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# DEFLUORIDATION USING A CONTINUOUS ELECTROCOAGULATION (EC) REACTOR

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## ABSTRACT

Electrocoagulation (EC) is a process of passing a steady electric current through liquid using aluminium or iron electrodes to remove impurities in water. When aluminium electrodes are used, the aluminium goes into solution at the anode and hydrogen gas is released at the cathode and dissolution of Al anodes produces aqueous aluminium species. Experiments were undertaken to investigate the effects of the different parameters such as: current density (12.5 – 50 A/m<sup>2</sup>), flow rate (150 – 400 mL/min), initial pH (4-8), and initial fluoride concentration (5-15 mg/L) on the fluoride removal efficiency in a continuous flow electrocoagulator. The experimental results showed that for an initial fluoride concentration of 10 mg/L when flow rates varied from 150 to 300 mL/min, the residual fluoride concentration reached to less than 1 mg/L when the current densities were respectively increased from 18.75 A/m<sup>2</sup> to 50A/m<sup>2</sup>. It appears that for higher defluoridation efficiency, the current density needs to be increased when flow rate is increased. The composition of the sludge produced was analysed using X-ray diffraction (XRD) spectrum. The strong presence of the hydroxyl-aluminium in the final pH range between 6 and 8, which maximizes the formation of fluoro-hydroxide aluminium complex, is the main reason for defluoridation by electrocoagulation. The results obtained showed that the continuous flow electrocoagulation technology is an effective process for defluoridation of potable water supplies and could also be utilized to the defluoridation of industrial wastewater.

## KEYWORDS

Electrocoagulation (EC), Fluoride removal, Continuous flow reactor, Water and wastewater treatment

## 1 INTRODUCTION

Fluoride has beneficial and harmful effects on the environment. The beneficial effect of fluoride is its capacity to help prevent dental carries in humans when an optimum amount of 1 mg/L is present in the drinking water. However, consumption of water containing excessive fluoride can lead to fluorosis of the teeth and bones. Long-term consumption of water containing 1.5 mg/L of fluoride leads to dental fluorosis (WHO, 2005). White and

yellow glistening patches on the teeth may be seen that may eventually turn into brown. Skeletal fluorosis is a bone disease caused by excessive consumption of fluoride. In groundwater, the natural concentration of fluoride depends on the geological, chemical and physical characteristics of the aquifer, the porosity and acidity of the soil and rocks, the temperature, the action of other chemical elements, and the depth of wells. Due to these variables, the fluoride concentrations in groundwater can range from less than 1 mg/L to more than 35 mg/L. In India and Kenya, concentrations up to 38.5 mg/L and 25 mg/L have been reported, respectively. The total number of people affected is not known, but an estimate would number in the tens of millions (Qian *et al.*, 1999). The fluorosis problem is most severe in the two largest countries of the world, China and India. In China, the World Health Organization (WHO) recently estimated that 2.7 million people have the crippling form of skeletal fluorosis, while in India, 15 of its 32 states have been identified as "endemic", with over 6 million people seriously afflicted (WHO, 2002). Fluoride has thus become a major public health problem in both of these countries. Waters with high fluoride content are usually found at the foot of high mountains and in areas with geological deposits of marine origin. Areas with these characteristics are found in the geographical belt from Syria through Jordan, Egypt, the Libyan Arab Jamahiriya, and Algeria to Morocco, and the Rift Valley through Sudan and Kenya. Another belt is the one stretching from Turkey through Iraq, the Islamic Republic of Iran, and Afghanistan to India, north Thailand, and China. Similar areas can be found in the Americas and in Japan and China (WHO, 2005). In Australia, the highest fluoride concentration was recorded 13 mg/L in a water bore near Indulkana that is not, however, used for human consumption (Fitzgerald *et al.*, 1999). The maximum acceptable concentration of fluoride in drinking water is 1.5 mg/L (NHMRC. and ARMCANZ, 2004). Health impacts from long-term use of high fluoride concentration in drinking water have been summarized in Table 1.

Fluoride also can be found in industrial wastewaters, such as in glass manufacturing industries (Sujana *et al.*, 1998) and in high concentrations in semiconductor industries (Toyoda and Taira, 2000). The discharge of these wastewaters without treatment into the natural environment would contaminate groundwater. There are several defluoridation processes that have been tested globally, such as adsorption (Lounici *et al.*, 1997), chemical precipitation (Parthasarathy *et al.*, 1986; Sujana *et al.*, 1998; Toyoda & Taira, 2000), electrodialysis (Amor *et al.*, 2001), and electrochemical method (Ming *et al.*, 1983; Li-Cheng, 1985; Mameri *et al.*, 1998). In the precipitation technology, alum or combination of alum and lime are added respectively to water with low and high concentration of fluoride. Fluoride is then removed by flocculation, sedimentation and followed by filtration. Using chemical coagulants for precipitation is one of the most essential processes in conventional water and wastewater treatment. Generation of large volumes of sludge, the hazardous waste categorization of metal hydroxides, and high costs associated with chemical treatments have made chemical coagulation less acceptable compared to other processes. If a new process can replace the conventional chemical coagulation, the process efficiency would increase with little modification to present infrastructure in water and wastewater treatment plants and many problems caused by chemical coagulation would also be solved. This new process, called electrocoagulation (EC), is an electrochemical technique with many applications and more recently it has been suggested that EC as an

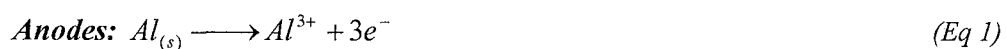
alternative to conventional coagulation (Mills, 2000). The previous results showed that the electrocoagulation technology is an effective process for defluoridation of potable water supplies and could also be utilized to the defluoridation of industrial wastewater (Ming et al., 1983; Drondina and Drako, 1994; Mameri et al., 1998; Yang and Dluhy, 2002; Emamjomeh *et al.*, 2003; and Shen et al., 2003). It has been suggested that the electrocoagulation process for fluoride removal does not require a big investment (Mameri *et al.*, 1998).

The previous results on the defluoridation using batch experiments by Emamjomeh *et al.*, (2003); and Emamjomeh and Sivakumar, (2004) showed that electrocoagulation using aluminium anodes is an effective defluoridation water treatment process. This research is directed towards improving this effective removal technology, for removing fluoride concentration in water and wastewater treatment processes by a continuous electrocoagulation system. This study elucidates the effects of current density, initial fluoride concentration, initial pH, and flow rate on the fluoride removal efficiency of a continuous EC reactor.

