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## STUDY THE EFFICIENCY OF ALUM COAGULANT IN FLUORIDE REMOVAL FROM DRINKING WATER

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### Abstract

Fluoride and its compound widely utilize in industries which disseminate in the environment through wastewater. Present study was conducted with the aim of studying the yield of alum coagulant in fluoride removal from drinking water. Present study was conducted as experimental in laboratory scale. Experiments were conducted based on variables such as fluoride concentration (3, 5, 7 and 10 mg/l), alum concentration (50, 100, 150, 200, 250, 300 and 900 mg/l), contact time (15, 30 and 45 min) and pH (6, 7, 8 and 9). Fluoride concentration measured by spectrophotometer and data analyzed using Excel software. Increasing alum coagulant concentration and contact time, fluoride removal efficiency increased, so that maximum removal rate achieved in 300 mg/l alum concentration, 45 min contact time and pH value of 6. Also, removal efficiency decreased by increasing fluoride content, since maximum removal efficiency obtained at 3 mg/l fluoride concentration at 900 mg/l alum concentration. Fluorine removal efficiency increased by increasing contact time and alum concentration and decreased by increasing fluoride initial concentration and pH value. Results showed that aluminum sulfate coagulant having acceptable efficiency, easy operation and access could be efficient way for fluoride removal from aqueous environments.

**Keywords:** fluorine, aluminum sulfate, drinking water.

**Introduction:** Recently, industrial activities like aluminum and steel production, metal's processing, fertilizer productions and so on resulted to entering considerable amounts of fluorine into environment (1). Fluorine considered

as a micronutrient in water sources which causes enameling tooth and bones, especially in children (2). However, excessive increase in fluoride concentration causes fluorosis event in which tooth and bones damaged according fluoride concentration. Also, long contact with  $F^-$  causes cancer, nervous distortions, osteoarthritis sclerosis, reduced growth and reduced IQ (3, 4). This event is intensified in warm areas in which water consumption amount is high and fluoride concentration increases due to evaporation (5). World Health Organization (WHO) determined maximum fluoride concentration in drinking water about 1.5 mg/l and optimum level recommended as 0.7-1.2 mg/l (6). Presence of high fluoride concentrations in drinking water sources in countries like India, China, Africa, Iran and United States of America was reported. So, removing excessive fluoride amounts from drinking water was considered as an important issue due to health aspects (7, 8). There are various methods for fluoride removal including reverse osmosis (2), ionic exchange (9), adsorption (10), nano-filtration (11) and electrolysis (12). Application of chemical coagulation using coagulants is one of the most common ways of wastewater treatment. The aim of coagulation is to coagulate suspended materials and especially semi-dissolved and colloid forms into large particles using chemicals which precipitate by their weight. In other words, coagulation accelerates the precipitation phase in mechanical treatment (13). Alum is one of the primary chemicals which is studied for fluoride removal from drinking water sources, so that currently utilize alone and combined with other chemicals for fluoride removal (14). Therefore, present study was conducted with the aim of fluoride removal from drinking water using aluminum sulfate coagulant.

