

Optimization of Parameters for Fluoride Removal by Electrocoagulation using Aluminum Electrodes in Monopolar Parallel Combination

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Abstract— One of the major challenges faced by mankind today is to provide clean drinking water to a vast majority of population. Fluoride in drinking water above permissible limit is responsible for causing fluorosis, a disease affecting the multiple tissues, organs and systems in the body. The present study was conducted to assess the ability of electrocoagulation process for removal of fluoride from synthetic fluoride sample in a batch reactor using four aluminum electrodes in monopolar parallel connection. Operating parameters such as initial fluoride concentration (4-10 mg/L), pH (5-9), voltage (5-25V), and electrolysis time (up to 60 minutes) was studied to evaluate their effect on fluoride removal efficiency. Results obtained revealed that effective fluoride removal within permissible limit (1.2 mg/L, WHO) was obtained at pH 6, 15V, within electrolysis time of 60 minutes.

Keywords—fluorosis, electrocoagulation, fluoride removal, aluminum electrode, electrolysis time

I. INTRODUCTION

Fluorine is one of the elements essential for humans. Naturally it occurs in geochemical deposits, minerals, and natural water systems and gets involved in food chains with drinking water, plants and cereals. Although the fluoride present in drinking water is essential for human health, an excessive intake of fluoride causes severe dental or skeletal fluorosis [1]. Prevalence of fluorosis in India is mainly due to hydro geochemical origin. Problem of fluoride contamination above permissible limit in groundwater has been reported in all 19 states of India [2]. A recent study conducted by UNICEF on the extent and magnitude of fluorosis problem tentatively indicates that 213 districts with affected 25 million people and 66 million people at risk [3]. As per Indian drinking quality standards (IS 10500-1991) fluoride concentration in drinking water has to be 1mg/L and the permissible limit in absence of alternate sources is 1.5 mg/L [4].

Various methods have been employed for fluoride removal from drinking water. Buroff [5] was the first to investigate variety of materials including activated alumina and alum for fluoride removal. NEERI initiated the defluoridation studies in 1961 using materials like clay

minerals, ion exchange resin, activated carbon and alum. Extensively studied and field tested methods for defluoridation are based on adsorption (activated alumina) and co-precipitation (nalgonda technique) which are dependent on aluminum salts where in aluminum compounds play a vital role in fluoride removal [6].

These conventional treatment processes like nalgonda technique, activated alumina & membrane processes have certain drawbacks like high sludge volume generation, extreme acidic /alkaline water & poor selectivity respectively. The process of electrocoagulation (EC) successfully applied to treatment of various wastewater [7] can be employed for fluoride removal from drinking water. This unique method of electrolytic defluoridation is emerging as a better treatment alternative owing to lesser production of sludge and high treatment efficiency at minimal cost [8, 9]. This paper presents outcome of, batch studies carried out to determine the optimum operating parameters for efficient fluoride removal from synthetic fluoride samples using aluminum electrodes in monopolar parallel combination.

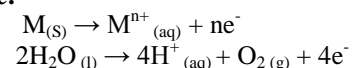
II. ELECTROCOAGULATION PROCESS

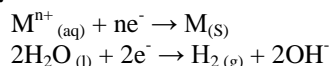
EC is a process in which the anode material undergoes oxidation and hence various monomeric and polymeric metal hydrolyzed species are formed. These metal hydroxides remove organics from water by sweep coagulation and/or by aggregating with the colloidal particles present in the water to form bigger size flocs and ultimately get removed by settling. Coagulating ions are produced in situ and it involves three successive stages [10]:

- Formation of coagulants at anode due to electrical oxidation
- Destabilizing pollutants and suspended substances and emulsion breaking
- Combining instable particles to form flocs.

In this process if M is considered as anode, the following reactions will occur:

At the anode:



At the cathode:

Freshly formed amorphous $M(OH)_3$ has large surface areas that are beneficial for rapid adsorption of soluble organic compounds and trapping of colloidal particles. If aluminum electrodes are used, the generated $Al_{(aq)}^{3+}$ ions will immediately undergo further spontaneous reactions to produce corresponding hydroxides and/or polyhydroxides [11].

