants and collect and provide the information related to environmental pollutants. Carbon nanotubes are categorized into two different groups: 1- single-walled carbon nanotubes and 2- multi-walled carbon nanotubes [1].

Nanoabsorbers, including carbon nanotubes, polymeric materials (such as dendrimers) and zeolites have special absorption properties and are used to eliminate heavy metals, organisms and biological impurities [3]. Carbon nanotubes have drawn a particular attention due to their specific capabilities in water treatment. They have been also proven to be effective in eliminating biological pollutants and chemical contaminants. Carbon nanotubes, as mediating absorbers, can absorb a wide range of polluting heavy metals, including Cr^{3+}, Pb^{2+}, and Zn^{2+}, organic materials such as polycyclic aromatic organic compounds (PAHs), atrazine and a range of biological pollutants, including bacteria, NOM viruses, and cyanobacterial toxins (cyanotoxins). The success of carbon nanotubes as absorbent mediators in eliminating biological pollutants, particularly the pathogens, is usually attributed to their functional, superficial, and physical properties and toxicity [4 – 9].

Carbon nanotubes form the condensed pores, through dozens of tubes connected together due to Van der Waals gravitational forces [10 – 12]. These condensed pores have the same dimensions of a Mesoporous or bigger ones [13 – 18] and can provide a wide external surface, that has the capability to destroy environmental pollutants, including the bacteria and viruses. The mechanisms through which metal ions are adsorbed into carbon nanotubes are very complicated and it seems that they are associated with some procedures including electrostatic gravity, adsorption-sedimentation, and chemical interactions between metal ions and the functional groups on the surface of the nanotubes. The Nano particles with high surfaces can be used to separate the heavy metals and deactivate the polluted surfaces. It is obvious that the chemical interactions with higher proficiency produce less pollutants and lesions.

Adsorption In carbon nanotubes takes place at four points: in the hollow interior spaces of nanotubes with open endings, in the interstitial space between the set of tubes, on the edges of the nanotubes' borders, or the external surface of the exterior carbon nanotubes [10, 19]. It is impossible to use the interior space of the carbon nanotubes for adsorption because first, carbon nanotubes have closed caps and second, even if they have open caps, the tubes' low diameter cannot comport a pollutant with normal macromolecular size. The interstitial space between the set of tubes provides the possibility to adsorb several small molecular adsorbents with low molecular weights (for example, metal ions) [12]. Thus, due to the adsorption of environmental pollutants in carbon nanotubes, the accessible external surface and the presence of condensed pores with dimensions bigger than those of the mesophorous is highly significant. The technology of using Nano materials is a recent topic which has been the center of attention in the last few decades. In the past, Nano materials were used in research conducted on removing chromate from liquids, checking the adsorption of cyanide and phosphorous, as well as adsorbing the petroleum and pharmaceutical pollutants and extracting several new synthetic Zeolite. For example, in 2015, Amira M. Mahmoud et al. explored the amount of lead removed from liquid in a paper, entitled as "The Different Methods of Adsorbing Heavy Metals from Liquids, Using Nickel Oxide Nano Catalysts"; in this laboratory research, they found that the Nickel Oxide Nanoparticles produced through using an organic solvent were more influential in removing the lead pollutants, compared to the Nickel Oxide produced through sedimentation method [20]. In another research conducted by Pragnesh N.Dave in 2014, entitled as "The Use of Iron Oxide Nano Materials in Removing the Heavy Metals", found that the magnetic Nano materials had a proper function in removing the heavy metals from liquids, due to their magnetic property in magnetic fields; further, the possibility of repeatedly using the magnetic Nano adsorbents helps to significantly reduce the expenses of this test [21]. In 2012, another researcher named Xiangtao Wang et al. in a paper called "Adsorbent Nano Materials in Removing Heavy Metals during Wastewater Treatment" concluded that adsorbent Nano materials have the capability to optionally remove some heavy metals [22].

