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Removal of fluoride from drinking water by using electrocoagulation (EC) with Aluminium (Al) electrodes

Pramod Kharmale, Sangramsingh Thakur
Assistant Professor, Civil Engineering, GHRCOEM Ahmednagar, Maharashtra, India

Abstract
Endemic fluorosis caused by fluoride in excess is a global geochemical disease. Maintaining fluoride concentration of 1mg/L in the dietary intake can prevent skeletal and dental problems (WHO, 1.5 mg/L max. limit). Recent attention of scientists has been devoted to the study of different types of low cost methods such as Electro coagulation (EC) is an emerging water treatment technology that has been applied successfully to treat various wastewaters and also for drinking water. Effects of different operating conditions such as pH, voltage, hydraulic retention time (HRT) and number of aluminum plates between anode and cathode plates on removal efficiency are investigated. A simple setup was fabricated made up of Acrylic for Electro coagulation along with sand filter has been used for fluoride removal. The fluoride laden synthetic water is passed through a reactor (1.5 L) in which electro coagulation (current passed 25 amp. to 80 amp.) is done and then effluent was filtered through sand filter and effluent after filtered was analyzed for different pH, Voltage, fluoride concentration. Results showed that there will be significantly reduction of fluoride concentration within time period of 30 to 40 minutes as specified by Indian Standard for drinking water. The objective of the present study is to examine the feasibility of using EC in fluoride removal from drinking water.

Keywords: electrocoagulation (EC) with Aluminium (Al), hydraulic retention time (HRT)

1. Introduction
Fluoride is a ubiquitous element present in earth's crust and is also being added to the environment anthropogenically. It is the most electronegative of all elements. Fluorine is found in the soil and the content of Fluorine in the lithosphere varies between 100 and 1500 g/ton. Fluoride has gained importance due to its dual influences on human beings. An excess amount of fluoride anions in drinking water has been known to cause adverse effects on human health. To prevent these harmful consequences, especially problems resulting from fluorosis, the World Health Organization (WHO) fixed the maximum acceptable concentration of fluoride anions in drinking water to 1 to 1.5 mg/L. Several processes, including electrodyalysis (Amor *et al*., 1998; Amor *et al*., 2001), Nanofiltration (Hu *et al*. 2006; Cohen & Conrad, 1998) [4], ion exchange membrane (Singh *et al*., 1999, Castel *et al*., 2000, Chubar *et al*., 2007; Tor, 2007), chemical treatment (Huang *et al*., 1999; Hu *et al*., 2005; Meenakshi *et al*., 2006), adsorption into materials (Srimulari *et al*., 1999; Fan *et al*., 2003; Wu *et al*., 2007), alum coagulation (M.G. Sujana *et al*., 1998), reverse osmosis (RO), electro dialysis (H. Nobuyuki, *et al*.,1999) and selective ion exchange (K.M. Popat, *et al*., 1994), can remove fluoride from water. Among these, coagulation with aluminum salt is one of the best. An electrochemical technique, electro coagulation (EC), has been applied to yield aluminum ions with aluminum anodes as a coagulant for removing fluoride (L. Ming, *et al*., 1983; N. Mameri, *et al*., 1998; F. Shen, X. *et al*., 2003). Some researcher has demonstrated that EC using aluminum anodes are effective in defluorination. The combined electro coagulation and electro flotation process is successfully applied in treating water containing fluoride by (Shen *et al*., 2003). As the electric current passes through the anodes, the aluminum metal is oxidized to aluminum ions. Afterward, the aluminum ions are transformed to polymeric species or Al(OH)3 flocs, which can co-precipitate or adsorb the fluoride ions, as presented by the following reactions L. Ming, *et al*., 1983; N. Mameri, *et al*.,1998; F. Shen, X. *et al*., 2003).

nAl^{3+} (aq) + 3n – mOH- (aq) + mF^{-}(aq) → AlnFm (OH) 3n-m(s) ……. (eq.1)

Aln (OH) 3n(s) + mF-(aq) → AlnFm (OH) 3n-m(s) + m (OH)-(aq). ……..(eq.2)

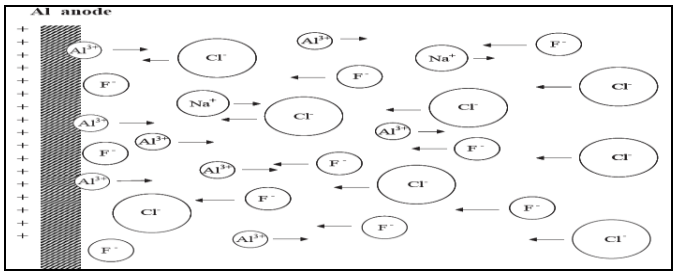

Fig 1: Diagram shows the electro-condensation effect.