III MATERIALS AND METHODS

Lab scale batch EC reactor was designed and fabricated for fluoride removal using acrylic sheet as shown in Fig. 3.1 having dimensions of 15cm×10cm×10cm with a working volume of 1 L and maximum working volume of 1.5 L. Four aluminum electrodes each having dimensions of 100mm×50mm×1mm with an active surface area of 0.0025m² were used in monopolar connection along with electrode spacing of 1 cm for all runs .

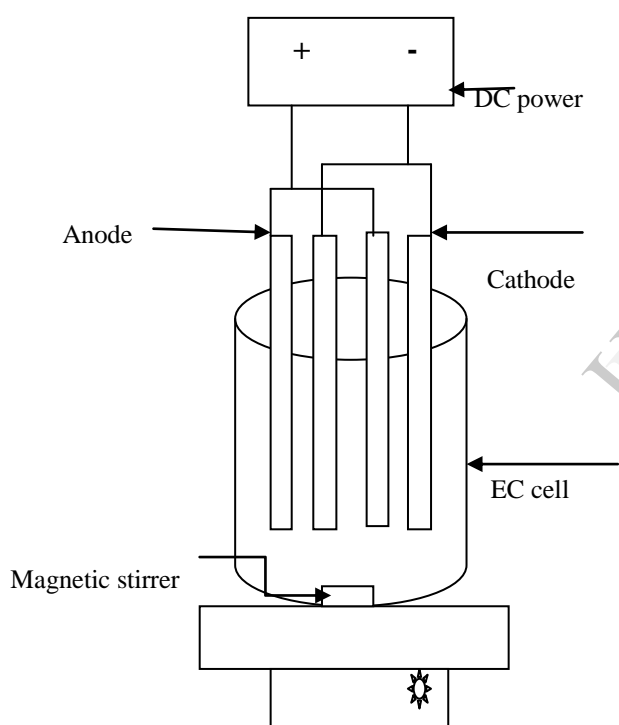


Fig 3.1 Schematic representation of electrocoagulation Process

Synthetic fluoride solutions were prepared by mixing analytical grade sodium fluoride (NaF) with double distilled water. pH and conductivity were measured using calibrated pH meter and conductivity meter . Before each run electrodes were washed and cleansed by HCl solution. Electrolysis was performed using a magnetic stirrer to ensure homogenous mixing and to aid floc formation as shown in Fig. 3.2. Digital DC power supply of 0-30 V and 0-2A was used to maintain constant current and voltage during all runs. To maintain homogenous mixing of the reactor contents magnetic stirrer was used at 120 rpm. Sample is collected every 10 minutes up to 60 minutes and filtered with Whatman

No.1 filter and residual fluoride is measured using fluoride ion meter (Orion, 920 A) using TISAB IV buffer solution to eliminate any aluminum interferences during fluoride measurement.



Fig 3.2 Floc formation in EC process

IV RESULTS AND DISCUSSION

This section includes information related to effects of various parameters such as, electrolysis time [ET], initial fluoride concentration, pH of solution and voltage which will be studied in various range as shown in Table 1 to evaluate their effect on defluoridation efficiency.

Sl No	Table 1. Parameter optimization specification	
	Specification	Remarks
1	Initial fluoride concentration	4,6,8 & 10 ppm
2	Electrolysis time(ET),	Up to 60minutes
3	pH	5,6,7,8 & 9
4	Voltage	5-25 V

A. Effect of Electrolysis time and Initial Fluoride Concentration

Electrolysis time (ET) or treatment time is one of the important parameters of defluoridation in batch EC runs. It basically involves determining the optimum residence time or detention time for removal of fluoride of a certain initial concentration to permissible limit. ET along with current density determines the coagulant dosage (Al^{3+} concentration) and rate of generating bubbles. In the study ET for various initial fluoride concentrations [C_0] of 4, 6, 8 and 10ppm was studied. Increased removal efficiency till the acceptable limit was obtained by increasing the duration of EC process. This is because as time progresses the dissolved coagulants from the aluminum electrode increased according to Faraday's law. A sufficient amount of coagulant dissolved from aluminum electrode trapped the fluoride ions and higher removal efficiency at longer duration was observed as shown in Fig.4.1.