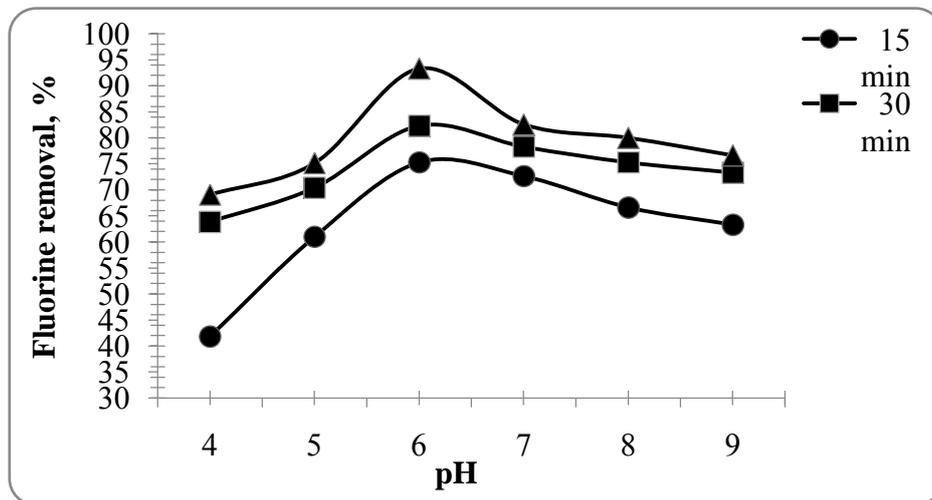
## **Materials and Methods**

The study was an experimental test in which aluminum sulfate was utilized for excessive fluoride removal from drinking water as a coagulant. In order to prepare a 100 mg/l fluoride stock solution, 221 mg sodium fluoride was dissolved in 1000 ml distilled water. Studied water samples were provided from Kermanshah urban water distribution system. The effect of various amounts of pH values (4, 5, 6, 7, 8 and 9), contact time (15, 30 and 45 min), coagulant concentration (50, 100, 150, 200, 250 and 300 mg/l) and fluoride concentration (3, 5, 7 and 10 mg/l) on aluminum sulfate coagulant efficiency were studied. It should be noted that all chemicals were purchased from Merck, Germany. In the first step, 3 mg/l fluoride and 300 mg/l aluminum coagulant were added to each sample. Then samples were poured into instrument flask (Hach) and rapid mixing was conducted at 80 rpm for one minute and calm mixing was conducted at 40 rpm for 15 minutes. After calm mixing, samples were kept for 30 minutes in order to allow precipitation and then a 256 ml sample was passed through filter paper. Fluoride was measured by spectrophotometer (Varian, UV-120-02) at 570 nm by SPANDS method and pH was measured by pH-meter (Microprocessor 537) according to standard methods (15). In the next step, in order to

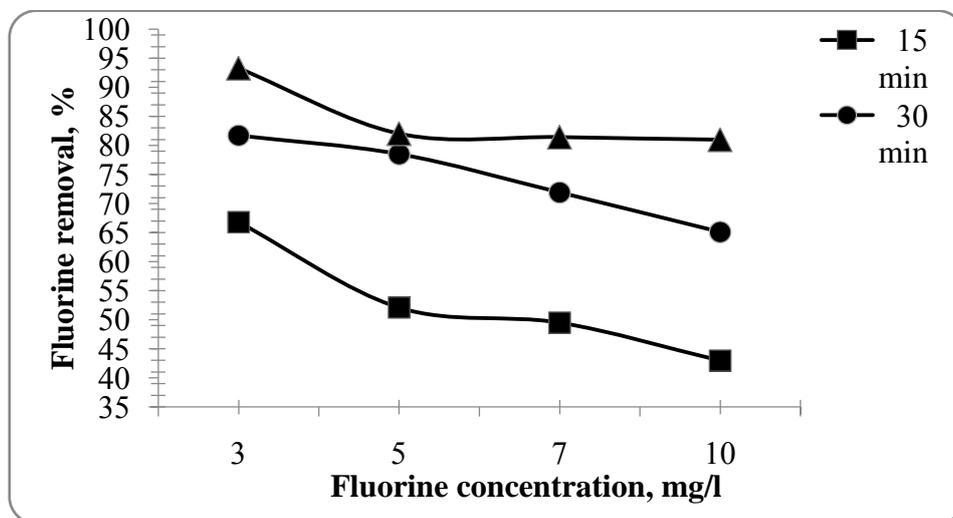
determination of optimum coagulant concentration, different aluminum sulfate concentrations added to solution having 3 mg/l fluorine and pH of 6 and results reported after 45 min. In the next step, various concentrations of fluorine (3, 5, 7 and 10 mg/l) and aluminum sulfate (300 and 900 mg/l) separately measured at pH value of 6 and 45 min contact time for determination of optimum concentration of coagulant.