2. Literature Re-view
1) ***Vaishali Tomar and Dinesh Kumar (2013)*** **[1]** found that Fluoride is a persistent and non-biodegradable pollutant that accumulates in soil, plants, wildlife and in human beings. Therefore, knowledge of its removal, using best technique with optimum efficiency is needed. The present survey highlights on efficacy of different materials for the removal of fluoride from water. The most important results of extensive

studies various key factors (pH, agitation time, initial fluoride concentration, temperature, particle size, surface area, presence and nature of counter ions and solvent dose) fluctuate fluoride removal capacity of materials are reviewed.

2) *Mine Taştaban Olcay Tünay N. Işık Kabdaşlı1 and Tuğba Ölmez-Hanc* (2013) *et al*. In this paper, experimental studies were conducted for the removal of fluoride by electrocoagulation process using aluminium electrodes and with synthetic samples. The purpose of the study was to investigate the removal potential of fluoride on the electrodes rather than onto the sludge. Fluoride removal via accumulation on the electrode is carried out in the pH range; where solubility of aluminum hydroxide near the optimum pH values. The main parameters were evaluated to find out their impact on the process and to define optimum treatment conditions. Fluoride removal was found to occur between the pH range of 4 to 5. The process efficiency increased with increasing current density up to 10 mA/cm2. The kinetic analysis of the data was employed to assess the effects of the process parameters on the rate of the process. The analysis of the material scraped off the electrodes was made to verify the mechanism of the process.

3. Materials and methods

3.1 Electrocoagulation process

Electrocoagulation (EC), the passing of electrical current through water, has proven very effective in the removal of contaminants from water. Electrocoagulation systems have been in existence for many years (Dietrich, patented 1906), using a variety of anode and cathode. This process consists of creating metallic hydroxide flocs within the water by electro dissolution of soluble anodes, usually constituted by aluminum. In the Electrocoagulation unit, two aluminum electrodes having opposite charges will undergo anodic reactions on the positive side while on the negative side cathodic reaction takes place. When current passes through an electrochemical reactor, cathodic over potential and ohmic potential drop of the solution. The anode over potential includes the activation over potential and concentration over potential, as well as the possible passive over potential resulted from the passive film at the anode surface, while the cathode over potential is principally composed of the activation over potential and concentration over potential. When aluminum is used as electrode material, three major reactions occur in electrochemical reactor.

The oxidation reaction at the anode,

$Al \rightarrow Al3^{+} + 3e$-…….…….. (1)

The reduction reaction at the cathode,

$2H^{+} + 2e \rightarrow H2$…….…….(2)

The hydrolysis reaction,

$Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^{+}$……(3)

The hydrolysis and polymerization of Al3+ under appropriate pH conditions subsequently give rise to the formation of such species as Al (OH) 2+, Al2 (OH)2 4+, Al(OH)3 and charged hydroxo cationic complexes which can effectively remove pollutants by adsorption.

