

# Development of Cost Effective Method for the Removal of Fluoride in Drinking Water Samples

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**Abstract:** Exceeding the level of fluoride (F<sup>-</sup>) in drinking water is responsible for skeletal fluorosis in human beings. Several methods were already been established, and in this works the authors moto is to find out the cost effective method for the fluoride removal in water. To evaluate this various concentrations of fluoride solutions were prepared and were subjected to various methods for the best removal. In process of that the authors selected some cost effective methods and their reliability to the cost was compared. The present study was carried out to assess the ability of electrocoagulation process with iron and aluminum electrodes in order to removal of fluoride from aqueous solutions. Several working parameters, such as concentration of fluoride, pH, applied voltage and reaction time, distance between electrodes, electrode reactive area and effect of Co- existing ions were studied to achieve a higher removal capacity. Variable concentrations (1, 5 and 10 mg/l) of fluoride solutions were prepared by mixing proper amount of sodium fluoride with tap water. The varying pH of the initial solution (5, 7 and 9) was also studied to measure their effects on the fluoride removal efficiency. Results obtained with synthetic solution revealed that the most effective removal capacities of fluoride could be achieved at 40V electrical potential. In addition, the increase of electrical potential, in the range of 10-40 V, enhanced the treatment rate. The effective reactive surface area found to be 40cm<sup>2</sup> whereas inter electrode distance was 1cm. Aluminum electrode could works effectively during defluoridation over iron electrode. This method could be effective up to a fluoride concentration level of 8ppm. Optimal results could be obtained at pH: 7. With an increase in electrode surface area the extent of defluoridation would be higher. The ability of Industrial grade alum to remove Fluoride from aqueous solution has also been investigated. The studies were carried out as functions of contact time, concentration and pH. The data indicate that Industrial grade alum surface sites are heterogeneous in nature and that fits into a heterogeneous site binding model. The optimum pH for complete removal of Fluoride from aqueous solution was found to be 6. The rate of fluoride removal was rapid during the initial 20 minutes, and equilibrium was attained within 50 minutes. The defluoridation process using alum could be optimal at a fluoride concentration of 2.5 ppm. Along with that the ability of Industrial grade aluminum oxide (Alumina) to remove Fluoride from aqueous solution has also been investigated. The studies were carried out as functions of contact time, concentration and pH. The data indicate that alumina surface sites are heterogeneous in nature and that fits into a heterogeneous site binding model. The optimum pH for complete removal of Fluoride from aqueous solution was found to be 8. The rate of fluoride removal was rapid during the initial 20 minutes. The defluoridation process using alumina could be optimal at a fluoride concentration of 8.0 ppm. With an increase in alumina dosage the extent of defluoridation could be optimal up to 8 gm. In addition to that the ability of activated carbon to remove Fluoride from aqueous solution has also been investigated at various concentrations and with different periods of time. The optimum pH for complete removal of Fluoride from aqueous solution was found to be 7. The rate of fluoride removal was rapid during the initial 40 minutes. The defluoridation process using alumina could be optimal at a fluoride concentration of 2.0 ppm. With an increase in activated carbon dosage the extent of defluoridation could be optimal up to 4 gm.

**Index Terms:** Defluoridation, electro coagulation, alumina, industrial grade alum, activated carbon, cost comparison

## I. INTRODUCTION

Fluorine belongs to the halogen group in the periodic table, known to be the lightest element in the group and the most reactive than all the other chemical elements. It is the most electronegative of the halogen group and has a strong tendency to transform into fluoride ion, which means that it exists as fluoride ion in aqueous solutions. This is the most available state of fluorine observed in nature than any other forms. Since the charge and the radius of hydroxide ions were almost similar to those of fluoride, they were capable of replacing the fluoride ions in the mineral structure of water. In addition hydroxide ions have the capability to form various mineral complexes with several cations and with various fluorides containing mineral species of low solubility. In the earth's crust, fluorine was found as its fluorides at about 0.06- 0.09 percent and the crustal abundance was found to be 300 mg/kg. It was established that fluorides were found in various types of minerals including hornblende, apatite, rock phosphate, cryolite, mica and in fluorspar at significant levels. In igneous and sedimentary rocks calcium fluoride was the common fluoride mineral which was found to have low solubility. The fumarolic acid and volcanic eruptions are generally associated with fluorine. Thermal water with higher pH value possess, higher amounts of fluorine. Cryolite and rock phosphates are the minerals of commercial importance, where in cryolite is used to produce aluminum. Phosphate fertilizers are prepared by converting rock phosphates by removing fluoride percentage up to 4.2 %. This removed fluoride was purified as its fluorosilicates and the purified fluorosilicates are added to drinking water as a protection against dental cavities in various African countries.

