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Review

Review of pollutants removed by electrocoagulation and electrocoagulation/flotation processes

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ABSTRACT

The word "electrocoagulation'' (EC) will be sometimes used with "electroflotation'' (EF) and can be considered as the electrocoagulation/flotation (ECF) process. Through the process of electrolysis, coagulating agents such as metal hydroxides are produced. When aluminium electrodes are used, the aluminium dissolves at the anode and hydrogen gas is released at the cathode. The coagulating agent combines with the pollutants to form large size flocs. As the bubbles rise to the top of the tank they adhere to particles suspended in the water and float them to the surface. In fact, a conceptual framework of the overall ECF process is linked to coagulant generation, pollutant aggregation, and pollutant removal by flotation and settling when it has been applied efficiently to various water and wastewater treatment processes. This review paper considers a significant number of common applications of EC and ECF processes which have been published in journal and conference papers.

1. Introduction

Electrocoagulation (EC) and electrocoagulation/flotation (ECF) processes can be applied to a broad range of water and wastewater treatment systems and are most effective in removing inorganic contaminants and pathogens. Because of their broad applicability, they have been used for groundwater and surface water remediation at several sites (Joffe and Knieper, 2000). These processes are characterised by ease of operation, reduced production of sludge, and no need to handle chemicals. They have been applied efficiently to various water treatment problems. Therefore, if EC can replace conventional chemical coagulation, very little modification is required to make the present treatment plants more efficient and resolve the many problems caused by chemical coagulation (Rajeshwar and Ibanez, 1997). This research presents information pertaining to the removal pollutants by electrocoagulation in water and wastewater.

Overall, electrocoagulation is an electrochemical technique with many applications, in which a variety of unwanted dissolved particles and suspended matter can be effectively removed from an aqueous solution by electrolysis. The main aim of this review is to present bench and field scale research studies for the EC and ECF technology to remove different pollutants from water and wastewater treatment plants.

2. Theoretical background on ECF process

2.1. Definition of electrocoagulation (EC)

Electrocoagulation is the process of destabilising suspended, emulsified, or dissolved contaminants in an aqueous medium by introducing an electric current into the medium. In its simplest form, an electrocoagulation reactor may be made up of an electrolytic cell with one anode and one cathode. The conductive metal plates are commonly known as 'sacrificial electrodes' and may be made of the same or different materials (anode and cathode) (Mollah et al., 2001). Electrocoagulation is the electrochemical production of destabilisation agents (such as Al, Fe) that brings about neutralisation of electric charge for removing pollutant. Once charged, the particles bond together like small magnets to form a mass. This process has proven very effective in removing contaminants from water and is characterised by reduced sludge production, no requirement for chemical use, and ease of operation (Rajeshwar and Ibanez, 1997). Colloid – destabilising agents that effect on-charge neutralisation are produced by electrolysis in the EC process. For example, aluminium anodes are used to produce aluminium cations which have the same effect as the addition of Al-based coagulants in conventional treatment systems.

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2.2. *Definition of electrocoagulation/flotation (ECF)*

The electrocoagulation/flotation process provides an alternative technique for removing pollutants from water and wastewater. This process involves applying an electric current to sacrificial electrodes inside a reactor tank where the current generates a coagulating agent and gas bubbles. In addition, electrocoagulation/flotation is a technique involving the electrolytic addition of coagulating metal ions directly from sacrificial electrodes. These ions coagulate with pollutants in the water, similar to the addition of coagulating chemicals such as alum and ferric chloride, and allow for easier removal of the pollutants by sedimentation and flotation. Interactions occurring within an ECF reactor have been shown in Fig. 1. In a general sense, electrocoagulation or electroflotation are technologies based on the concepts of electrochemical cells, specifically known as "electrolytic cells". In an electrolytic process a source of direct current is connected to a pair of electrodes immersed in a liquid that serves as the electrolyte. To understand the electrochemical behaviour of the system, it is important to describe the general electrolytic reactions. The basis of electrocoagulation is the in situ formation of a coagulant species that can remove various pollutants from the water and wastewater being treated. There are three main mechanisms in the whole of electrocoagulation/flotation process:

Electrode oxidation
Gas bubble generation
Flotation and sedimentation of flocs formed

The electrode connections in an electrocoagulation reactor are monopolar and bipolar. A simple arrangement of the electrode connections is shown in Fig. 2, where the electrodes are monopolar and bipolar connections in the electrocoagulation reactor. Cell voltage and current are measured as digital and need to be controlled in all these experiments. With monopolar connections an electric potential is connected between *n* pairs of anodes and cathodes (Mollah et al., 2004a; Jiang et al., 2002a). Parallel connections to each electrode cause current (I_0) to pass across each electrode and solution but if an electrical potential (U_0) is applied between two feeder electrodes, a series connections to bipolar electrodes cause the same current to pass through "*n*" electrode pairs.