In 2009, studying some specific metal samples and using the different methods of producing Nano materials, Y.C sharma in a paper entitled as "Using Nano Adsorbents for Removing Heavy Metals from Wastewater and Water" found that these Nano materials had a good performance in removing heavy metals and the liquids' treatment, but these materials should be removed from the liquid after the adsorption process is over [23].

Carbon nanotubes are divided into two single-wall (SWCNT) and multi-wall (MWCNT) groups. The multi-wall nanotubes are composed of interleaving layers and the distance between the layers is often equal to 0.375-0.342 nm. The thinnest synthesized nanotube has a diameter of about 0.23 nm [3].

Carbon nanotubes are different from each other depending on the type, technique, synthesis technology and the number of walls, but they are similar regarding the following properties [3].

Electrical and thermal conductivity and semi-conductivity: In carbon nanotubes, the carbon atoms are bound together in a six-faceted form and act like a wire. The torsion angle of the nanotubes, which is the angle between the axis of its hexagonal pattern and the tube axis, as well as the change in the radius, are important in the conductivity and semi-conductivity of the nanotubes. In addition, the carbon nanotubes have a high thermal conductivity [3]. Electron transfer by a throwing method: By entering an electron from one side, another electron is pulled out of the other side of the nanotube. The wall smooth surface: This feature increases the passage rate of gas from the nanotube compared to the microporous membranes used to separate gases and can be useful for separating nitrogen and oxygen from the air. Sensitivity to force and pressure: Pressure can reduce or increase the conductivity in the nanotubes since it changes the quantum structure of the electrons. Unique mechanical properties: High flexibility, high-range vibration, and radiation with a steep angle Diffusion and absorption of light: The carbon nanotubes absorb or dispose of infrared light. Surface condensation: The surface condensation of nanotubes is very high, which results in their high strength. In other words, they have a very high surface-to-volume ratio. Magnetic moment: Nanotubes represent magnetic properties by placement in a magnetic field. Storage of energy and atoms: In carbon nanotubes, all three carbon atoms have the storage capacity of 3 lithium atoms. Also, the energy storage capacity of nanotubes is several times that of the graphite electrodes. Electric voltage generation: This feature is used to make liquid flow sensors since a high electrical voltage is generated by the passage of a liquid through the single-wall nanotubes. Tensile strength: At high temperatures, the strength and tensile strength of the nanotubes and their conductivity increase. Electrical mobility factor: At ambient temperature, the nanotubes have an electrical mobility coefficient higher than any material. Young modulus: The nanotubes have a high Young modulus and are very elastic [3].

In this research, the amount of metal ions pollutant adsorbed through single- and multi-walled carbon nanotubes was explored. Different tests were conducted in six different levels to explore the parameters of time effect, liquid concentration effect, as well as the effect of adsorbent amount on removing ions such as Nickel (II), Cadmium (II), Lead (II), and Iron (II), using the two carbon nanotubes 1) single-walled and 2) multi-walled.

MATERIALS AND METHODS

To conduct the test, first, a certain amount of a liquid with certain concentration of heavy metals was chosen as the standard solution. Then, a certain amount of the adsorbent is added to the solution and after some time, when some of the pollutants were adsorbed by carbon nanotubes, the standard solution is filtered and the concentration of the obtained solution, which also contains the remaining amount of adsorbent, is measured.

Adsorption Spectroscopy Analyzer (UV-VIS) was employed to measure the amount of remaining pollutants in the solution and exploring the adsorbed materials. The amount of adsorbed metal ions, measured through Beer-Lambert law, is also shown on the analyzer.

1) $R = C / C0 \times 100$

In this equation, parameters are defined like this:

C: The concentration of the solution under evaluation

C_0: The concentration of standard solution

Evaluating the parameter of initial concentration amount effect, with the fixed amount of adsorbent on the amount of pollutants adsorption

After conducting the tests in six levels, the data obtained for the concentration of the new solution, with pollutants adsorbed in part, is presented in the following tables (Tables 1 & 2).