Fluoride exists in certain concentrations in all natural waters. The fluoride concentration in sea water was found to be 1mg/ l, where as in several lakes and rivers the concentrations of fluoride were found to be less than 0.5 mg/l. The concentration of fluoride

in groundwater is mainly due to the chemical, geological, physical characteristics of the aquifer, the acidity and porosity of the soil and rock lattice, temperature, and the chemical action of several elements and depth of the aquifer. On account of the large number of differences in soil, temperature and physical properties, the fluoride concentrations in groundwater vary from country to country. The concentrations of fluoride varies from less than 1 mg/l to more than 35 mg/l, in different countries like Kenya, South Africa and India. The concentrations of fluoride in water depends on fluoride solubility. So in groundwater high fluoride concentrations are expected where poor concentrations of calcium aquifers are present. The ion exchange process also increases the fluoride concentration in groundwater where the sodium is replaced by calcium. Continuous consumption of more or equal to 1 mg/l or higher concentrations of fluoride causes dental fluorosis. Extreme concentrations of fluoride lead to skeletal fluorosis. The Rift Valley system and the soda lakes were found to have extreme fluoride content of about 1,640 mg/l and 2,800 mg/l respectively. Elmentaita and Nakuru are the Kenyan lakes consisting of about 690 mg/l fluoride concentration. Nair. et al (1984) made a detailed survey of fluoride content in the groundwater of Kenya by collecting over 1000 groundwater samples throughout the nation. Over 61% of the samples exceeded the concentration of 1 mg/l, about 20% of the samples were found to have a concentration over 5 mg/l and about 12% of the samples were found to have 8 mg/l concentration. The higher concentrations of fluoride was found in volcanic areas of the Rift Valley and Central Provinces of Nairobi, where the groundwater fluoride concentrations were 30-50 mg/l. Wells and boreholes are known to be the common sources of drinking water. Since water fluoridation causes serious health problems, one cannot justify the costs. The adverse effect found in children's teeth during teeth development has been identified as dental fluorosis. Dental fluorosis is observed to be very high in fluoride affected regions of the world. In United Republic of Tanzania the fluoride concentration of drinking water exceeded 1.5 mg/l in 30% of the areas (Latham and Grech, 1967). High fluoride concentrations are associated with different rock soils, for instance various metamorphic and igneous rocks like gneisses and granites have high concentrations of fluorine in countries like India, West Africa, Pakistan, Sri Lanka, China, Southern Africa and in Thailand. In China 28 provinces are found to have fluoride in groundwater, But Shanghai is an exception to this. Here the concentration of fluoride is higher in both shallow and deeper layers. Dissanayake et al. (1991) observed that the dry zone was found to have a fluoride concentration up to 10 mg/l, leading to dental and skeletal fluorosis. Endemic fluorosis has become a national health problem in India, where 13 states contain high concentrations of fluoride in groundwater as reported by Mangla et al. (1991). According to UNICEF the fluoride, affected states in India has increased to 17, in which Andhra Pradesh, Telangana, Punjab, Haryana, Rajasthan, Gujarat, Tamil Nadu and Uttar Pradesh were found to have high fluoride concentrations in groundwater. According to UNICEF, Rewari District of Haryana has extreme concentration of 48 mg/l of fluoride in groundwater owing to distribution and the extent of dissolution of fluorite, topaz and apatite in the local bedrock.