3. Discussions on application of EC and ECF processes

EC and ECF processes are electrochemical techniques with many applications when each application has been considered separately in this section.

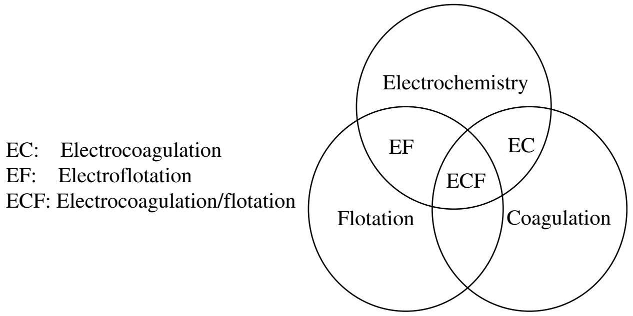


Fig. 1. Interactions occurring within an ECF process.

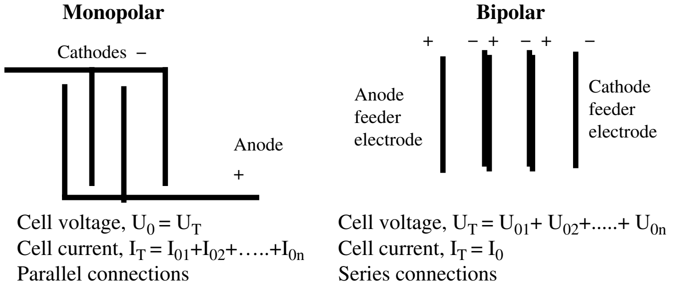


Fig. 2. Monopolar and bipolar electrode connections in the EC reactor (adapted from Jiang et al., 2002a).

3.1. *Oily wastewater treatment*

Electrocoagulation/flotation process to treat refinery wastewaters and remove emulsified oils was studied by Kaliniichuk et al. (1976). Their results showed that wastewater treated with aluminium hydroxide formed by dissolution of aluminium anodes when hydrogen evolved at the cathode floats, the hydroxide floc adsorbed the oil. An electrolytic process to treat oily wastewater was considered by Weintraub et al. (1983). A pilot plant with iron electrodes was designed to treat between 300 and 7000 mg L^{-1} of emulsified oil at flow rate of 4 L min^{-1}. The results illustrated that ferric ion destabilises the emulsion by neutralising the charge on the droplets and then precipitates as ferric hydroxide, while the oil is adsorbed onto the flocculent precipitate. It was found that the effluent oil could be reduced to 10 mg L^{-1} or less when the dissolution rate of iron was increased in this process.

An electroflocculation-electroflotation system was used by Balmer and Foulds (1986) to separate oil from oil-in-water emulsions. They described how a particularly effective separation could be achieved by electrochemical dissolution of a sacrificial iron anode. The main conclusions of their investigation are summarised as follows:

- Electroflocculation can be combined with electroflotation to separate dispersed oil from aqueous emulsions.
- The production rate of a generated reagent is controlled by the electrical current rate.
- Several different anode materials are effective for electrocoagulation.
- The electrical energy consumption is optimised by using high conductivity electrolytes (i.e. high salt content) with small electrode spacing in a low current.
- The consumption rate of the sacrificial anode depends on the current applied.

Ibanez et al. (1995) also described a simple laboratory experiment based on an electrolytic process for treating oily wastewater. The results showed that an oil layer is formed at the top of the solution after a few minutes of electrolysis. The solution became clear at the bottom after almost half an hour and the top layer of oil can be skimmed off. Free oil can generally be removed from water by skimming.

Using chemical coagulation with ferric chloride and lime is one of the most successful methods for destabilising oil water emulsions. An attempt was made by Ogutveren and Koparal (1997) to remove oil from synthetic and oily wastes through an electrochemical method using iron or aluminium electrodes. These experiments were performed with initial oil concentrations of 50, 200, and 500 mg L^{-1}. The effects of applied potential, initial oil concentration, and supporting electrolyte concentration on the rate of efficient removal of oil have been investigated. The experimental results showed that aluminium electrodes are more efficient than iron electrodes. It may be because of high adsorption capacity of hydrous aluminium oxides.