The pollutants under evaluation: Nickel, Cadmium, Lead, Iron ions.

Table 1. The comparison of for ion adsorption efficiency through single carbon nanotubes in different initial concentrations and a fixed adsorbent concentration (w=1)

C_0	swnt							
	Ni^{2+}	R (%)	Cd^{2+}	R (%)	Pb^{2+}	R (%)	Fe^{2+}	R (%)
0.15	0.03	20.00	0.04	23.33	0.03	21.33	0.04	23.33
0.25	0.09	36.00	0.10	40.40	0.10	39.60	0.11	43.20
0.43	0.16	37.21	0.17	39.77	0.17	38.37	0.18	41.40
0.55	0.29	52.73	0.32	58.36	0.32	57.64	0.32	58.91
0.67	0.41	61.19	0.44	65.67	0.43	64.33	0.45	67.31
0.89	0.63	70.79	0.63	71.12	0.63	70.90	0.64	72.13

Table 2. The comparison of for ion adsorption efficiency through multi-walled carbon nanotubes in different initial concentrations and a fixed adsorbent concentration (w=1)

C_0	mwnt							
	Ni^{2+}	R(%)	Cd^{2+}	R(%)	Pb^{2+}	R(%)	Fe^{2+}	R(%)
0.15	0.04	25.87	0.04	29.07	0.04	29.53	0.04	27.33
0.25	0.10	39.52	0.11	43.84	0.12	46.92	0.11	43.20
0.43	0.17	39.26	0.18	41.77	0.19	43.56	0.17	40.47
0.55	0.30	54.33	0.33	59.93	0.33	60.60	0.33	59.27
0.67	0.42	62.51	0.45	66.96	0.46	68.70	0.44	65.67
0.89	0.64	71.78	0.64	72.09	0.65	73.18	0.64	71.92

Looking at the tables 1-2, one may conclude that with the increase in the initial solution concentration and the fixed amount of concentration in single- and multi-walled carbon nanotubes, the adsorption rate increases for Nickel, Cadmium, Lead, and Iron ions. Furthermore, according to the adsorption efficiency (R %) presented in these tables, it was proved that in the exploration of initial solution concentration parameter, the multi-walled carbon nanotubes had a better performance and higher metal adsorption rate, compared to the single-walled carbon nanotubes.

To have a better review, the diagram showing the difference in single- and multi-walled nanotubes' efficiency for different ions are presented in Figure 1.

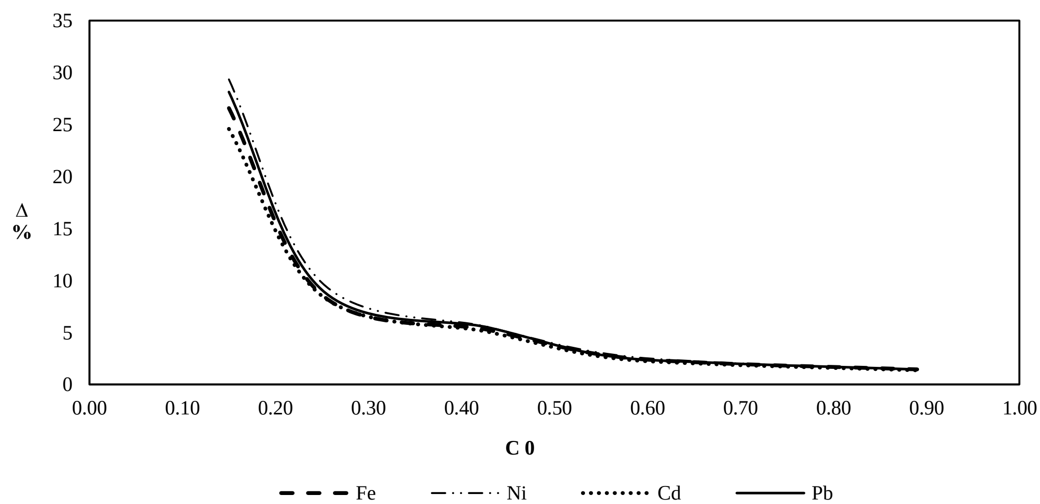

Figure 1. The diagram showing the difference rate of removing ions in single- and multi-walled nanotubes, based on the initial concentration.