**Results**

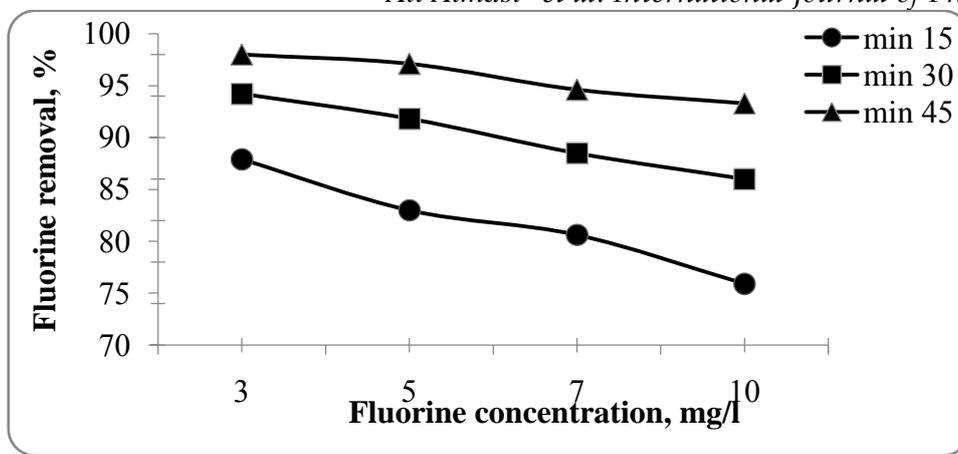
Results of the study summarized in Figure 1 to 4. Results showed that maximum removal rate achieved in 33 mg/l alum concentration, contact time of 45 min and pH value of 6. Also by increasing fluorine concentration, removal efficiency decreased so that maximum removal efficiency at 3 mg/l fluorine, obtained at alum concentration of 900 mg/l. in the present study, effect of different pH values on fluorine removal from water investigated. According to results, maximum removal efficiency (at constant alum concentrations) obtained at pH value of 6 and 45 min contact time (93.3%) indicating maximum fluorine removal at this pH.( Figure 1 to 4).



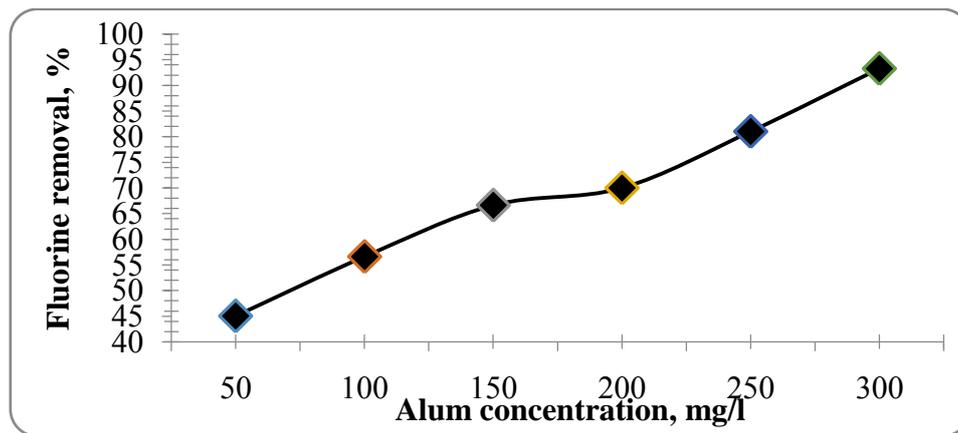
**Figure-1: effect of pH on fluorine removal efficiency at different contact time (initial fluorine and aluminum sulfate concentration of 3 and 300 mg/l, respectively).**



**Figure-2: effect of various concentrations of fluorine on fluorine removal efficiency at various contact times (pH: 6 and aluminum sulfate: 300 mg/l).**



**Figure-3: effect of various concentrations of fluorine on fluorine removal efficiency at various contact times (pH: 6 and aluminum sulfate: 900 mg/l).**



**Figure-4: Effect of alum coagulant on fluorine removal efficiency (initial fluorine concentration: 3 mg/l, pH: 6 and contact time: 45 min).**