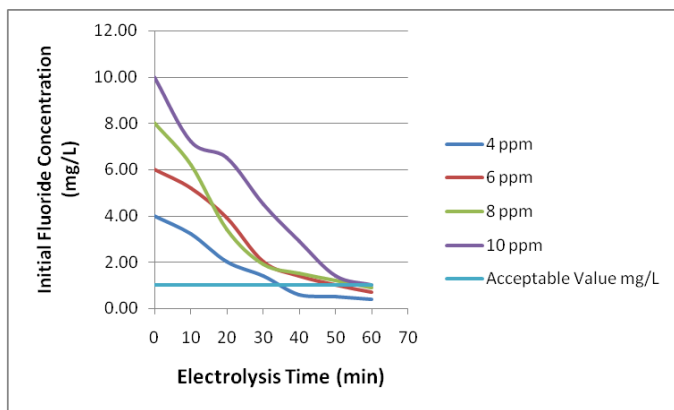


Fig.4.1: Effect of ET on Fluoride removal at various concentration [V=20 V, pH=6]

The result suggested that there is no direct correlation between pollutant concentration and removal efficiencies. And also it was observed that for higher initial fluoride concentration longer time is needed for removal of fluoride than compared to lower initial fluoride concentration. Higher concentration of fluoride was reduced significantly in relatively less time than lower concentration. This can be explained by theory of dilute solutions. In dilute solutions, formation of the diffusion layer has no effect on rate of diffusion or migration of metal ions to the electrode surface [12]. Most of 60-85% of fluoride was removed in the first 30 - 40 minutes as shown in Fig. 4.2.

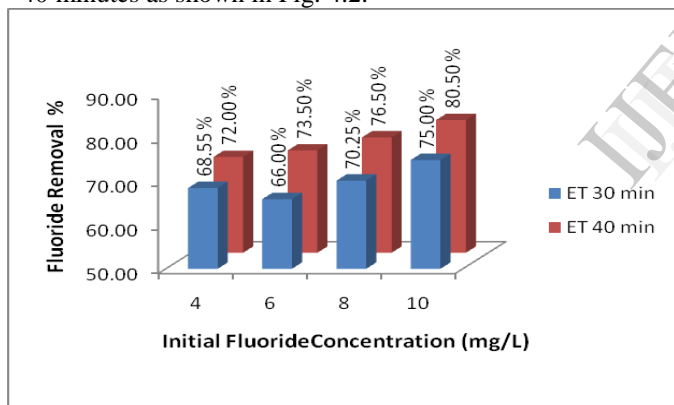


Fig. 4.2: Fluoride removal within 30 and 40 minutes of ET for various Initial Fluoride Concentrations

It was observed that defluoridation efficiency decreases with increasing fluoride concentration as shown in Fig.4.3..

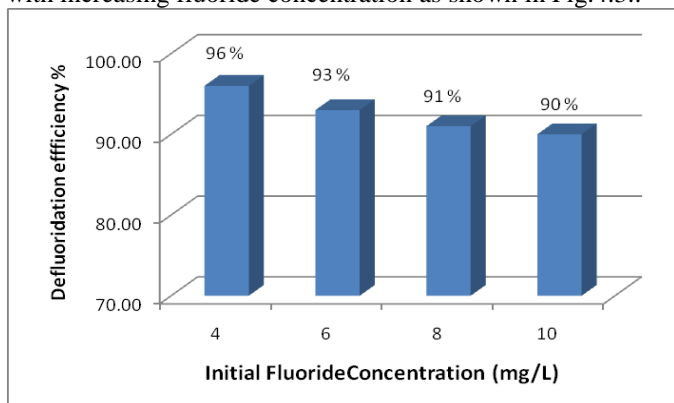


Fig 4.3: Defluoridation Efficiency for various fluoride concentrations

For initial fluoride concentration of 4, 6, 8 and 10 ppm efficiency of 96%, 93%, 91 %& 90% was obtained respectively within an ET of 60 minutes.

B. Effect of pH

The coagulation process depends on pH of solution [13]. It has been established from previous studies that pH has a considerable effect on efficiency of EC process as the rate of coagulation depends on pH of solution. In the study pH is varied in the range 5 - 9 in an attempt to investigate the influence of acidic and basic pH on fluoride removal with increasing ET as shown in Fig 4.4.