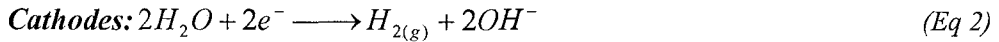
## 2 FUNDAMENTALS OF ELECTROCOAGULATION (EC):

Electrocoagulation process provides an alternative technique for the removal of pollutants from wastewater. The process involves the application of an electric current to sacrificial electrodes inside a reactor tank. The current generates a coagulating agent as well as gas bubbles. In addition, electrocoagulation, electroflocculation, and electroflotation are three techniques involving the electrolytic addition of coagulating metal ions directly from sacrificial electrodes. These ions coagulate with pollutants in the water, in a similar manner to the addition of coagulating chemicals such as alum and ferric chloride, and allow the easier removal of the pollutants by both sedimentation and flotation process. Interaction occurring within an EC reactor is shown in Figure 1. In the EC process, the aluminium goes into solution at the anode and hydrogen gas is released at the cathode when aluminium electrode is used. The aluminium coagulates with the pollutants that can then be removed, usually by either settling or flotation.

Aluminium is naturally present in some waters but arises in drinking water predominantly through the use of aluminium sulphate in the coagulation process of water treatment. The added aluminium precipitates under the most common conditions as aluminium hydroxide, removing suspended material and some dissolved substances by adsorption and enmeshment within the precipitate. The precipitated aluminium hydroxide is removed by settlement and filtration. Although there has been a move away from using Alum as a coagulant in the water industry due to its possible link to Alzheimer's disease (Khachaturian and Radebaugh, 1996), it still remains in use as a coagulant agent in many countries. The electrolytic dissolution of Al anodes by oxidation in water produces aqueous  $Al^{3+}$  species (Sposito, 1996) and the electrochemical reactions are outlined below:



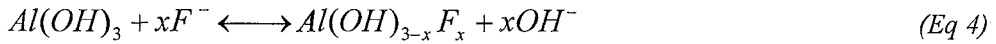
At the aluminium cathodes reduction takes place which results in hydrogen bubbles being produced by the following reaction:



The H<sub>2</sub> bubbles float and hence drive the flotation process. The Al<sup>3+</sup> ions further react as shown in Eq 3 to a solid Al(OH)<sub>3</sub> precipitate. These precipitates form flocs that combine water contaminants as well as a range of coagulant species and metal hydroxides formed by hydrolysis:



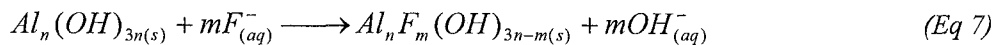
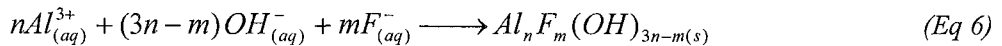
These coagulants destabilise and aggregate suspended particles or precipitate and adsorb dissolved contaminants. For example, the Al(OH)<sub>3</sub> floc is believed to adsorb F<sup>-</sup> strongly (Shen *et al.*, 2003) as shown by equation (4):



Freshly formed amorphous Al(OH)<sub>3</sub> precipitates that are required for “sweep coagulation” have large surface areas, which are beneficial for a rapid adsorption of soluble compounds and trapping of colloidal particles. These flocs polymerize usually at high Al concentrations as:



An investigation into the mechanisms of aluminium salt for adsorption of fluoride was conducted by Hu *et al.*, (2003). They have found that precipitation (Eq. 6) or adsorption (Eq. 7) reaction may occur when aluminium salt is used for fluoride removal.



The characteristics of destabilization mechanism of the contaminants, and particulate suspension has been summarized (Mollah *et al.*, 2001; Shen *et al.*, 2003) as follows:

- Colloid destabilizing agents that effect charge neutralization are produced by electrolysis in an electrocoagulation system.
- The floc formed as a result of coagulation creates a sludge blanket that entraps and bridges colloidal particles, such as the metallic cation interacts with OH<sup>-</sup> to form a hydroxide, which has high adsorption properties thus enabling bonding with the pollutant.

- The fluoroaluminium complexes were strongly present inducing an efficient aluminium complexation by fluoride.

Competitive adsorption between the hydroxide ion  $\text{OH}^-$  and  $\text{F}^-$  may exist. Sludge characterization results showed that the fluoridation is a chemical adsorption process with  $\text{F}^-$  replacing the  $\text{OH}^-$  group from the  $\text{Al}_n(\text{OH})_{3n}$  flocs (Shen *et al.*, 2003)

### 3 MATERIALS AND METHODS

#### 3.1 EXPERIMENTAL PROCEDURE

A continuous flow pilot scale electrocoagulation reactor was designed and constructed for fluoride removal as shown in Figure 2. For the electrochemical cell, two aluminium (purity of Al 95–97%, Ullrich Aluminium Company Ltd, Sydney) plate anodes and cathodes (dimension 950×200×3 mm) were used as electrodes. The electrodes were dipped 800 mm into an aqueous solution (net volume 7.9 L) in a Perspex reactor (dimension 1000×210×48 mm). The reactor could be configured with a sedimentation and flotation tank (net volume 49 L). The gaps between the two neighbouring electrode plates were kept constant at 5 mm for all the experiments.

Current was varied over the range 2 – 8A, however, it was held constant for each run. The initial pH of the water was varied for each experiment within the range of 4-8. 1 M sodium hydroxide and 1:5 hydrochloric acid solutions were used for pH adjustment. The influence of the various parameters on the defluoridation process was achieved using “synthetic” water (Wollongong city tap water + NaF salt + NaCl) in a continuous reactor. Fluoride solutions (5 -15 mg/L as  $\text{F}^-$ ) were prepared by mixing sodium fluoride in tap water. The conductivity of the solution was increased with 0.005 M NaCl to 50mS/m. Fluoride concentration was determined using the ionometric standard method (AWWA and WEF, 1998) with a fluoride selective electrode (Metrohm ion analysis, Fluoride ISE 6.0502.150, Switzerland). To prevent interference from the  $\text{Al}^{3+}$  ion, TISAB buffer [58 g of NaCl, 57 mL of glacial acetic acid, 4 g 1,2 cyclohexylenediaminetetraacetic (CDTA), 125 mL 6N NaOH were dissolved in 1000 mL distilled water with stirring until pH 5.3 – 5.5 was reached] was added to the samples. Direct current from a DC power supply (0–18 V, 0–20 A, ISO-TECH, IPS-1820D) was passed through the solution via the electrodes. Cell voltage and current were readily monitored using a digital power display. Conductivity and pH were measured using a calibrated conductivity meter and pH meter, respectively. The composition of sludge was analysed by X-ray diffraction (XRD) technique. The XRD is a method for determining the mineralogical structure of a compound. The XRD measurements were carried out using Philips no RN 1730 with  $\text{CuK}_\alpha$  source. The analysers were fitted by ICCD standard patterns and Trace 5 software. Pass voltage of 40 kV and current of 20 mA were used for its spectra.