The percentage of the difference between the adsorbtion of single and multi-walled nanotubes are calculated as follows:

2) $\Delta=\frac{||R_m-R_s||}{R_s}*100$

In this equation,parameters are defined like this:

R_m: the adsorbtion efficiency of ion for multi-walled nanotubs

R_s: the adsorbtion efficiency of ion for single-walled nanotubs

According to the figure, it can be observed that with the increase in Nano adsorbent concentration, the difference in adsorption rates in the single- and multi-walled carbon nanotubes decreases. The effects of other parameters affecting the adsorption of ions under evaluation as well as the efficiency measured for each of them are presented in the Tables 3-8.

Table 3. The comparison of Nickel ion adsorption efficiency through single- and multi-walled carbon nanotubes in a fixed initial concentration and different adsorbent concentrations (c=0.591)

W	**swnt**		**mwnt**	
	Ni^{2+}	**R (%)**	**Ni^{2+}**	**R (%)**
0.10	0.11	18.78	0.11	18.19
0.20	0.14	23.01	0.13	22.42
0.30	0.20	34.01	0.20	33.42
0.40	0.24	40.95	0.24	40.36
1.00	0.36	61.08	0.36	60.49
1.50	0.41	69.54	0.41	68.95

Table 4. The comparison of Cadmium ion adsorption efficiency through single- and multi-walled carbon nanotubes in a fixed initial concentration and different adsorbent concentrations (c=0.325)

W	**swnt**		**mwnt**	
	Cd^{2+}	**R (%)**	**Cd^{2+}**	**R (%)**
0.10	0.09	27.38	0.08	25.91
0.20	0.12	37.23	0.12	35.75
0.30	0.14	43.38	0.14	41.91
0.40	0.17	52.00	0.16	50.52
1.00	0.19	59.69	0.19	58.22
1.50	0.24	72.31	0.23	70.83

Table 5. The comparison of Lead ion adsorption efficiency through single- and multi-walled carbon nanotubes in a fixed initial concentration and different adsorbent concentrations (c=0.248)

W	**swnt**		**mwnt**	
	Pb^{2+}	**R (%)**	**Pb^{2+}**	**R(%)**
0.10	0.07	26.61	0.06	25.69
0.20	0.10	39.52	0.10	38.59
0.30	0.13	50.81	0.12	49.88
0.40	0.15	59.68	0.15	58.75
1.00	0.16	65.32	0.16	64.40
1.50	0.19	76.61	0.19	75.69

Table 6. The comparison of Lead ion adsorption efficiency through single- and multi-walled carbon nanotubes in a fixed initial concentration and different adsorbent concentrations (c=0.366)

W	**swnt**		**mwnt**	
	Fe^{2+}	**R (%)**	**Fe^{2+}**	**R%**
0.10	0.10	27.05	0.10	25.98
0.20	0.11	30.87	0.11	29.81
0.30	0.14	39.34	0.14	38.28
0.40	0.19	52.46	0.19	51.39
1.00	0.23	63.66	0.23	62.60
1.50	0.27	74.04	0.27	72.98

According to the Tables 3-6, it can be concluded that in case of having a fixed initial solution concentration, with the increase in the Nano adsorbent concentration for adsorbing certain ions, the adsorption rate increases as well. Further, based on these Tables and the comparison of the adsorption efficiency of single- and multi-walled carbon nanotubes, it can be observed that the single-walled carbon nanotubes showed a better performance, compared to the multi-walled carbon nanotubes and the amount of heavy metals' adsorption is higher in these tubes. Similarly, the diagram showing the difference rate of nanotubes' efficiency is presented in Figure 2.