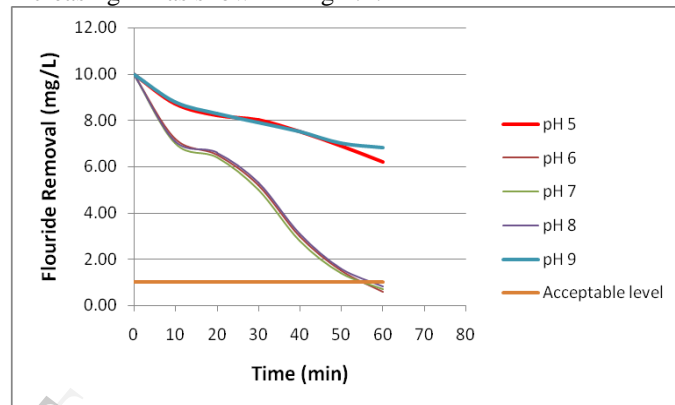


Fig. 4.4: Variation of residual fluoride at different pH [C₀=10ppm V=20 V]

During the process it was observed that there was a considerable change in pH post treatment i.e. an increase in pH was identified as shown in Fig.4.5.

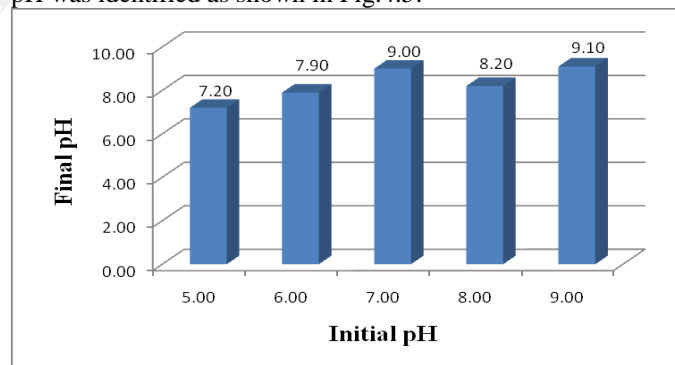


Fig. 4.5: Variation of final pH post treatment [C₀=10ppm, i=18.75m², EC=0.9mS/cm]

pH shift occurs towards alkaline conditions and for higher initial pH of 7 the final pH reaches a value of 9 which is way beyond the limit for drinking water. However at pH 6 the final pH post treatment was within the limits and also had reasonable defluoridation efficiency of 92% and the residual fluoride reached the acceptable limit within 60 minutes of ET. The fluoride removal efficiency between pH 6, 7 and 8 was found to be almost similar (90%, 93% & 92 %) as shown in Fig.4.6.

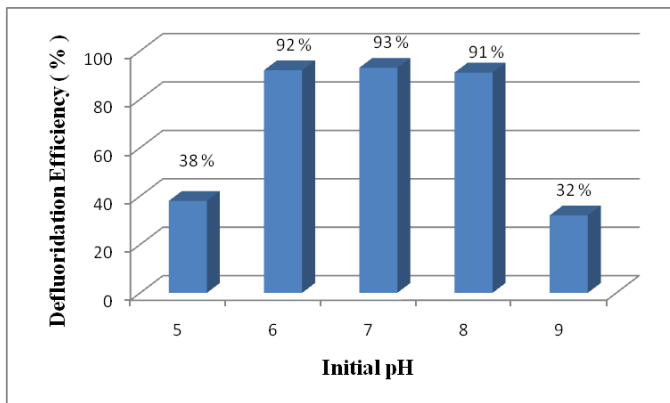


Fig 4.6: Defluoridation efficiency at different pH [$C_o=10\text{ppm}$, $i=18.75\text{m}^2$, $EC=0.9\text{mS/cm}$]

The fluoride removal efficiency at neutral condition (pH - 7) is better than acidic and basic condition due to strong presence of aluminum hydroxide which maximized the fluoro-hydroxide aluminum complex formation [14]

C. Effect of Applied Voltage

The voltage supplied has a direct effect on bubble production rate and on floc growth because of its strong influence on current density [15]. Therefore it has a significant influence on fluoride removal efficiency. As expected it appears that for a given time, the removal efficiency increased significantly with the increase in current density as shown in Fig. 4.7.

