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TO CONFISCATE FLUORIDE FROM GROUNDWATER

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Abstract

Presence of fluorides in drinking water has become a public health problem. The adverse effects have increased the need to defluoridate water. Several methods like adsorption, coagulation, membrane processes and electrochemical techniques have been studied for defluoridation of water. However, the generation of large volumes of sludge, hazardous waste categorization of metal hydroxides, and high costs associated with chemical treatments have made them less acceptable. Membrane processes are effective, but they increase the operational cost and also remove the beneficial contents of water. Researchers have investigated the influence of various parameters like solution pH, current density, flow rate, residual aluminium, and operational cost on the process of electrocoagulation (EC). Experimental results also suggest that using a multi stage treatment over single stage treatment would be a better option as it increases the defluoridation efficiency and reduces the energy requirements. Identifying the potential of this process can make it the distinct economical and environmental choice for defluoridation of water.

Introduction

Fluoride is a geogenic contaminant of water and its toxic effects are considered to be irreversible. Drinking water with excess fluoride will produce adverse effects that range from mild dental fluorosis to crippling skeletal fluorosis. More and more areas are being discovered which are affected by the problem of fluorides. Therefore the need of the hour is to identify a defluoridation method which is best suited for the purpose. Various techniques have been tested in the past for defluoridation which suffered from few drawbacks mentioned in Table 1. In recent years, there is growing interest in EC. This technique can be used to treat restaurant wastewater (Chen et al., 2000), textile wastewater (Bayramoglu et al., 2004), electroplating wastewater (Adhoum et al., 2004), and fluoride-containing wastewater (Hu et al., 2004 & 2005) effectively. It has also proven its good efficacy for drinking water defluoridation (Mameri et al., 2001).

Fluoride removal mechanism

In the EC process electrolytic dissolution of Al electrodes takes place by oxidation and the reactions are outlined below

Anode: $Al(s) \rightarrow Al^{3+} + 3e^-$ (1)

Cathode: $2H_2O + 2e^- \rightarrow H_2(g) + 2OH^-$ (2)

The H_2 bubbles float and hence drive the flotation process. The Al^{3+} ions further react to form a solid $Al(OH)_3$ precipitate:

$Al^{3+} + 3H_2O <-> Al(OH)_3(s) + 3H^+$ (3)

The basic principle of the process is adsorption of fluoride at precipitated Al $(OH)_3$ as depicted in reaction (Qianhai et al, 2008) (4) :

$Al(OH)_3 + x F^- <-> Al(OH)_{3-x} F_x + x OH^-$ (4)

The above process is pH dependent. The fluoride complexes AlF^{2+}, AlF_2^+ and AlF_4^- dominate in the acidic solution till the pH of the solution reaches 6 and Al $(OH)_3$ precipitates (Emamjomeh et al., 2011). As the solution turns alkaline, Al $(OH)_4^-$ species are formed.

Emamjomeh et al, (2011), have described that mechanism of fluoride removal is not only the competitive adsorption between OH^- and F^- but also the formation of solid cryolite in the final pH range of 5-8. Thus a pH range of 6-8 was considered to be suitable for an efficient defluoridation process.

Parameters affecting EC process

Effect of current density and flow rate

Current density is an important parameter for controlling the reaction rate in most electrochemical processes, because it determines the aluminium dosage within EC process. Also for a fixed volume, the higher the flow rate the lower will be the detention time. Thus the combination of current and detention time determines the electric charge given to the system that drives the aluminium dissolution. From the results in Fig.1 it can be observed that for same current density, as the flow rate is increased the defluoridation efficiency decreases. But when the current density is increased for a constant flow rate, the defluoridation efficiency increases.

Residual aluminium in the effluent

Samples collected from the outlet of an EC reactor were analyzed for aluminium concentration and the results are listed in Table 2. The residual aluminium concentration was in the range of 0.039 mg/l – 0.417 mg/l, which is less than the reported residual Aluminium concentration in Activated Alumina process and Nalgonda technique, 0.16-0.45 ppm and 2.01-6.86 ppm respectively, and is a major drawback of these conventionally used techniques (George et al, 2010).