Total aluminium concentration was determined based on standard method (AWWA and WEF, 1998) by acidifying a sample with concentrated nitric acid (HNO<sub>3</sub>) to pH<2. The samples were transferred to a beaker, 5 mL of concentrated acid HNO<sub>3</sub> was added and it was boiled without allowing sample to dry. The samples were cooled, filtered and then transferred to a 100 mL volumetric flask and make up to the mark with distilled water. Once samples are prepared, total aluminium concentration was determined using Atomic Absorption Spectrophotometer. Dissolved aluminium concentration was determined based using standard method (AWWA and WEF, 1998) by Eriochrome cyanin R method. It provides a means for estimating aluminium with low concentration (less than 0.5 mg/L). After preparation of a calibration curve, absorbances of filtered samples were read on a spectrophotometer, using a wavelength of 535 nm. Negative errors can be caused by fluoride interference in solution. A simpler correction can be determined from the family of curves (Figure 306:1 from AWWA and WEF, 1998) for estimation of dissolved aluminium concentration in the presence of fluoride.

## 4 RESULTS AND DISCUSSION:

### 4.1 EFFECT OF CURRENT DENSITY AND FLOW RATE

In most electrochemical processes, current is the most important parameter for controlling the reaction rate in the reactor, because current determines the coagulant dosage within electrocoagulation. It is important to determine the Al<sup>3+</sup> dose achieved within the ECF process. Faraday's law can be used to describe the relationship between current value (I), electrolysis time (t) in second, volume of reactor and the amount of aluminium, which goes into solution (gr/m<sup>3</sup>) (Scott, 1995).

$$C_{Al(T)} = \frac{It.M_{(Al)}}{Z.F.V} \quad (Eq\ 8)$$

where C<sub>Al(T)</sub>, Z, F, V, and M<sub>(Al)</sub> are the theoretical concentration of Al<sup>3+</sup> (gr/m<sup>3</sup>), aluminium valance, Faraday constant (96500 C/mol), volume of electrobox (m<sup>3</sup>), and molecular weight of aluminium (27 gr/mol), respectively.

In Eq. 8, where t is the mean hydraulic retention time (min), using flow rate (Q) in mL/min and current density (I/A), Eq. 8 can be rearranged to give:

$$C'_{Al(T)} = \frac{i.M_{(Al)}}{Z.F.Q} \quad (Eq\ 9)$$

Where C'<sub>Al(T)</sub> and i are the amount of aluminium concentration goes to into volume of solution per unit area of electrode (gr/m<sup>2</sup>.mL) and current density (A/m<sup>2</sup>), respectively. The rate of an electrochemical reaction is measured as current density defined as current per unit area of electrode. For an initial fluoride concentration of 10 mg/L, the effect of current density on the fluoride removal efficiency is shown in Figure 3. The fluoride removal efficiency is increased from 89% at 12.5 A/m<sup>2</sup> to 99% at 50 A/m<sup>2</sup> when the flow rate and initial fluoride concentration are respectively kept at 150 mL/min and 10 mg/L. The current density determines the rate of

dissolution of  $\text{Al}^{3+}$  concentration. In lower current density, the less aluminium is released from the anode and hence the fluoride reduction is low.

In the EC process, the amount of the aluminium ion produced is proportional to the amount of current supplied and flow rate (Eq. 8). The effect of flow rate on defluoridation efficiency is studied by varying flow rates in the ranges 150 to 400 mL/min at different initial fluoride concentrations and current densities.

As seen in Figure 4, for a flow rate of 400 mL/min, the fluoride removal efficiency was increased from 55% at 12.5  $\text{A/m}^2$  to 86% at 50  $\text{A/m}^2$ . It can be seen that when the flow rates are increased, the fluoride removal efficiency decrease for the same current densities. So, it appears that for higher defluoridation efficiency, the current density needs to be increased when flow rate is increased.

From Eq. 8, the highest current (8A) produced the quickest fluoride removal due to the ready availability of  $\text{Al}^{3+}$  ions in the solution. However at higher currents, more coagulant (aluminium) is available per unit time, which may be unnecessary, because not only excess residual aluminium is unsafe for drinking water but also high current is also uneconomic in terms of energy consumption. The optimum current densities for different flow rates have been summarised in Table 2 for an initial fluoride concentration of 10 mg/L and initial pH of 6.

#### **4.2 EFFECT OF INITIAL FLUORIDE CONCENTRATION AND FLOW RATE**

In order to find out the effect of initial fluoride concentration on defluoridation efficiency, the experiments were conducted by changing initial fluoride concentration from 5 mg/L to 15 mg/L at the same current densities and flow rates. The results are shown in Figure 5 and it indicates that when the initial fluoride concentration is low the removal efficiency is highest. The residual fluoride concentration was decreased from 1.94 mg/L at 12.5  $\text{A/m}^2$  to 0.47 mg/L at 50  $\text{A/m}^2$  when the flow rate and initial fluoride concentration were respectively kept at 400 mL/min and 5 mg/L. The effect of initial fluoride concentrations on defluoridation at different current densities from 12.5 to 50  $\text{A/m}^2$  is shown in Figure 6. In this experiment, the flow rates are increased from 150 to 400 mL/min. It can be seen that for the same current density (12.5  $\text{A/m}^2$ ) and flow rate (150 mL/min), the residual fluoride concentration increases from 1.1 to 1.6 mg/L when initial fluoride concentration is increased respectively from 10 to 15 mg/L. At higher values of flow rate (400 mL/min) and current density (50  $\text{A/m}^2$ ), the residual fluoride concentration also increase from 1.39 to 2.1 mg/L when initial fluoride concentration is increased from 10 to 15 mg/L, respectively. This is possibly due to the formation of insufficient amount of aluminium hydroxide complexes. It appears that higher initial fluoride concentration needs higher current for enhancing the defluoridation efficiency.

#### **4.3 EFFECT OF PH**

The electrocoagulation process is highly dependent on the pH of the water or wastewater to be treated (Mameri *et al.*, 1998; Ming *et al.*, 1983). The initial pH is one of the important solution chemistry factors in terms of raw water but also as a potential operational variable. pH will affect the speciation of Al and it will have a significant



influence on the defluoridation mechanism. Since aluminium hydroxide is an amphoteric hydroxide, pH is a sensitive factor for the formation of  $\text{Al}(\text{OH})_3$  flocs.

