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Comparison of carbon nonotubes and activated alumina efficiencies in fluoride removal from drinking water

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Abstract

Fluoride is a kind of anions, which makes its way to the water resources through diverse natural and human activities. Moreover, its health effects are so important and can vary based on the amounts of the intakes to the body. The aim of this study was to investigate fluoride removal efficiency of single and multi-wall carbon nanotubes (SWCNs & MWCNs) from water, and its comparison with the removal efficiency of two types of fine powder and 150 mesh activated alumina. Results show that, using SWCNs, the highest removal efficiency was achieved in pH=5 and sorbent concentration of 0.5 g/L. Moreover, with the increasing of pH the removal efficiency decreased. Meanwhile, fluoride removal efficiency increased with the increasing of the sorbent dosage and decreasing of the initial concentration of fluoride. In conclusion, the SWCNs had higher removal efficiency in comparison with the removal efficiency of both types of the activated alumina in optimum conditions.

Keywords: Fluoride, carbon nanotubes, activated alumina, water treatment

Introduction

Fluorine is one of the elements of halogens and exists abundantly in crust, especially in some organics and stones (WHO, 2004). Fluoride deficiency may cause dental caries and excessive use of its standard may cause dental disease, liver and skeletal fluorosis (Harrison, 2005; Xiong, 2007). Fluorosis can cause weakness of dental and skeletal structure and stagnate the growth. WHO has determined 0.7 mg/L for optimum range of flouride to reduce dental caries and avoid fluorosis in tropics and up to 1.2 mg/L in cold regions (Nanbakhsh & Saei, 2002).

Major sources of fluoride entrance to water supplies include water contact with mineral compounds containing fluoride and discharge of industrial wastewater such as effluents from semiconductors glass factories (Rasheed & Jamhour, 2005). Production in terms of fluoride toxicity and hazards of additional dosage, fluoridation of drinking water has been stopped in some countries (Rasheed & Jamhour, 2005; Chidambaram *et al.,* 2003). Ecological study by Amini and Colleagues showed that in areas where water fluoride levels were higher the prevalence of hypertension and systolic blood pressure was also higher. High fluoride waters in large parts of the geographical belt with marine sediments in the mountains volcanic and granitic rocks. For example, the first range can be seen in Iran, Iraq, Turkey, and Mediterranean or from Algeria to morocco. Other items can also be seen in southern United States of America, Southern Europe the best example for high fluoride waters of volcanic origin belongs to eastern Africa from Jordan valley to Sudan, Ethiopia, Uganda, Kenya, and Tanzania. In areas such as India, Pakistan, China, Sri Lanka, Thailand, South and West of Africa with volcanic alternative rocks high levels of fluoride is reported in groundwater.

In 28 states of China numerous fluorosis are reported. Overall, it seems that about 60-70 million people in India and 2.7 million in China are at risk of fluorosis. In the study conducted by UNICEF, fluorosis was confirmed in at least 27 countries of the world (AWWA, 1999). In rural parts of North Rajastan of India, average of fluoride was 2.82 mg/L in drinking water (Suther *et al.,* 2008). In North of Africa, the amount of fluoride in groundwater is reported more than 20 mg/L and in south parts of California this amount is more than 5 mg/L (Tor, 2007). In some parts of Iran, fluoride concentration is higher than standard in drinking water, including the provinces of Hormozgan (Bandar Abbas, Bandar Langeh, and Queshm), Yazd (Ardakan), Hamedan, Tehran, Kerman (Shahre babak, Kuh Bonan area), Khorasan – e – Razavi, Southern Khorasan, Bandar Boushehr (especially Borazjan & Dashtestan), Semnan, Zabol and Zahedan in Sistan and Balouchestan.

Considering undesirable health effects resulting from fluoride increasing in water, especially ground water resources and due to the high use of ground water in many cities, it is necessary to remove excess fluoride from water with appropriate methods. Many methods including adsorption, chemical sedimentation, membrane processes, and ion exchange have been used so far for excess fluoride removal (Rasheed & Jamhour, 2005; Tor, 2007). However, many of these methods cannot be applied in developing countries and low-income areas, because of their high cost and complexity (Chidambaram *et al.,* 2003). For example, the chemical deposition technique is used widely in refineries of the country. However, the amount of excess sludge produced in this method requires special attention for disposal. Membrane processes as a successful method has been considered to remove fluoride from drinking water, but this method is also very costly. Anionic exchange resins are less considered for fluoride removal as they have less

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Table 1. Profile of nanotubes used in the study

*Type of NCTs**	*Internal Diameter (nm)*	*External Diameter (nm)*	*Length (µm)*	*Specific Surface (BET)m^2/gr*	*(Heat conductivity w/mv)*
Single wall	*0.8-1.1*	*1-2*	*10*	*700*	*3000*
Multiwall	*----*	*10-30*	*10*	*270*	*1500*

**NCT: Nanocarbon tubes*

capacity and are expensive (Rasheed & Jamhour, 2005).