## Discussion

According to results of the investigating fluorine concentration on removal efficiency, it can be concluded that results are in agreement with Govush et al. Govush et al. showed that increasing fluoride concentration from 4 to 10 mg/l decreased system efficiency from 90% to 83.8 percent (16). In fact, aluminum sulfate cations caused to neutralizing fluoride particles charge as iso-electric point in coagulation process. By increasing fluoride concentration, available cations would be decreases and fluoride surplus precipitated into solution (16). Also, residual fluoride at contact times less than 45 min and amounts higher than 5 mg/l are more than range recommended by WHO (6). pH of the solution is one of the important parameters in coagulation process in water treatment. Using optimum pH of coagulant, maximum removal would be acquired. On the other hand, if chemical coagulant utilized in optimum pH, coagulation process develops in earliest time (17). Maximum removal rate (at constant aluminum sulfate concentration) achieved at pH 6 and 45 min contact time (93.3 percent). Kushwaha showed that use of coagulation process with aluminum sulfate, at pH of 8, has maximum removal efficiency (18). It can be stated that increasing chemical reaction pH range of at 4 to 7 and higher causes increase in OH concentration and more production of

aluminum hydroxide. In other word, increasing the pH resulted to considerable increase in aluminum hydroxide production, fluoride removal and more fluoride precipitation in the system. Results are in agreement with findings of Sakar et al (19). In the present study, best efficiency achieved at pH 6. Isadeki et al. (20) found pH value of 7 as best pH value for fluoride removal. Kan et al. confirmed that best conditions for alum coagulation were in pH range of 6 to 7 (21). On the other hand, at pH of 5.5 to 6 it is possible that sorption coagulation or reactive coagulation occur. However, in the present study there was no considerable difference in efficiency at various pH amounts from 6 to 9. Changes in pH are an indicator of formation of aluminum hydroxide plaques. Increase in efficiency coincide with increase in contact time could be attributed to this point that by increasing contact time, size and growth of aluminum hydroxide plaques would increase. Also, production of higher amounts of precipitates and aluminum hydroxides causes increase in system efficiency. By increasing contact time, micro flocks would be grater and heavier and would remove through sweep coagulation (13). Therefore, more removal efficiency requires more reaction time, so removal efficiency after 45 min reached to maximal amount. Results are in agreement with Dargahi results (22). Coagulant dose is one of the important parameters for determination of optimal performance in coagulation and flocculation. Determination of optimum dose for minimizing sludge production, decreasing cost and achieving favorable yield is important (19). Present study shows that fluorine removal efficiency increased as coagulant dose increased. Maximum removal efficiency obtained at 300 mg/l concentration. As aluminum sulfate concentration increased, particles precipitation rate increased which is effective in decreasing reaction time (23). Also, increasing fluorine removal by increase in coagulant dose was due to increase in hydrolyzed species which destabilizes the colloidal particles. Results of Kushuwaha et al. confirm our findings. Kushuwaha et al. showed that increasing coagulant dose from 100 to 300 mg/l, increased COD removal efficiency in dairy wastewater. Present study showed that if favorable conditions provided for coagulation process, fluoride contents could be reduced to less than standard limit.

## **Conclusion**

Overall results showed that fluorine removal from drinking water is possible with alum coagulant. Considering to needing high concentrations of aluminum sulfate for fluorine removal, consequently high amount of aluminum remained in the water. It is recommended that absorbents like activated alumina utilized for removing residual aluminum after fluorine removal treatment. Also, future studies recommended for providing and economic and appropriate fluorine removal, measuring residual aluminum and sludge production rate and most appropriate dehydration of the sludge.