Operational cost

The continuous flow EC reactor's operational cost (AUD/m^3 of treated water) was estimated by Emamjomeh & Sivakumar (2009) by adding the specific costs of electrical energy, aluminium plate consumption, pH adjustment, and sludge treatment. Total operational cost for defluoridation by Nalgonda process was reported to be AUD 1/m^3 of treated water (TNWSDB, 2005) when initial fluoride concentration was 5 mg/L. At the same initial fluoride concentration, the maximum total operational cost for EC process is found to be AUD 0.6/m^3 of treated water.

Experimental Results

Single *vs* Double Stage electro coagulation process

Energy consumption has been used to compare the performances of single stage and double stage treatment continuous EC treatment process. Chemical analysis of fluoride contaminated sample is listed in Table 3. For flow rate of 150 ml/min and

200 ml/min and applied current of 0.15A. 0.30 A and 0.45A, samples were collected for 20 minutes after the pseudo steady state from the outlet of the reactor. The energy consumed in 20 minutes reaction time was measured and fluoride content was determined for the collected samples. It can be observed from the results that double stage treatment provides better removal than single stage. It can be clearly observed from results that at flow rate of 200 ml/min, spending 2.7 J/mg in single stage treatment does not lower the fluoride concentration to permissible levels but spending 2.6 J/mg in double stage treatment lowers the fluoride concentration to permissible levels. Hence double stage treatment is energy efficient. At high applied current of 0.45 A, more energy per unit fluoride removal is required but the fluoride removal is not significant. The reason is that with increase in current the reaction rate increases which cause rapid increase in pH, affecting the overall fluoride removal. Therefore to avoid energy wastage, the current which reduces fluoride to desirable levels should be used. For all the experiments of double stage treatment, fluoride content in the effluent is well within WHO, 2004 prescribed limit (less than 1.5) (Refer Figure 3).

Conclusion

EC is attractive strategy in which no contaminants are introduced. Beneficial contents present in raw water can be remained during defluoridation. Reported advantages of this process are reduced sludge production, no chemical handling and lesser cost than the Nalgonda technique. But the process has its own drawbacks of periodical replacement of the sacrificial anodes and minimum requirement of conductivity necessary to facilitate the flow of current. Identification of the potential of this process can make it the distinct economical and environmental choice for defluoridation of water.

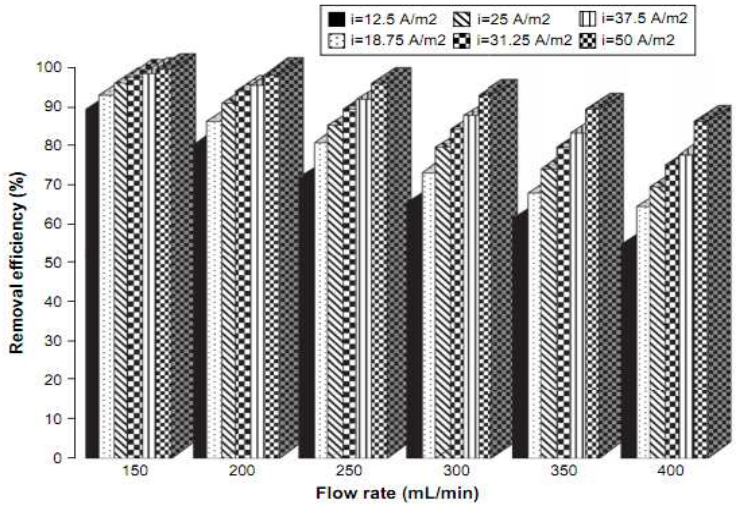

Figure 1. Effect of flow rate on the defluoridation efficiency at different current densities from 12.5 to 50 A/m^2 in a continuous flow electro coagulator

(Initial F- =15 mg/L, pH in = 6, Electrical conductivity (Ec) = 50 mS/m)

[Source: Emamjomeh & Sivakumar, 2009]