## II. LITERATURE REVIEW

Fluoride removal from water has been a challenge to scientists for over a decade. Several researchers have proposed some efficient and cost effective methodologies for the removal of fluoride from drinking water. The following is the account of the methodologies proposed by them over a period of time. Sujana. M. G. et al (2011), dwelt on removal of fluoride from contaminated water using bauxite. The effectiveness of his methodology has been established on both natural and synthetic groundwater samples. The impact of several ions over fluoride removal was also determined by them. They reported that competition of the surface sites of  $\text{CO}_3^{2-}$  ions was less when compared to those of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{PO}_4^{3-}$  at a fluoride concentration of 5-20 ppm levels of fluoride. Zhijie Zhang et al (2011), proposed the method of removal of fluoride from water using calcium chloride modified natural Zeolite. The experiments were carried out over various batch sizes. Several parameters like pH, co-ions, time and dose were also examined. The maximum absorption was observed to be 1.766 mg g<sup>-1</sup>, at initial fluoride concentration of 100 mg L<sup>-1</sup>. X. Fan et al (2003), reported that the fluoride uptake by the ion exchange resin Hydroxyapatite has higher fluoride adsorption compared to quartz, calcite and fluorspar. The initial concentration ranges from  $2.5 \times 10^{-5}$  to  $6.34 \times 10^{-2}$  mg/l. Approximately 90% of the fluoride was removed using this methodology at pH of 6. Krishna Biswas et al (2010), developed a mixed oxide of iron(III)-aluminum(III)-chromium(III) ternary hydrated oxide, and the applicability and effectiveness of this oxide was investigated. The conditions looked highly pH sensitive, the optimum pH level was 4.0-7.0 and equilibrium was obtained at about 1.5 h. Sanjay P. Kamble et al (2010), proposed fluoride removal from drinking water using alumina from alkoxide origin. The effectiveness of its applicability was established over several batch sizes and for continuous operations. In addition, the impact of several parameters like pH, dose and fluoride concentration that help fluoride removal was also well established. This method can be effectively used for commercial sources of water as well. Sneha Jagtap et al (2011), dealt with the applicability of lanthanum impregnated chitosan flakes, and their usage for the removal of fluoride from water. The effective removal of fluoride using chitosan was well established whereas a modified impregnation of lanthanum further enhanced the effectiveness of fluoride removal. In addition, the impact of several parameters over fluoride removal was also studied. Mohammad M. Emamjomeh et al (2011), used electrocoagulation process and its analysis during fluoride removal. Defluoridation was found to be most effective at pH levels ranging between 6-8. The removal was examined by XRD analysis. Youbao Sun et al (2011), reported fluoride removal from water using (III) modified natural stilbite zeolite, where Stilbite zeolite is a high crystalline open pore natural zeolite. By adding  $\text{FeCl}_3$  solution the effectiveness of the process removal of fluoride was enhanced. The modification was analyzed by using FTIR. Various involved parameters were also examined. At pH 6.9 the level of fluoride in water was reduced to less than 1 ppm from 10 ppm, with 2.31 mg/g adsorption capacity. S. Chakraborty et al (2013), described the effective fluoride removal from impacted groundwater using cross flow nanofiltration, which was considered to be an economical model for fluoride removal. The system was found to be most accurate and the errors during its performance was found to be fewer than 0.1. More than 98% of fluoride from water was successfully removed. This is considered to be most effective for the industrial usage. Kamel Babaeiveli et al (2013), described the effective fluoride removal using adsorption over crystalline titanium dioxide. The impact of various parameters like pH, concentration, dosage and other ions was also studied. Maximum removal was observed in the pH range of 2. At about pH range 7-8, about 75% of removal was observed. Higher the concentration of carbonate ion, lower was the percentage of fluoride removal.