Coagulation with aluminium salts occurs at a wide range of pH due to different mechanisms, the amorphous aluminium hydroxide is least soluble at a pH close to 8 (Sposito, 1996). The solubility of aluminium in equilibrium with solid phase  $\text{Al}(\text{OH})_3$  depends on the surrounding pH; between pH 5 and 6, the predominant hydrolysis products are  $\text{Al}(\text{OH})^{2+}$  and  $\text{Al}(\text{OH})_2^+$ ; between pH 6 and 8, the solid  $\text{Al}(\text{OH})_3$  is most prevalent; and above pH 9, the soluble species  $\text{Al}(\text{OH})_4^-$  is the predominant species. MINEQL<sup>+</sup> model (Environmental Research Software, 1999) was utilized to show in Figure 7 how different pH would influence the solubility of Gibbsite [ $\text{Al}(\text{OH})_3$ ].

Effect of influent pH on the fluoride removal by EC process is shown in Figure 8. When the influent pH value increased from 4 and 6, the final pH was increased respectively from 6 to 8 and the residual fluoride concentration reached to an equilibrium value of 0.74 mg/L. By increasing initial pH from 6 to 8, the residual fluoride concentration increased from 0.74 to 1.05 mg/L and the final pH reached to 9. It is clear that the defluoridation efficiency decreases when initial pH is increased from 6 to 8. It is because very high pH will lead to the formation of  $\text{AlO}_2^-$ , which is soluble and not useful for defluoridation. The results, reported in Figure 8, show that the defluoridation process is more efficient for a final pH ranging from 6 to 8. Therefore, it can be concluded that at this pH range, the majority of aluminum complexes (coagulants) are formed and it is the optimum pH range for carrying out the electrocoagulation.

#### 4.4 CHARACTERISTICS OF SLUDGE

The presence of fluoride can produce marked changes in the progress of Al in water. Since fluoride ion forms strong complexes with aluminum, it can considerably increase the solubility of Al (Townsend and Bache, 1992). In the presence of fluoride, the distribution curves in Figure 9 show that the fluoride complexes  $\text{AlF}^{2+}$ ,  $\text{AlF}_2^+$ ,  $\text{AlF}_3$ , and  $\text{AlF}_4^-$  predominate in acid solution until  $\text{Al}(\text{OH})_3$  precipitates. As noted in section 4.3, based on the formation of  $\text{Al}(\text{OH})_4^-$ , which are soluble and not useful for defluoridation, the fluoride removal efficiency is decreased when the final pH is beyond 10.

The composition of the sludge produced in the continuous flow reactor was analysed using X-ray diffraction (XRD) spectrum at the final pH range of 6-8. The compositions of the sludge were studied in two steps, first by XRD and then by a trace analysis software. As seen in Figure 10 and Figure 11, the strongest peaks appeared at degree 18 and 20, which were identified to be the aluminium fluoride hydroxide and aluminium hydroxide, respectively. This result is in agreement with the results obtained by (Shen *et al.*, 2003). Shen *et al.* (2003) also reported that other peaks were identified to be  $\text{Al}(\text{OH})_2$ ,  $\text{Al}(\text{OH})\text{F}$ ,  $\text{AlFO}_2\text{H}$ , and  $\text{AlF}_2$  when the composition of the sludge was analysed using X-ray photoelectron spectroscopy (XPS) and time-of-flight secondary ion mass spectroscopy (TOF-SIMS).

In order to explore the mechanism of fluoride removal, the results from XRD spectrum showed that competitive adsorption between the hydroxide ion  $\text{OH}^-$  and  $\text{F}^-$  may exist. In the final pH range of 6–8, sludge characterization results showed that residual fluoride in such solution may occur in precipitated solid particles such as  $\text{Al}(\text{OH})_{3-x}\text{F}_x$ .

## 5 CONCLUSIONS

Continuous flow experiments were designed to investigate the effects of the different parameters including current density, initial fluoride concentration, initial pH and flow rate on the fluoride removal efficiency by EC process. The removal efficiency was found to be dependent on the applied current density, the flow rate and the initial fluoride concentration of the solution for final pH ranging between of 6 and 8. The highest treatment efficiency was obtained for the largest current density. However, it is advisable to limit the current density in order to avoid excessive energy consumption provided a desirable effluent fluoride concentration range is achieved. It can be concluded that for achieving residual fluoride concentration for reaching health standards, the optimum current density is considered to be  $18.75 \text{ A/m}^2$  for a flow rate of 150 mL/min and for an initial fluoride concentration of 10 mg/L. The optimum current density can be increased to  $50 \text{ A/m}^2$  if flow rate is to be increased to 400 mL/min. XRD analysis of the composition of the dried sludge obtained by EC process shows the formation of  $\text{Al}(\text{OH})_{3-x}\text{F}_x$  and it is the main reason for defluoridation by electrocoagulation process.