Today using nanotechnology has expanded in all fields and has been considered as a powerful technology. Carbon nanotubes have been caught attention of experts in different fields of nanotechnology, including environment. In fact, carbon nanotubes are pages of carbon atoms that move in some parts such as rollers. High specific surface, high reactivity, abundant heat resistance and mechanical and chemical characteristics are some properties of these materials. Carbon nanotubes are hollow cylindrical structures that they can be imagined in the form of graphene rolled pages. These Materials, based on type of construction and number of carbon layers, are divided into two groups: single-wall structures' and multi-wall (Bahari *et al.,* 2010).

Multi-wall types are made of graphite fibers, while single-wall nanotubes are made of fiolern fibers. The single wall type is made of a graphene page, which is formed as a flat array of benzene molecules with single and double bonds (Rasheed & Jamhour, 2005; Chidambaram *et al.,* 2003; Majengera & Mkongo, 2003). According to various studies, these nanotubes are able to remove organic and inorganic materials, heavy metals, some semi metals, and some microbial and viral contaminations from human environment.

Activated alumina is one the substances used for fluoride removal successfully. It is considered as a good sorbent of fluoride and is one of the most essential materials used for fluoride removal with high switching capacity for this ion Also its ion exchange capacity will not be affected by sulfate and chlorine ions of water (Majengera & Mkongo, 2003).

Adsorption action by activated alumina is a physicochemical process, during which existing ions in water inlet will be adsorbed on oxidized surfaces of activated alumina. Although the chemical reactions involved in the activated alumina are in fact, a kind of ion exchange, but activated alumina is considered as an adsorption process. In this study the performance of single-wall and multi-wall carbon nanotubes and activated alumina in reducing fluoride of water resources and the impact of different parameters on nanoparticles performance are studied.

Materials and methods

This study was conducted in lab scale and in Water and Wastewater laboratory of public health school, in Tehran University of Medical Sciences in 2010-11. Single-wall and multi-wall carbon nanotubes were provided from the national Institute of Petroleum Industry; their specifications are shown in Table 1.

Activated alumina used as an adsorbent in this study, was purchased from Aldrich Co. Code No. 19, 944-3 with mesh size of 150, and fine activated alumina was prepared from Passargad Shimi-e-Novin co. The raw material of the activated alumina is aluminum hydroxide, which the hydroxyl on its surface will be removed and small pores will be created. Specific area surface of activated alumina was more than 200 m^2/g. the operational parameters were: pH = (5, 7, 9); initial fluoride concentration = (1, 2, 4 mg/L); concentration of single-wall and multi-wall carbon nanotubes added to water = (0.25 – 0.5 g/L); contact time = (5, 15, 30, 60, 70 min).

Synthetic water samples containing fluoride with specific were prepared and were exposed different conditions (pH, contact time, and concentration of fluoride) with a certain amount of single-wall and multi-wall carbon nanotubes. The contact was provided in a shaker, Hidolph Unimax 1010 with 470 rpm. Solution containing the absorbent was passed from filtering paper and then was centrifugal with 5000 rpm for 10 minutes. Final concentration of fluoride was optional with Spectrophotometer of Hach Co. Model D.R-5000 the optimal conditions of fluoride removal by fine activated alumina and activated alumina with 150 mesh were compared.