Yanhui Li et al (2013), reported the usage of combination of manganese dioxide and graphene oxide as a defluorinating agent for water. Where the graphene oxide coated with manganese dioxide was synthesized and characterized with XRD and FTIR. The optimum removal was reported to be at pH of 5.5-6.7. Various conditions and time and dosage were also described. Yuxin Maa et al (2017), reported effective fluoride removal from aqueous medium using acid treated bentonite. The applicability of this method was made for column and batch sizes as well. A more effective removal was reported at pH of 4.95 and about 40 mins of time was taken for the equilibrium. Vinod Kumar Gupta et al (2007), described effective fluoride removal from water using waste carbon slurry obtained from fertilizer companies. The best pH conditions for this method was noted as 7.58 and the equilibrium was obtained at 1h. Maximum adsorption capacity of fluoride was reported as 4.861 mg/g. It was repeated when the experiments were carried out over column and batch size. It was reported to be the best in cost and performance for the effective removal of fluoride. Eva Kumar et al (2009), reported that granular ferric hydroxide had the capability of removing fluoride from water. The batch experiments started with an initial concentration of 1-100 mg/L and the contact time was 1–24 h. About 95% of adsorption took place within 10 mins and the equilibrium was obtained at 60 min. The ideal pH condition was prescribed as 4-8. Dominique Claveau-Mallet et al (2013), proposed the removal of fluoride, metal and phosphorous with steel slag filters. The experiments were done on batch size starting with water having 1.7-8.2 mg/L aluminum 11-107 mg/L orthophosphates, 0.24-0.83 mg/L manganese, 9-37 mg/L fluoride and 0.20-3.3 mg/L zinc. Experiments were conducted over a period of 145-222 days and the samples were collected twice a week. Within 34 h 85.3% of fluoride, 99.9% of phosphorus, 99.3% of zinc and 98.0% of manganese were removed. Liyuan Chai et al (2013), reported fluoride removal from drinking water with nanoparticles of sulfate doped Fe<sub>3</sub>O<sub>4</sub>/Al<sub>2</sub>O<sub>3</sub>. About 90% of removal attained in a time lag of 10 mins. At an initial concentration of 10 mg/L of fluoride removal of 90% and at 50 mg/L, about 70% of fluoride removal were reported. Several parameters involved in the process were also mentioned at length. Hari Paudyal et al (2013), worked on drinking water defluorination using dried orange juice residue at minimal concentrations of fluoride, after a chemical loading of Zr(IV), Ce(IV) and Al(III), over the orange juice pulp of about 0.90, 0.85 and 0.97 mmol/g respectively. This method was effective only at a very low levels of fluoride. Xiaolin Yu et al (2013), describe a novel process for the removal of fluoride using cellulose hydroxyapatite nanocomposited from water samples. By the application of this methodology the drinking water meets the WHO norms as the initial fluoride concentration of 10mg/L has been reduced to 3g/L. The effect of coexisting ions were also described. Hongtao Wang et al (2007), developed defluorination of drinking water mechanism using Mg/Al hydrotalcite and calcinated compounds. The main factors described were pH and the crystal structure of the calcinate. Within 15 mins the maximum adsorption was attained. The initial fluoride concentration was 5 mg/L and by the application of this methodology the fluoride concentration was reduced to 1 mg/L. Guocheng Lv et al (2012), suggested the use of red mud sintered porous materials, and their applicability as an effective defluorination agent. Zirconium coated red mud pots were used for the defluorination with a pore size of 50-100µm. Maximum adsorption of fluoride was reported at pH .3.0 and the equilibrium was obtained within 60mins.

### III. METHODOLOGY

Experimental Methodology for fluoride removal from water using electrocoagulation. All the chemicals used in this research work are of analytical grade. Variable concentrations (1-10 mg L<sup>-1</sup>) of fluoride solutions were prepared by mixing proper amount of sodium fluoride with de-mineral water. The pH of initial solution was adjusted (5, 7 and 9) by using sulfuric acid solution (1N) and sodium hydroxide (1N). Experiments were performed in a batch reactor. The internal size of the cell was 5 cm × 10 cm (width × length) (Fig. 4.1, and 4.3) with an effective volume of 250ml. The active area of each electrode was 4× 5 cm. The distance between electrodes was 4 cm. Power supply pack had an input of 220V and variable output of 0–40V (10, 20, 30 and 40 V for this study). At different time intervals (10, 20, 30 and 40 min) 25ml of treated sample was collected and filtered before being analyzed to determine the residual fluoride. The residual fluoride concentration was determined using spectrophotometric method according to the standard method (APHA, 21st Edition).

### IV. RESULTS & DISCUSSION

The electrocoagulation process is quite complex and may be affected by several parameters like concentrations, pollutants, initial pH and electrical potential (voltage). In the present study, electrocoagulation process has been evaluated as a treatment technology for fluoride removal from synthetic solutions, fluoride removal efficiency at different conditions (pH, electrical potential and various initial concentrations) in various reaction times has been evaluated as follows.