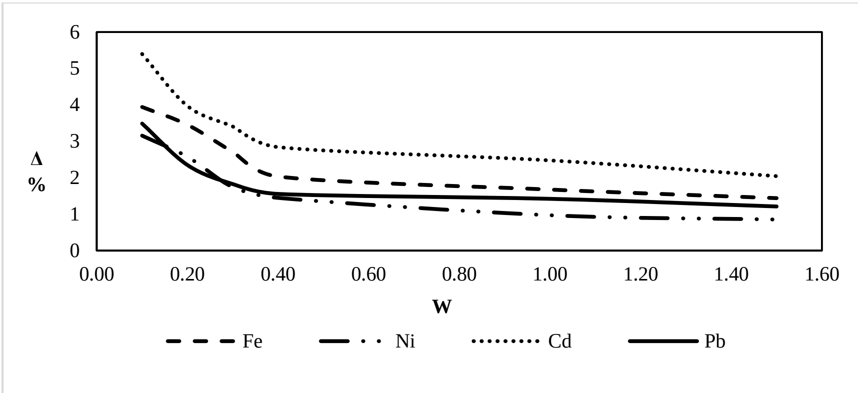

Figure 2. The diagram showing the difference rate of removing ions in single- and multi-walled nanotubes, based on the initial concentration

According to Figure 2, it is observed that with the increase in the Nano adsorbent concentration, the difference rate of adsorption in single- and multi-walled nanotubes decreases. In Tables 7 & 8, the effect of time factor is reported

Table 7. The comparison of for ion adsorption efficiency through single carbon nanotubes in fixed initial and adsorbent concentrations and varying times (w=1) (c=0.6)

T(min)	swnt							
	Ni^{2+}	R (%)	Cd^{2+}	R (%)	Pb^{2+}	R (%)	Fe^{2+}	R (%)
10	0.13	20.83	0.13	22.33	0.121	20.17	0.129	21.50
30	0.17	28.17	0.17	28.50	0.167	27.83	0.184	30.67
60	0.24	39.17	0.25	41.50	0.232	38.67	0.241	40.17
90	0.40	66.83	0.513	85.50	0.399	66.50	0.411	68.50
120	0.41	68.83	0.514	85.67	0.401	66.83	0.415	69.17
150	0.42	70.50	0.515	85.83	0.404	67.33	0.417	69.50

Table 8. The comparison of for ion adsorption efficiency through multi-walled carbon nanotubes in fixed initial and adsorbent concentrations and varying times (w=1) (c=0.6)

T (min)	mwnt							
	Ni^{2+}	R%	Cd^{2+}	R%	Pb^{2+}	R%	Fe^{2+}	R%
10	0.13	22.10	0.14	23.47	0.13	21.40	0.14	22.70
30	0.18	29.47	0.18	29.67	0.17	29.05	0.19	31.90
60	0.24	40.43	0.26	42.63	0.24	39.90	0.25	41.40
90	0.41	68.12	0.520	86.65	0.407	67.77	0.418	69.70
120	0.42	70.12	0.521	86.80	0.408	68.07	0.422	70.35
150	0.43	71.78	0.522	86.97	0.412	68.60	0.424	70.73

Looking precisely at tables 7-8, it can be perceived that having fixed initial and nanotube concentrations, with the increase in the contact time of the solution with Nano adsorbent, the adsorbent rates for all the four ions increase as well. Based on the comparison of adsorption efficiency presented in these tables, it is perceived that in this test, the multi-walled carbon nanotubes showed a better performance, compared to the single-walled carbon nanotubes, and had a higher amount of heavy metals adsorption. Besides, in the cases taking longer than 90 minutes, the adsorption increasing trend slowed down and the adsorption rate diagram based on time had an almost steady, similarly, the diagram showing the difference rate of nanotubes' efficiency is presented in Figure 3.