## References

1. Tang Y, Guan X, Wang J, Gao N, McPhail MR, Chusuei CC. Fluoride adsorption onto granular ferric hydroxide: effects of ionic strength, pH, surface loading, and major co-existing anions. *Journal of Hazardous Materials*. 2009;171(1):774-9.
2. Maheshwari R. Fluoride in drinking water and its removal. *Journal of Hazardous Materials*. 2006;137(1):456-63.
3. Jagtap S, Thakre D, Wanjari S, Kamble S, Labhsetwar N, Rayalu S. New modified chitosan-based adsorbent for defluoridation of water. *Journal of Colloid and Interface Science*. 2009;332(2):280-90.
4. Tchomgui-Kamga E, Ngameni E, Darchen A. Evaluation of removal efficiency of fluoride from aqueous solution using new charcoals that contain calcium compounds. *Journal of colloid and interface science*. 2010;346(2):494-9.
5. Tor A, Danaoglu N, Arslan G, Cengeloglu Y. Removal of fluoride from water by using granular red mud: batch and column studies. *Journal of hazardous materials*. 2009;164(1):271-8.
6. Organization WH. *Guidelines for drinking-water quality: recommendations*: World Health Organization; 2004.
7. Asgari G, Roshani B, Ghanizadeh G. The investigation of kinetic and isotherm of fluoride adsorption onto functionalize pumice stone. *Journal of hazardous materials*. 2012;217:123-32.
8. Gao X, Zhang F, Wang C, Wang Y. Coexistence of high fluoride fresh and saline groundwaters in the Yuncheng Basin, northern China. *Proc Earth Planet Sci*. 2013;7:280-3.
9. Ho LN, Ishihara T, Ueshima S, Nishiguchi H, Takita Y. Removal of fluoride from water through ion exchange by mesoporous Ti oxohydroxide. *Journal of colloid and interface science*. 2004;272(2):399-403.
10. Chen N, Zhang Z, Feng C, Zhu D, Yang Y, Sugiura N. Preparation and characterization of porous granular ceramic containing dispersed aluminum and iron oxides as adsorbents for fluoride removal from aqueous solution. *Journal of hazardous materials*. 2011;186(1):863-8.
11. Hu K, Dickson JM. Nanofiltration membrane performance on fluoride removal from water. *Journal of Membrane Science*. 2006;279(1):529-38.
12. Amor Z, Bariou B, Mameri N, Taky M, Nicolas S, Elmidaoui A. Fluoride removal from brackish water by electrodialysis. *Desalination*. 2001;133(3):215-23.
13. Pirsahab M, Zinatizadeh AA, Dargahi A. Performance Evaluation of Coagulation Process in Removal of Low Turbidity and Color from Water Using Different Inorganic Coagulants. *water and wastewater journal*. 2012 (1):111-8.

14. Aguilar M, Sáez J, Lloréns M, Soler A, Ortuno J, Meseguer V, et al. Improvement of coagulation–flocculation process using anionic polyacrylamide as coagulant aid. *Chemosphere*. 2005;58(1):47-56.
15. Carranzo IV, editor APHA, AWWA, WEF." Standard Methods for examination of water and wastewater." *Anales de Hidrología Médica*; 2012.
16. Ghosh D, Medhi C, Purkait M. Techno-economic analysis for the electrocoagulation of fluoride-contaminated drinking water. *Toxicological & Environ Chemistry*. 2011;93(3):424-37.
17. Chu W. Dye removal from textile dye wastewater using recycled alum sludge. *Water Research*. 2001;35(13):3147-52.
18. Kushwaha JP, Srivastava VC, Mall ID. Treatment of dairy wastewater by inorganic coagulants: Parametric and disposal studies. *Water research*. 2010;44(20):5867-74.
19. Sarkar B, Chakrabarti P, Vijaykumar A, Kale V. Wastewater treatment in dairy industries—possibility of reuse. *Desalination*. 2006;195(1):141-52.
20. Essadki AH, Gourich B, Vial C, Delmas H, Bennajah M. Defluoridation of drinking water by electrocoagulation/electroflotation in a stirred tank reactor with a comparative performance to an external-loop airlift reactor. *Journal of hazardous materials*. 2009;168(2):1325-33.
21. Chichuan K, Chihpin H. Coagulation monitoring in surface water treatment facilities. *Water science and technology*. 1998;38(3):237-44.
22. Dargahi A , Taghi Savadpour M, Sharafi K. The study of coagulation process in medium turbidity removal drinking water. *Arch Hyg Sci*. 2014;3(4):192-200.
23. Sharafi K, Mansouri AM, Zinatizadeh AA, Pirsahab M. Adsorptive removal of methylene blue from aqueous solutions by pumice powder: process modelling and kinetic evaluation. *Environmental Engineering & Management Journal (EEMJ)*. 2015;14(5): 1067-1078.
24. Pirsahab M, Karami A, Dargahi A, Ejraei A. Evaluation the performance of inorganic coagulants (poly aluminium chloride, ferrous sulfate, ferric chloride and aluminum sulfate) in removing the turbidity from aqueous solution. *International Journal Of Pharmacy & Technology*. 2016;8(2):13168-81.

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