Variable concentrations (1-10 mg/ L) of fluoride solutions were prepared by mixing proper amount of sodium fluoride with distilled water. The pH of initial solution was adjusted (5, 7 and 9) by using sulfuric acid solution (1N) and sodium hydroxide (1N). Experiments were performed in a batch reactor. The internal size of the cell was 5 cm × 10 cm (width × length) with an effective volume of 200ml. The active area of each electrode was 4×5 cm. The distance between electrodes was 4 cm. Power supply pack having an input of 220V and variable output of 0–40V (10, 20, 30 and 40 V for this study). At different time intervals (10min) 25ml of treated sample was collected and filtered before being analysed to determine the residual fluoride. The residual fluoride concentration was determined using spectrophotometric method according to the standard method (APHA, 21st Edition). The time dependence of fluoride removal by electrocoagulation process at different electrodes was shown in Table.5.1. It can be seen from the figure that up to 50-87 % (Fig.1) of the initial concentration of fluoride decreased within 10-30 min of electrocoagulation processing for both the electrodes.

The optimum time for fluoride removal was observed to be 20 min and above that contact time the fluoride would desorb from the flocs. A series of experiments were performed with different initial concentrations of fluoride. The efficiency of electrocoagulation for the removal of fluoride was also studied and results were obtained. The results obtained at different concentration levels of fluoride are given as follows. The initial concentrations of fluoride were started with 2 ppm and the studies were carried at various levels of fluoride concentration i.e 4, 6, 8 and 10 ppm (Fig.2), taken from groundwater.

The results obtained during the defluoridation of impacted water were as follows.

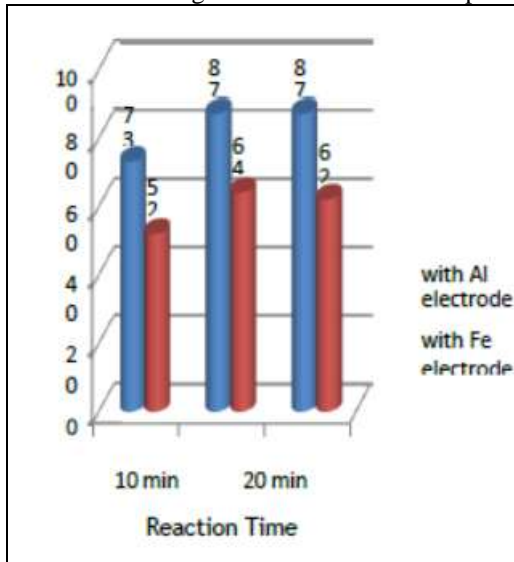


Figure 1: Effect of reaction time on fluoride removal

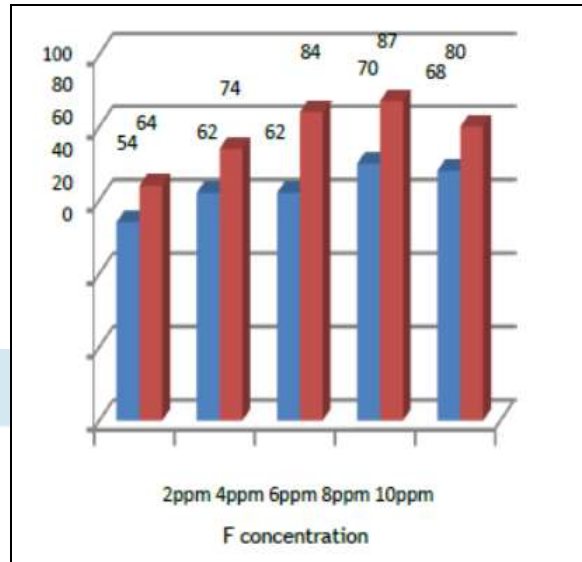
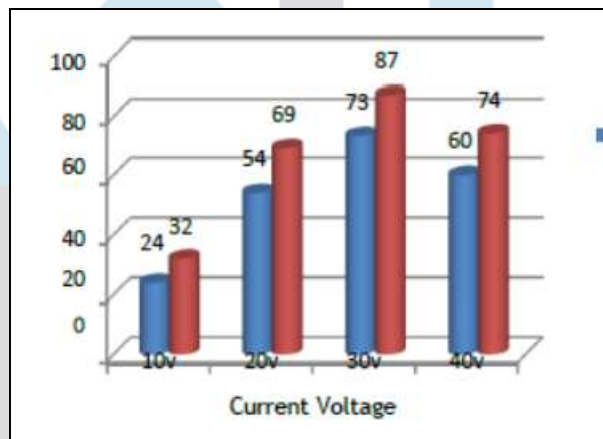