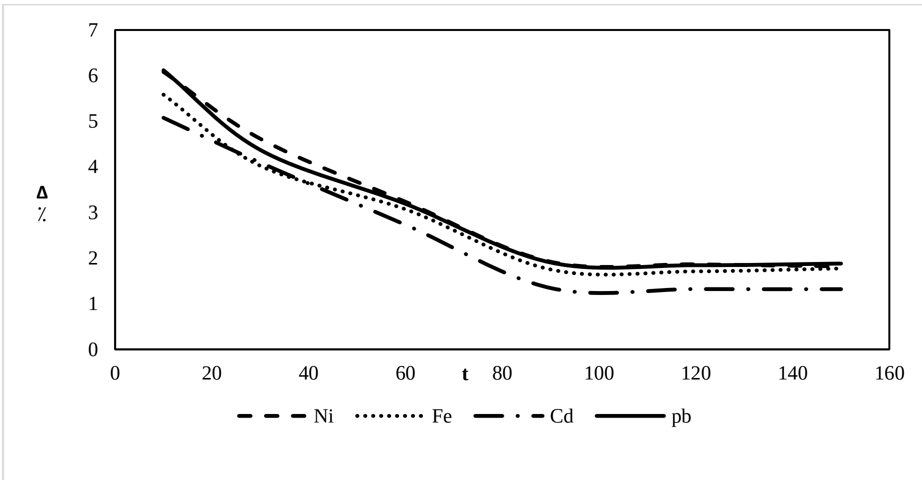

Figure 3. The diagram showing the difference rate of removing ions in single- and multi-walled nanotubes, based on the contact time

Furthermore, according to diagram 3, showing the difference rate of adsorption in single-walled nanotubes, compared to multi-walled nanotubes, one may find that with the increase in contact time, the difference rate decreases.

CONCLUSIONS

After studying the tests, diagrams, and observations on the adsorption of heavy metals, four metal ions present in polluted water and wastewater were selected as the sample and the standard solutions with certain ion volume and concentrations and single- and multi-walled carbon nanotubes were prepared and the parameters of solution initial concentration, Nano adsorbent concentration, solution and Nano adsorbent contact time were tested. The obtained result is as follows:

A) When the Nano adsorbent concentration and time are fixed and the initial solution concentration is selected as the test's variable, for all the four different heavy metals, the adsorption efficiency is directly associated with initial solution concentration. Besides, in these conditions, multi-walled carbon nanotube had a more proper performance in pollutant adsorption, compared to the single-walled carbon nanotube.

B) When the initial solution concentration and time are fixed and the Nano adsorbent concentration is selected as test's variable, for all the four different heavy metals, the increase in the amount of Nano adsorbent is accompanied by the rise of adsorption efficiency. Further, in these conditions, single-walled carbon nanotube had a more proper performance in pollutant adsorption.

C) When the initial solution concentration and carbon nanotube concentration are fixed and the contact time of solution and Nano adsorbent is selected as test's variable, for all the four different heavy metals, the adsorption efficiency diagram based on time has an ascending trend. Further, multi-walled carbon nanotube had a more proper performance in pollutant adsorption, compared to single-walled carbon nanotube.

D) According to the difference rate of nanotubes' adsorption diagrams, the maximum differences in the performance of single- and multi-walled are obtained as 30 percent for the initial solution concentration, around 5 percent for Nano adsorbent concentration, and around 6 percent for contact time parameter; based on these numbers, the initial solution concentration is introduced as the most influential test variable.

Generally, it can be estimated that compared to single-walled carbon nanotubes, the multi-walled carbon nanotubes has a higher adsorption capability due to their porous form and having more surface rippling. It should be taken into consideration that the different nanotubes' cells, being double open-ended, double close-ended, and open-ended, play a significant role in the amounts and kinds of pollutants adsorption. Nowadays, taking this capability into consideration and categorizing the nanotubes based on it is not possible but it is expected that the double open-ended carbon nanotube has a higher interior adsorption, due to the possibility of passing the pollutants through itself and the double close-ended nanotube has a higher surface adsorption.

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