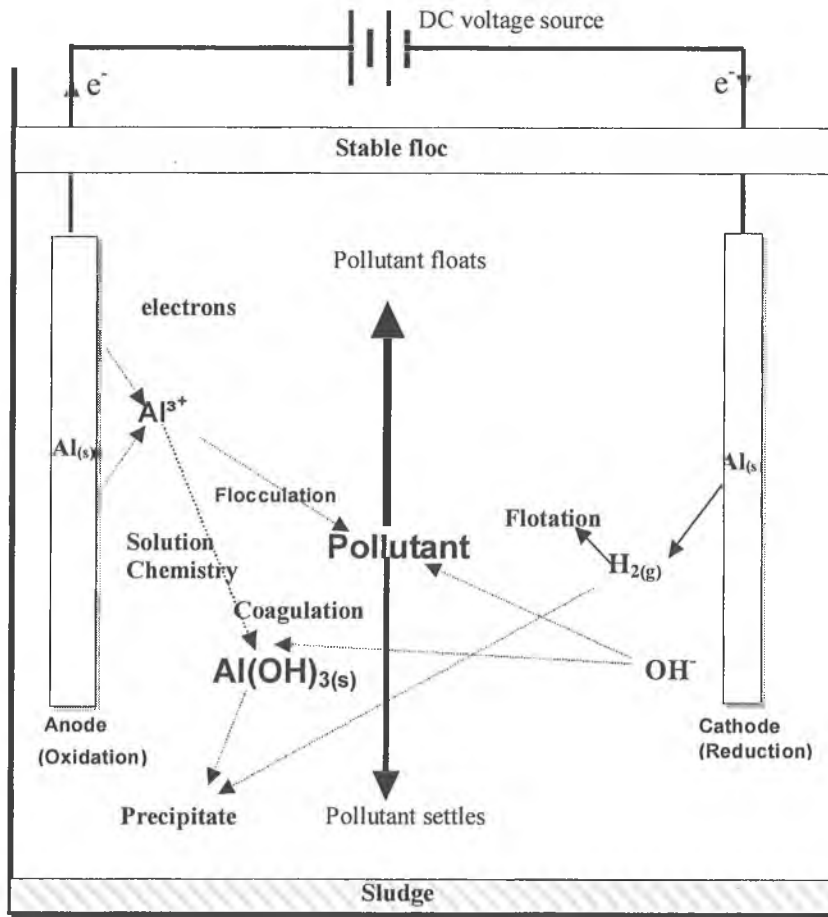


Figure 1: Pollutant separation processes in an electrocoagulation reactor (adapted from Holt et al., 2002)

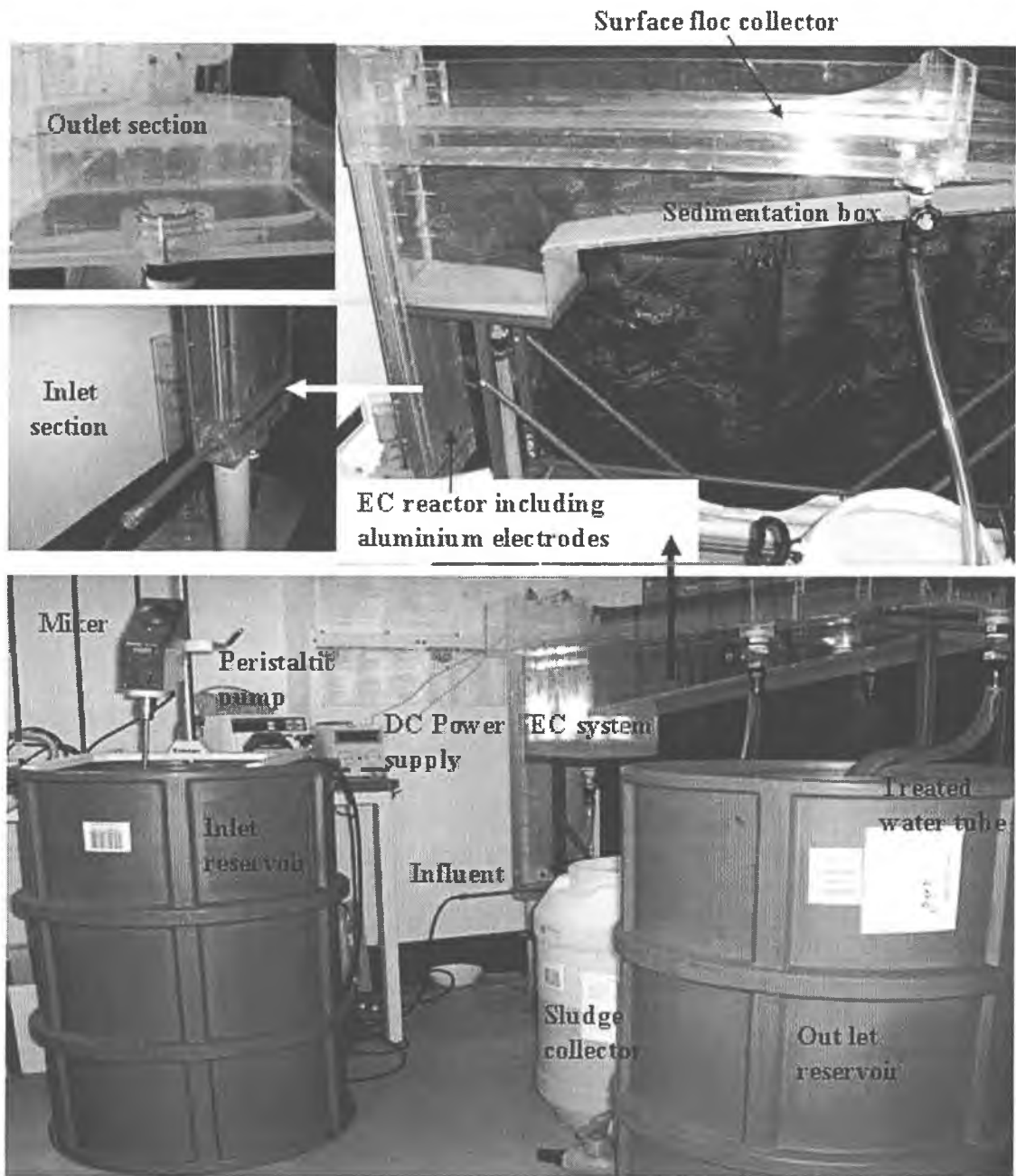


Figure 2: Experimental set up of a continuous flow electrocoagulation system

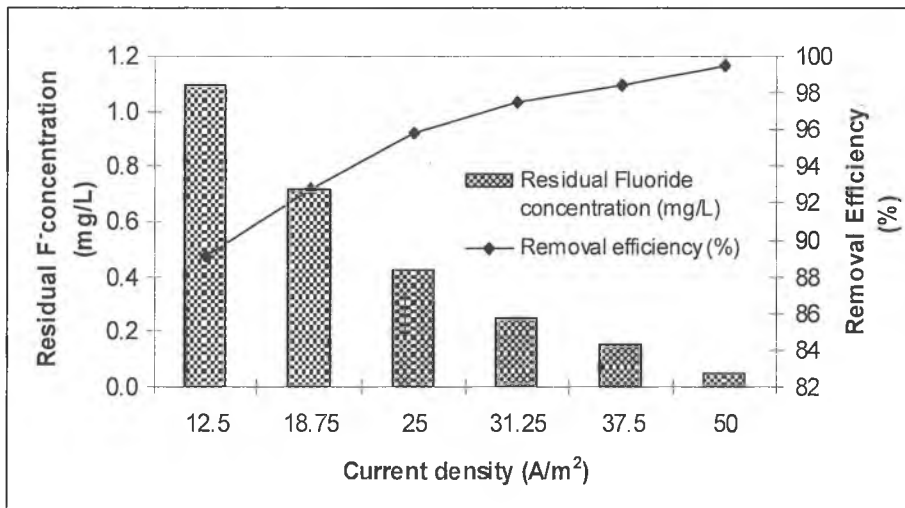


Figure 3: Effect of current density on the fluoride removal efficiency in a continuous flow electrocoagulation (Initial fluoride concentration = 10 mg/L, Flow rate= 150 mL/min, Initial pH =6, Conductivity=50 mS/m)