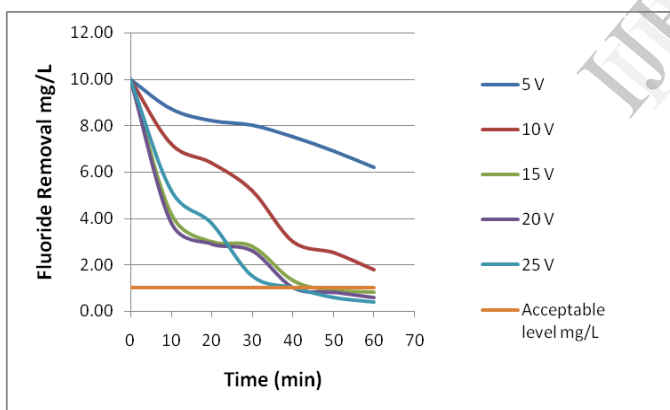


Fig 4.7: Variation in fluoride removal at different voltage range [$C_o=10\text{ppm}$, $\text{pH}=6$]

About more than 50 % of the fluoride removal was observed within only 30 minutes of ET at 25V and lowest fluoride removal at 5V as shown in Fig.4.8. This is because at higher voltage the amount of aluminum oxidized increases

therefore greater amount of coagulant is released for precipitation of fluoride.

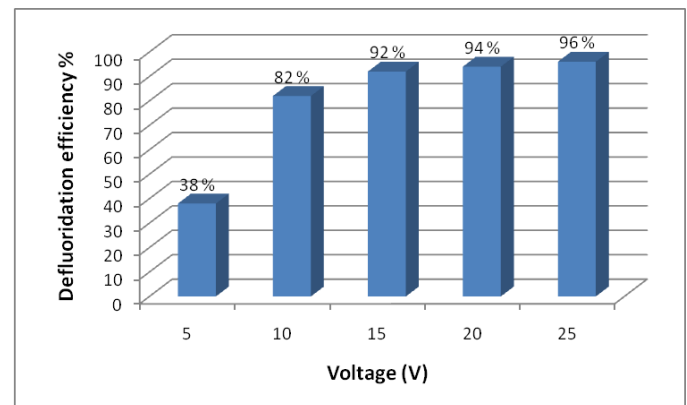


Fig.4.8: Defluoridation efficiency at different voltage ranges [$C_o=10\text{ppm}$, $\text{pH}=6$, $EC=0.9\text{mS/cm}$]

Literature also supports the fact that the bubble density increases and their size decrease with increases voltage which results in greater upward flux and faster removal of fluoride [16]. Specific electrical energy consumption (SEEC) in an electrocoagulation process depends on operating time, voltage and current density.

With increasing voltage the efficiency increases and acceptable fluoride level is reached within short ET but also there is increase SEEC with increased voltage which adds to the cost of the treatment [17] as shown in Fig.4.9.

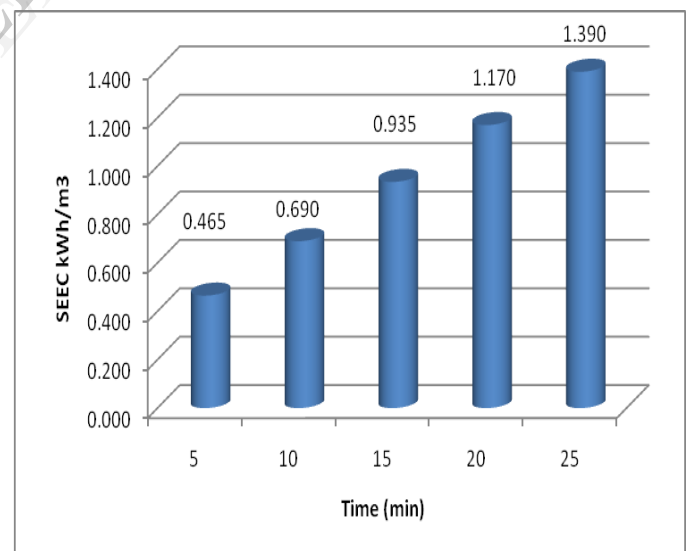


Fig 4.9: SEEC at different voltage range

VI CONCLUSION

Based on the experimental findings, the electrolysis duration of 60 minutes, pH 6 and 15V were found to be the critical operating parameters for removal of fluoride within acceptable limit (1.2 mg/L, WHO) using four aluminum electrodes in monopolar parallel combination. Hence, it can be concluded that the electrocoagulation technology using combination of aluminum electrodes appears to be a feasible alternative for removal of fluoride from drinking water. Thus electrocoagulation holds the potential to be an efficient

defluoridation system is which is fast, economical and can be operated using less equipment and limited space.

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