Figure 2: Effect of Con. Of defluoridation in electrocoagulation of 10 ppm

It is well-known that electrical current not only determines the coagulant dosage rate but also the bubble production rate and size and the floc growth, which can influence the treatment efficiency of the electrocoagulation. Thus current density or applied voltage (electrical potential) has an effect on the fluoride removal.

As expected, it appears that for a recorded time, the removal efficiency increased significantly with the increase in current density. This proves the fact that at a higher voltage level the amount of Al oxidation increased resulting in the formation of greater amount of precipitate which made the electrode not to react to the fluoride. The maximum fluoride removal was observed at 30V (Fig.3), while the lowest fluoride removal efficiency occurred in the lowest electrical potential (10V).



## V. CONCLUSION

All the experiments were carried out using variable fluoride dosages, and the optimal results were obtained at an initial fluoride concentration of 8 ppm. Maximum defluoridation was observed at a 20 min, that is, when the equilibrium was attained and after that no considerable defluoridation was observed. Aluminum electrode is found to be the more efficient than iron electrode. The distance between the electrodes was well impacted during the defluoridation process. Lesser the differentiation between the electrodes, the better would be the amount of defluoridation. The surface area of the electrode strongly impacts the defluoridation process. Defluoridation increases with the increase in the surface area of the electrode. The optimal pH condition for the best defluoridation was found to be 7. For an applied voltage of 30 V optimal defluoridation was observed. The co-existing ions of sulfate, nitrate and chloride highly impact the defluoridation process. The cost of the process per 1000 liters at a fluoride concentration of 8ppm and at an applied voltage of 30V with aluminum electrode, at a time equilibrium of 20 min and at a pH of 7, was calculated as 35.6 INR or 0.57 USD. All the experiments were carried out using variable fluoride dosages, and optimal results were obtained at an initial fluoride concentration of 8 ppm. Optimal defluoridation was observed at 40 min, when the equilibrium was attained and after that no considerable defluoridation was observed. The best alum dosage for optimal defluoridation was found to be 2.5 g/ liter. The optimal pH condition for the best defluoridation was found to be 7. The co-existing ions sulfate, nitrate and chloride highly impacts the defluoridation process. The cost of the process per 1000 liter at a fluoride concentration of 8ppm and at a time equilibrium of 40 min and at a pH of 7, was calculated to be 50.68 INR or 0.8 USD.

All the experiments were carried out using variable fluoride dosages, and optimal results were obtained at an initial fluoride concentration of 8 ppm. Optimal defluoridation was observed at 40 min, when the equilibrium was attained and after that no considerable defluoridation was observed. The best alum dosage for optimal defluoridation was found to be 2g/ liter and there was

no use of increasing the dosage of carbon. Optimal defluoridation was observed at an initial fluoride concentration of 2 ppm. At higher fluoride concentrations this method was not found to be much effective for defluoridation.

The optimal pH condition for defluoridation was found to be 7. The coexisting ions sulfate, nitrate and chloride highly impacts defluoridation process. The cost of the defluoridation process per 1000 liters at a fluoride concentration of 2 ppm and at a time equilibrium of 40 min and a pH of 7 was calculated to be 101.56 INR or 1.6 USD.

Using alumina All the experiments were carried out using variable fluoride dosages, and optimal results were obtained at an initial fluoride concentration of 8 ppm. Optimal defluoridation was observed at 40 min, when the equilibrium was attained and after that no considerable defluoridation was observed. The best alum dosage for optimal defluoridation was found to be 8g/ liter and the higher the dosage the highest will be defluoridation. The optimal defluoridation was observed at an initial fluoride concentration of 8 ppm. The optimal pH condition for the best defluoridation was found to be 8. The cost of the process per liter at a fluoride concentration of 8 ppm at a time equilibrium of 40 min and at a pH of 8 was calculated to be 32.64 INR or 0.52 USD.

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