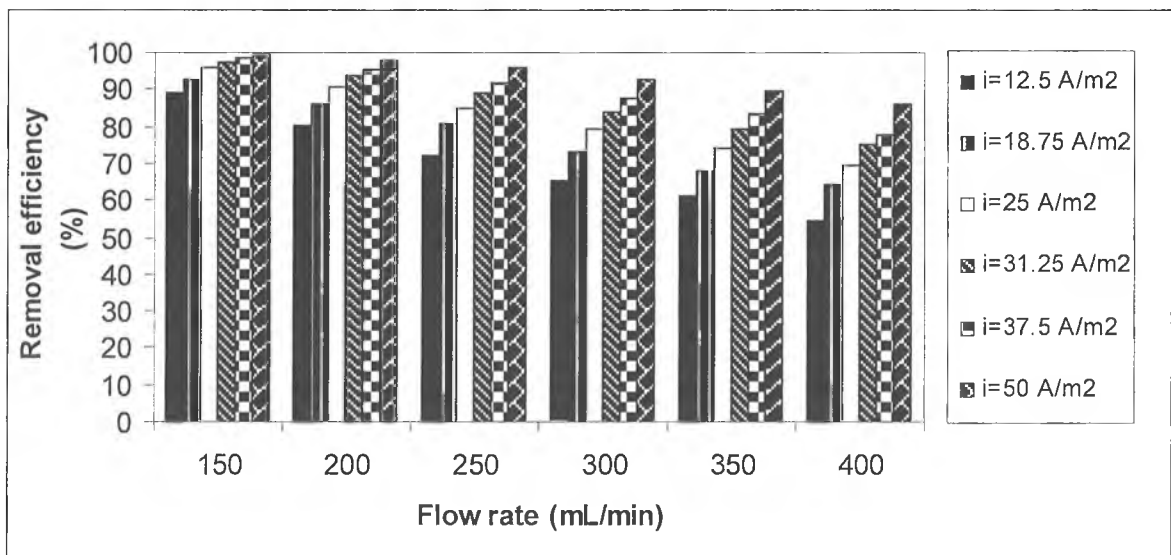


Figure 4: Effect of flow rate on the defluoridation efficiency at different current densities from 12.5 to 50 A/m<sup>2</sup> in a continuous flow electrocoagulation (Initial fluoride concentration = 10 mg/L, Initial pH =6, Conductivity =50 mS/m)

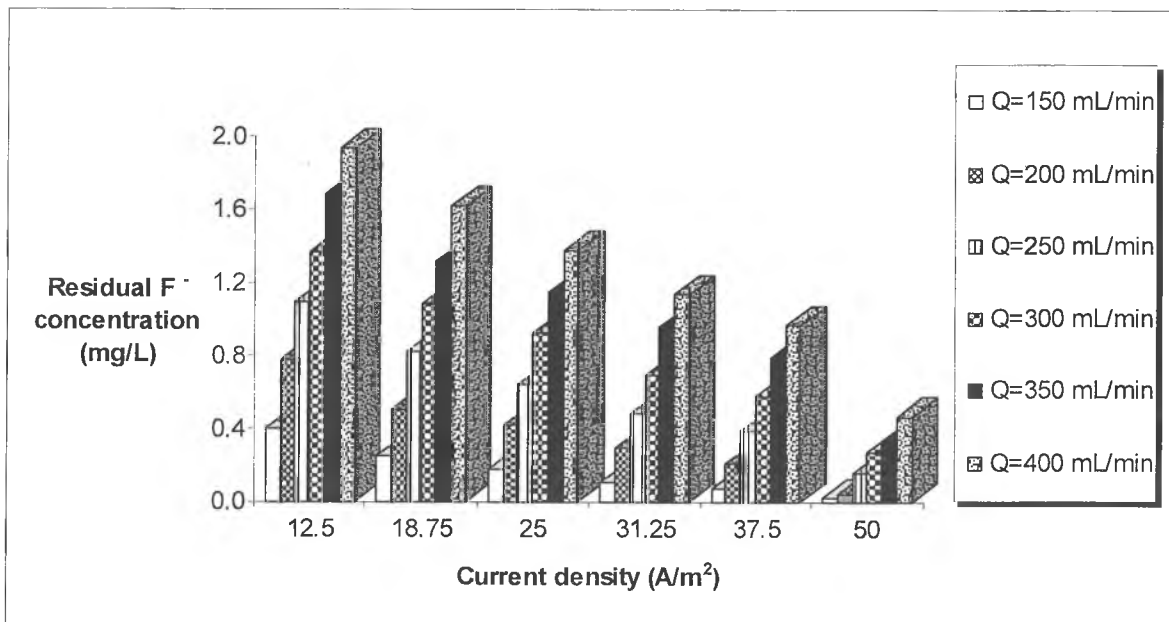


Figure 5: Effect of current density on the residual fluoride concentration at different flow rates in a continuous flow electrocoagulation (Initial fluoride concentration = 5 mg/L, Initial pH =6, Conductivity=50 mS/m)