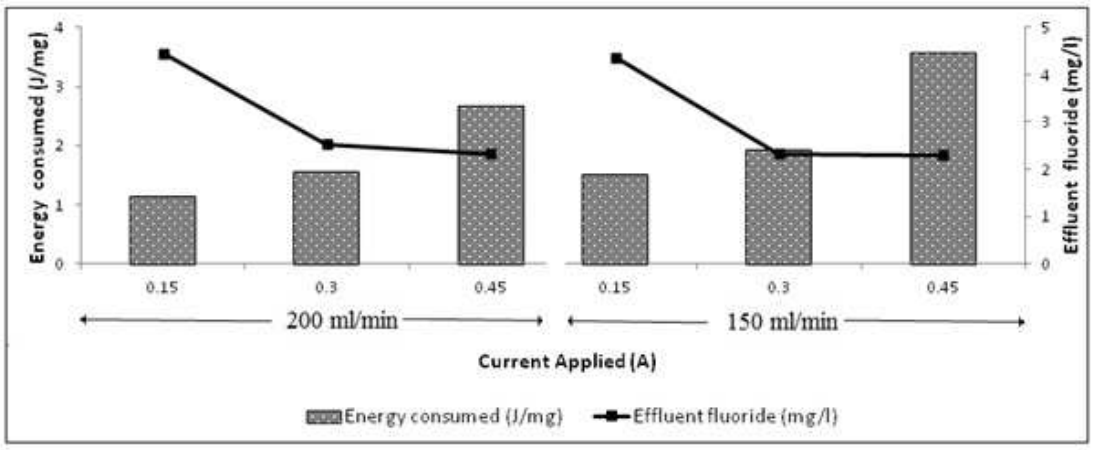

Figure 2. Effect of energy consumption on effluent fluoride concentration for single stage treatment at flow rate of 200 ml/min and 150 ml/min.

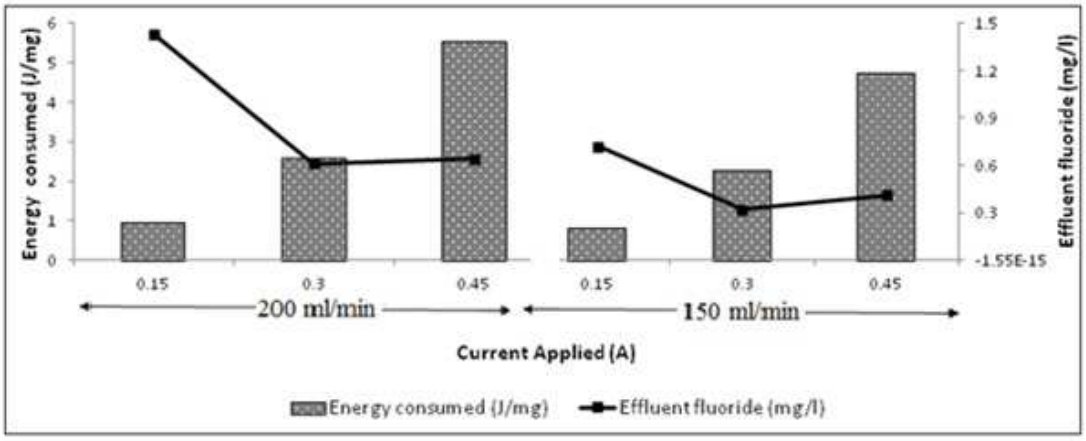

Figure 3. Effect of energy consumption on effluent fluoride concentration for double stage treatment at flow rate of 200 ml/min and 150 ml/min.

Table 1:

Limitations of Defluoridation techniques (Meenakshi & Maheshwari, 2006)

Techniques	Limitations
Activated Alumina	• Presence of sulphate, phosphate or carbonate results in ionic competition • Low adsorption capacity, poor integrity and needs pretreatment • Effectiveness of adsorbent reduces after each regeneration • Disposal of fluoride laden sludge and concentrated regenerant
Nalgonda technique	• Presence of soluble aluminium fluoride complex. • Regular analysis of feed and treated water. • High maintenance cost & Sludge disposal
Membrane processes	• Expensive • Disposal of brine water

Table 2.

Observations of residual Al at varying applied current and initial fluoride concentration

Sample	Initial Fluoride Conc (mg/l)	Current (A)	Residual Al (mg/l)
1	6	0.15	0.340
2	6	0.30	0.356
3	6	0.45	0.417
4	4	0.15	0.039
5	4	0.30	0.089
6	4	0.45	0.134

Table 3.

Chemical analysis of fluoride contaminated sample water

Chemical Parameters	Concentration
Alkalinity (total), mg/l	394
Total dissolved solids, mg/l	1120
Conductivity, mS/cm	1.90
pH	8.35
Fluoride, mg/l	6

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