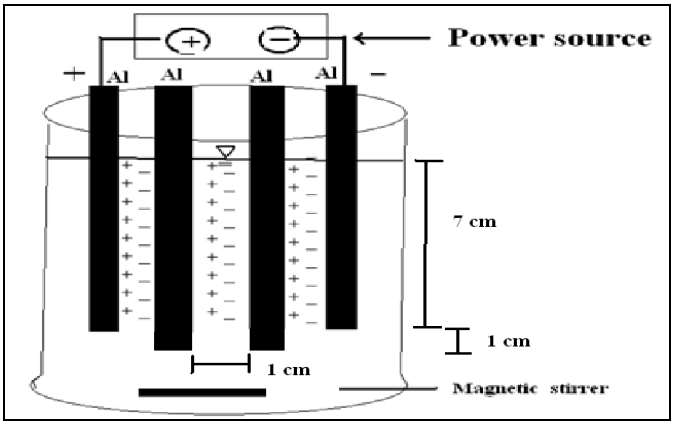

Fig 2: Schematic diagram of the experimental setup

3.2 Electrocoagulation experiment

The experiment was conducted in a bipolar batch reactor (as shown in fig.2.) with 2Al electrodes and some numbers of aluminum plates ware connected in a parallel, as used by C.Y. Hu *et al*. Only the outer electrodes were connected to the power source, and anodic and cathodic reactions occurred on each surface of the inner electrode when the current passed though the electrodes. A constant current is maintained and Magnetic stirring at 400rpm is carried out for a homogenous solution in the batch reactor which contains 1.5 L of water sample. In this experiment, sample from ground water was used, which was collected in some part of Maharashtra, Madhya Pradesh and in local area ground water some amount of fluorine add and this sample was used. The concentration of fluoride is normally 5 to 10 mg/L and the initial pH is 7. Experiments was carried out, at laboratory scale, in cell equipped by 2 aluminum electrodes (anode and cathode with 150cm2 area) in addition with some extra aluminum plates (2 to 5) in order to improving coagulation. The active area of each electrode, excluding the outer electrodes, was 8 cm x 9 cm=72 cm2 and the dimensions of the outer electrodes (7 cm x 7 cm) were reduced to prevent the electric field dispersing. The aluminum electrodes were installed vertically, and connected in a bipolar mode. The net spacing between the electrodes was 10 mm. The current was maintained constant, by means of a precision DC power supply. It was turned on with a voltage kept at a desired value of 10, 20 and 30 V. In each run, 1.5 L treated water was fed to the electrolytic cell. At the end of the each experiment, the solution is filtered through sand filter before analysis. pH of each sample after each runs is determined by a pH meter. The absorbance of fluoride was measured by using spectrophotometer and prepared a standard graph of absorbance vs. concentration.

4. Results and Discussions

4.1 Effect of HRT on fluoride removal

The interval of 5–10 mg/L. was used to determine the influence of the fluoride concentration on the Electro coagulation process with two aluminum electrodes (anode and cathode). The test was first performed at fixed potential of 30V and at difference electrolysis times. The variation of fluoride with electrolysis time is shown in fig.4 the concentration of fluoride dropped from8.2 to 0.6 mg/L in 70 min with increasing electrolysis time and a removal efficiency

of 90%. As shown in fig.4. The rate of fluoride reduction decreases with time and eventually the fluoride becomes near zero.

4.2 Effect of Electro coagulation time on pH

The pH of a solution is one of the most important parameters that govern the removal of fluoride in both the Electro coagulation and flotation processes (Lui *et al*., 1983; Mameri *et al*., 1998; Mollah *et al*., 2001; Shen *et al*., 2003; Matis and Mavros, 1991; Lin and Liu, 1996). Controlling the pH of the solutions, however, is very difficult during the ECF process because the defluoridation reaction changes the pH and the applied current may interfere with the measurement of pH. Moreover, controlling the initial pH in the ECF system is not very useful to vary the final pH. The final pH and residual fluoride concentration did not change very much in the range of initial pH = 4–8 in the EC defluoridation process because of the buffer capacity of aluminum hydroxide (Mameri *et al*., 1998; Shen *et al*., 2003). Therefore, changes in pH value is measured during the test in different times and also, with different number of plates.

Concentration	Absorbance
0	0.181
1	0.177
2	0.173
3	0.165
4	0.16
5	0.156
6	0.151
7	0.144
8	0.14

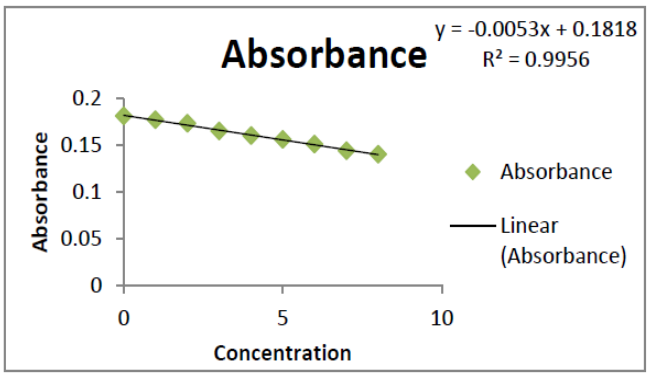

Fig 3: Standard graph of conc. 8 mg/L (Absorbance Vs Concentration)

Standard graph for 2mg/L conc. was made. It was observed that graph is having R2 value is equal to 0.995, which indicate that it is best fitting line. This data was used for calculating the conc. with respect to absorbance.