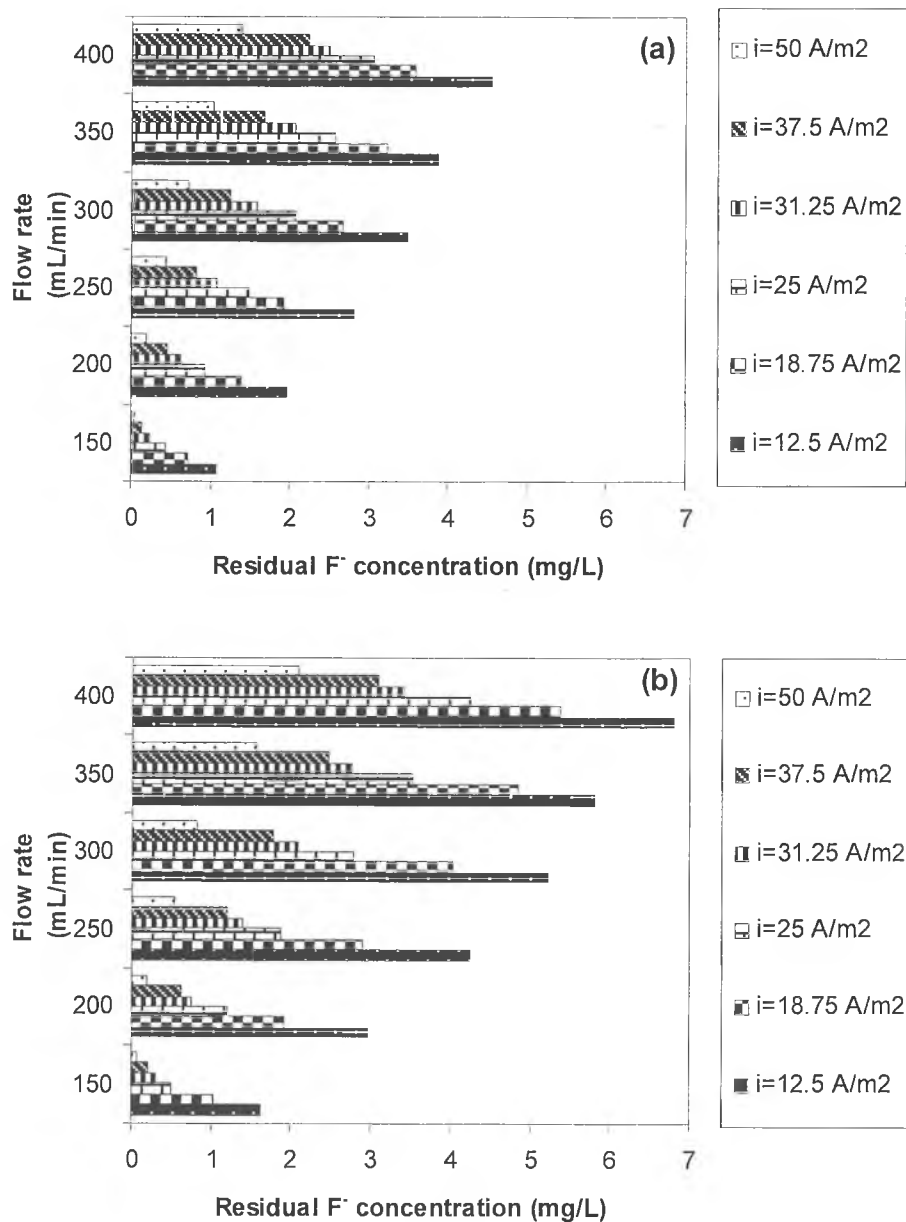


Figure 6: Effect of Initial fluoride concentration on the residual fluoride concentration at different current densities in a continuous flow electrocoagulation: [a] (Initial fluoride concentration = 10 mg/L) and [b] (Initial fluoride concentration = 15 mg/L), Initial pH = 6, Conductivity = 50 mS/m

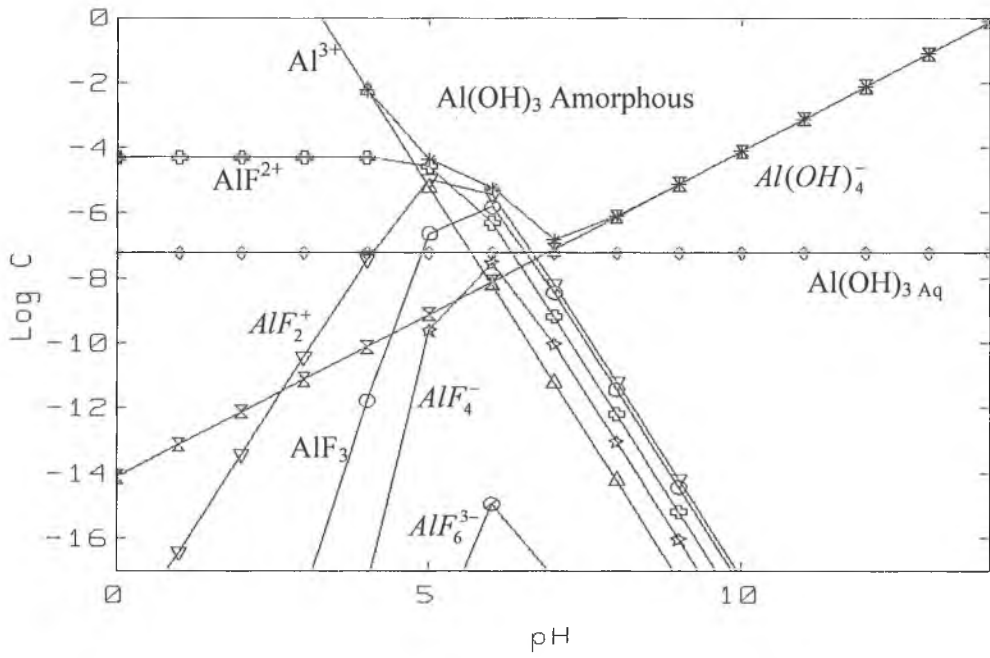


Figure 7: Solubility of aluminium hydroxide at various pH values (using MINEQL+ model)

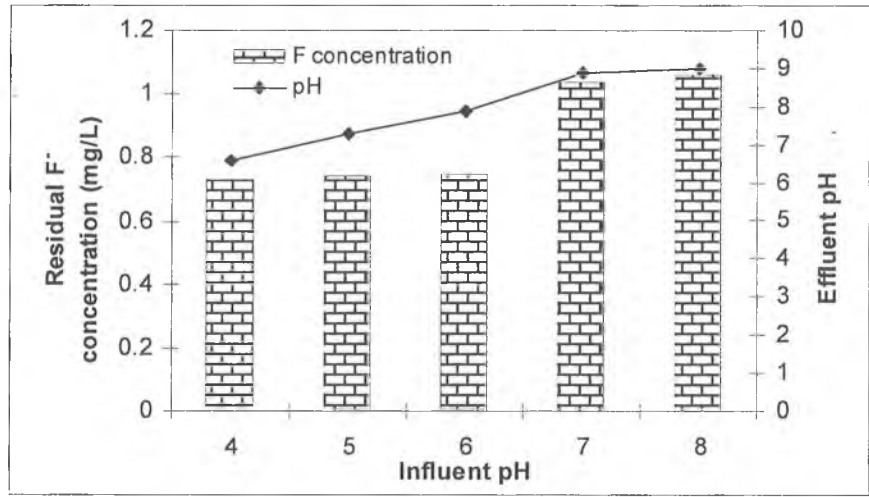


Figure 8: Effect of influent pH on the fluoride removal in a continuous flow electrocoagulation (Initial fluoride concentration = 10 mg/L, Current density = 18.75 A/m<sup>2</sup>, Flow rate= 150mL/min, Conductivity=50 mS/m)