Fig. 1. Fluoride removal efficiency compared to the concentration of 0.25g/L of absorbent (multi-wall carbon nanotube) at pH=5, 7, 9 and fluoride initial concentration of 4 mg/L

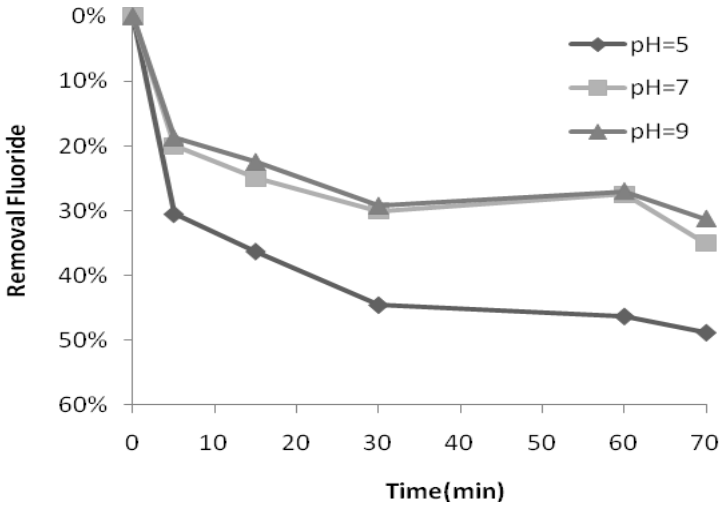

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Fig. 2. Fluoride removal efficiency compared to the concentration of 0.5g/L of absorbent (multi-wall carbon nanotube) at pH=5, 7, 9 and fluoride initial concentration of 4 mg/L

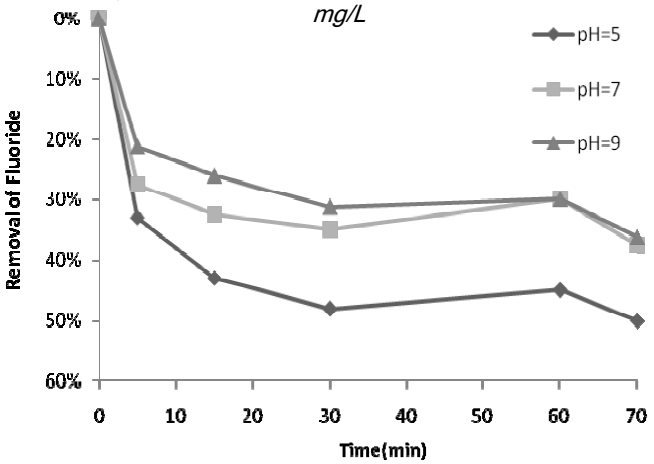


Fig. 3. fluoride removal efficiency compared to the concentration of 0.25g/L of absorbent (single-wall carbon nanotube) at pH=5, 7, 9 and fluoride initial concentration of 4 mg/L

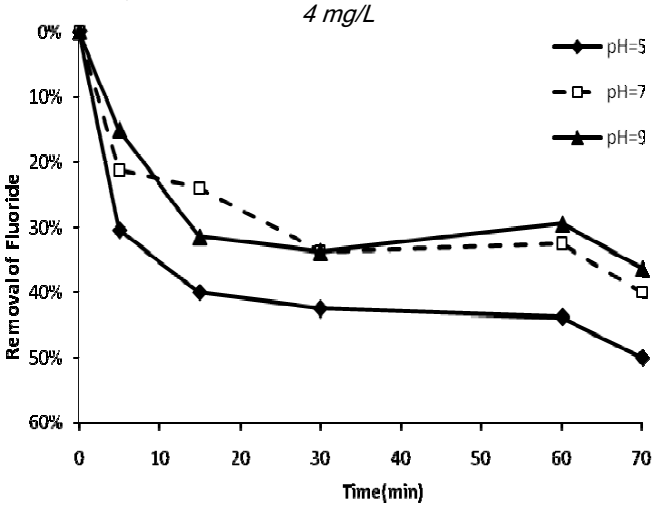


Fig. 4. Fluoride removal efficiency compared to the concentration of 0.5g/L of absorbent (single-wall carbon nanotube) at pH = 5, 7, 9 and fluoride initial concentration of 4 mg/L