Time	Concentration	pH
0	8.2	8.03
5	7.6	8.14
10	7.4	8.2
15	7	8.29
20	6.1	8.22
25	5.2	8.03
30	4.8	8.25
35	4.6	8.13
40	4.4	8.39
45	4	8.31
50	3.6	8.3
55	2.6	8.1
60	1.8	7.95
65	1	7.36
70	0.6	7.26

Effect of voltage and time 30 V.

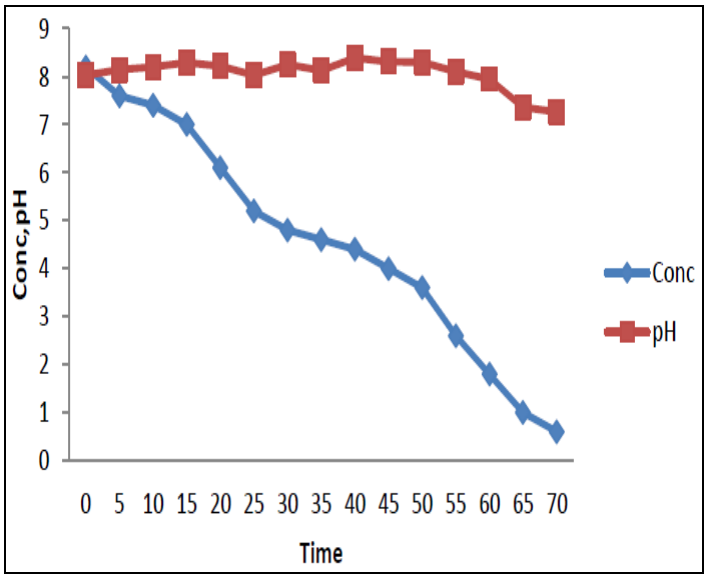

Fig 4: Concentration & pH Vs Time (Conc. = 8.2 mg/L at Volt = 30V)

It is observed that at 30V conc. is decreasing 8.2 to 0.6 within 70 min. It is also can that initially pH uses to 8.39 however it becomes normal at the end of treatment.

4.3 Effect of voltage on fluoride removal

Voltage is the only operational parameter which can be controlled directly because considering reactions, current will change with time. In other word, due to increase of ohmic resistance according to mentioned reactions, electric current would change. Moreover, generation of salt sediments like carbonate salts on aluminum plate could affect the electrical current. In this system electrode spacing is fixed and voltage is

a continuous supply. Fig. 5 shows the fluoride removal as a function of voltage. It is found that the removal of fluoride was increased with increasing voltage.

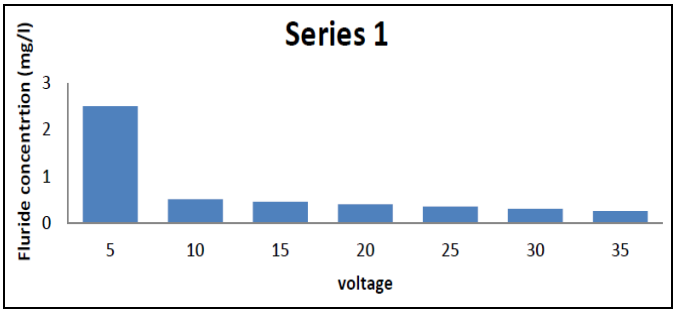

Fig 5: Effect of voltage on fluoride removal.

5. Conclusion

1) Performance of a parallel-plate electro coagulation process with aluminum electrodes is investigated for the removal of fluoride from drinking water.
2) Effect of various parameters like pH, voltage, hydraulic retention time (HRT) and number of aluminum plates between anode and cathode are studied in detail.
3) Electro coagulation is carried out for initial fluoride concentration 5.00-10.00 mg/L and satisfactorily, after 50 min 90% removal is obtained.
4) It could be seen that Variations of pH in EC reactor pH increases from 7 to 8.38. After certain time the pH reaches to permissible range and remains relatively constant.
5) Generation of salt sediments like carbonate salts on aluminum plate could affect the electrical current. It is found that the removal of fluoride was increased with increasing voltage. From this parameters we can conclude that the batch mode process is very efficient.

6. References

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