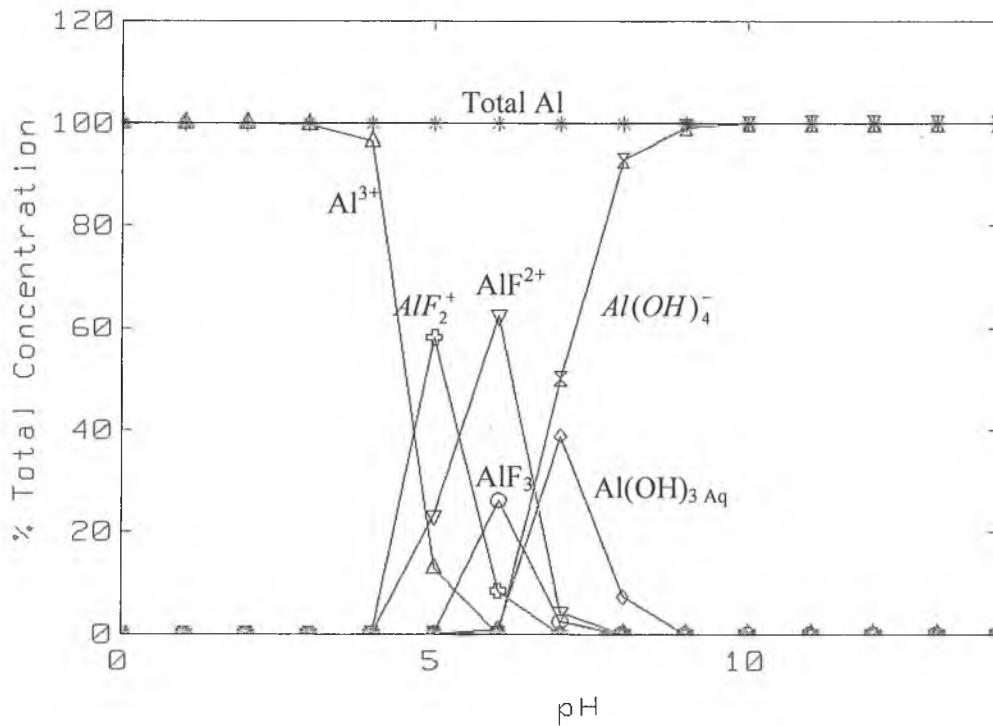


Figure 9: Composition of chemical species formed from  $1 \times 10^{-6} M$  aquo  $Al^{3+}$  ion;  $1 \times 10^{-5} M$  fluoride ion as a function of pH (using MINEQL<sup>®</sup> software)

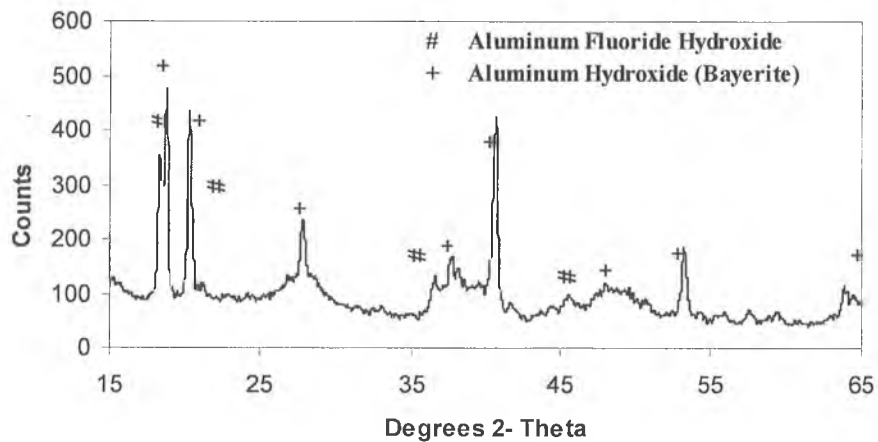


Figure 10: Composition of dried bottom sludge analyzed by XRD spectrum (Initial fluoride concentration = 10 mg/L, Current density = 18.75 A/m<sup>2</sup>, Flow rate = 150mL/min, Initial pH = 6, Final pH = 8, Conductivity = 50 mS/m)

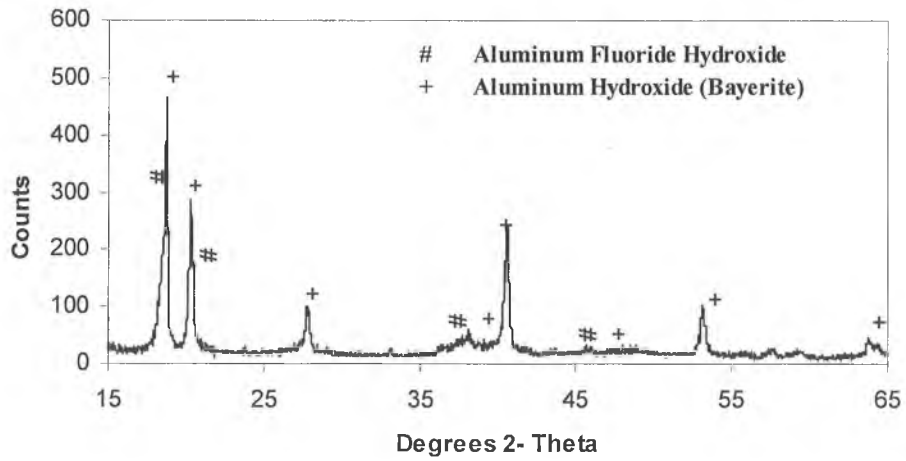


Figure 11: Composition of dried settled sludge analyzed by XRD spectrum (Initial fluoride concentration = 15 mg/L, Current density = 25 A/m<sup>2</sup>, Flow rate= 200mL/min, Initial pH =6, Final pH=8, Conductivity=50 mS/m)

Table 1: Health impacts from long-term use of high fluoride concentration in drinking water (WHO, 2005)

Fluoride concentration (mg/L)	Effects
0.8 - 1.2	Prevention of tooth decay, strengthening of skeleton
Above 1.5	Fluorosis: pitting of tooth enamel and deposits in bones
Above about 10	Crippling skeletal fluorosis

Table 2 : Optimum current densities at different flow rates (Initial fluoride concentration = 10 mg/L, Initial pH =6, Conductivity=50 mS/m)

Flow rate (mL/min)	Current density (A/m <sup>2</sup> )	Fluoride removal efficiency (%)
150	18.75	93
200	25	91
250	37.5	92
300	50	93

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