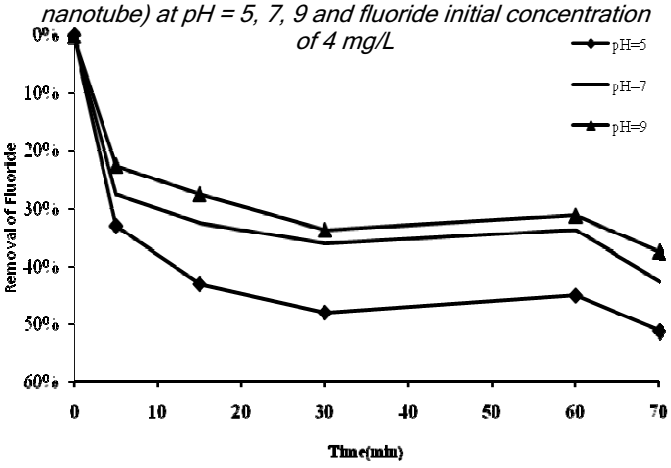
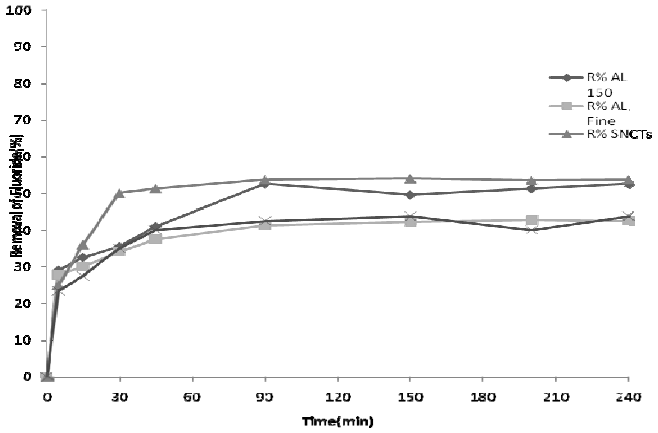


Fig. 5. Comparing reduction of fluoride concentration by fine activated alumina and 150 mesh activated alumina with single-wall and multi-wall carbon nanotubes (absorber concentration is 0.375mg/L, fluoride initial conc. = 4 mg/L and pH=6.15)



Results and discussion

Fluoride removal efficiencies for the concentration of 0.25 g/L and 0.5 g/L of absorbent (single-wall carbon nanotubes) in pH = 5, 7, 9 with initial concentration of 4 mg/L of fluoride (Fig. 1 & 2). Fluoride removal efficiencies for the concentration of 0.25 g/L and 0.5 g/L of absorbent (multi-wall carbon nanotubes) in pH = 5, 7, 9 with initial concentration of 4 mg/L of fluoride (Fig. 3 & 4). In Fig. 5, levels of fluoride decrease with fine activated alumina and 150 mesh activated alumina with single-wall and multi-wall (concentration of absorbents = 0.375 mg/L and fluoride = 4 mg/L) in pH = 6.15 are compared. In this study, fine activated alumina was compared with 150 mesh-activated alumina for fluoride removal with 4 types of absorbents (Fig. 1).

This study showed that the highest removal efficiency was obtained of fluoride in pH = 5 and fluoride initial concentration of 1 mg/L using single-wall carbon nanotube with 0.5 g/L concentration, as 58% and occurred in 70 minutes. In addition, the highest removal efficiency with multi-wall carbon nanotube is 54% and occurs in 70 minutes. As it was mentioned the highest removal level with both types of carbon nanotubes, was in pH = 5, The solution is more acidic, adsorption capacity is higher. In the first 30 minutes is high. Then slowly absorption and desorption happens and it is maximum level in 90 minutes, which reaches the higher absorption level in compare with two other adsorbents.

In a report, the adsorption of fluoride from water by aligned carbon nanotubes was studied. The results showed that uptake of fluoride in the first 30 minutes is fast and adsorption capacity reaches quickly to 3mg/g. Then, gradually in 180 minutes this will appears to moderate. The study showed that fluoride adsorption depends on pH of the solution and the maximum

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adsorption occurs at pH=7. In the present study the best removal efficiency was at pH=5 (Xiong, 2007; Bahari *et al.*, 2010).

Previous work also led to understand that reinforced carbon nanotubes with alumina, bar alumina and pH factors were used for fluoride removal; the highest adsorption level occurred in 30% weight of alumina load. The adsorption ability of reinforced carbon nanotubes is very high compared alumina. Results showed that removal efficiency reduced by pH increase. Optimum pH was 5-7 similar to the present study. However, in higher range of pH> 7-9, single-wall or multi-wall carbon nanotubes has shown no appreciable difference in removal efficiency. As per the Water Quality Association, the best performance of activated alumina is at pH = 5.5-6.5 (Tor, 2007; Bahari *et al.,* 2010).

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