A combined process of electrocoagulation and electroflotation in a continuous flow reactor was used by Chen et al. (2000) to treat wastewater from restaurants in Hong Kong with a high oil and grease content. Here, electrocoagulation destabilised and aggregated the fine particles while electroflotation was responsible for floating the flocs produced in the electrocoagulation unit. The effects of operational variables on the system, including: detention time, influent pH, conductivity, sludge production, characterisation of restaurant wastewater, energy consumption, and electrocoagulation and electroflotation charge loading ($C_L = (I \times t)/(F \times V)$), were investigated, where I is Total current (A), V is Volume of cell (m^3), t is Time (min), F is Faraday's constant ($F = 96\,500\ C\,mol^{-1}$).

The results showed that the electrocoagulation/electroflotation system was a feasible process for this treatment. Electrical conductivity had no significant effect on the efficient removal of pollutant when the water product pH was kept constant between 6 and 7. The loading charge and detention time were the most important operational variables for efficient treatment in either the electrocoagulation or electroflotation process.

The results on the investigation of the electrocoagulation process for treatment of oily wastewater indicated that using an EC reactor successfully destabilises oil-in-water emulsions. Dispersed gas bubbles formed from electrolysis are extremely fine and uniform. The effect of the most important operating parameters such as current inputs, electrolysis time, and initial oil concentration were investigated upon the performance of the EC and EF processes. Several different anode materials are effective for electrocoagulation. However, the results showed that aluminium electrodes have better treatment performance than iron electrodes.

3.2. Dye and textile wastewater treatment

Decolourization of dye solution by electrocoagulation has become an attractive method in recent years. A bipolar packed bed electrochemical reactor consisting of soluble electrodes was used in the batch mode to remove dye by Ogutveren et al. (1992). The effects of pH, current rate, electrolysis time, initial concentration of dye, and applied potential on the removal rate of dye have been investigated. Dye removal rate of 100% was achieved in a very short time (3–5 min) while the maximum power consumption was found to be 2.24 kWh m^{-3}.

An electrocoagulation/flotation process was developed to remove colour from wastewater by Ibanez et al. (1998). The gas bubbles can carry the pollutant to the top of the solution where it can be more easily concentrated, collected, and removed. The metallic ions can be reacted with the OH^- ions which are produced at the cathode. Insoluble hydroxides adsorb pollutants and are then removed by precipitation and flotation.

Decolourisation of a strongly coloured solution containing reactive textile dyes using the electrocoagulation process was investigated by Gurses et al. (2002). The 4 × 4 cm iron and aluminium plates, and cylindrical graphite, were applied in the electrocoagulator. Cell voltage and current rate were digitally measured. The experimental results showed that effective parameters on the decolourisation process were found to be cell voltage, electrolysis time, and current density. They also reported that several interactive mechanisms are possible between the dye molecules and hydrolysis products when the rate of these interactions depends on the pH of the solution. Two main mechanisms were being considered in the research: precipitation and adsorption. Each was being proposed for a separate pH range, as follows:

Precipitation:

$$\text{DYE} + \text{Monomeric} \rightarrow \text{Al}\ [\text{DYE Monomeric Al}]_{(s)} \quad \text{pH} = 4\text{–}5$$

$$\text{DYE} + \text{Polymeric} \rightarrow \text{Al}\ [\text{DYE polymeric Al}]_{(s)} \quad \text{pH} = 5\text{–}6$$

Adsorption:

$$\text{DYE} + \text{Al(OH)}_{3(S)} \rightarrow [\text{particle}] \quad \text{pH} > 6.5$$

$$[\text{DYE polymeric Al}]_{(s)} + \text{Al(OH)}_{3(S)} \rightarrow [\text{particle}] \quad \text{pH} > 6.5$$

Under experimental conditions, the negatively charged dye molecules were neutralised. The positive charge of the aggregates may be attributed to the adsorption of dye monomeric and dye polymeric colloidal particles on the $Al(OH)_3$ precipitates.

An electrocoagulation process for the decolourisation of reactive dyes was developed in Korea (Kim et al., 2002). They investigated the effects of different operating parameters including current density, number of electrodes, electrolyte concentration, electrode gap, dye concentration, pH of solution, and the inlet flow rate on decolourisation using the continuous electrocoagulation process. However, the removal mechanism has not been considered in the study by the authors. The results showed that removing dye using Al electrodes was more efficient than Fe electrodes. It is because of low absorption capacity of ferrous ions which is caused by the iron sacrificial anode. Also, the power consumption for electrocoagulation increased proportionally when the current density, electrode gap, and concentration of electrolyte in the solution were increased. The dye removal efficiency was reported to be between 60 and 98.5%.

An electrocoagulation technology using aluminium electrodes has been proposed by Can et al. (2003) for decolourising reactive dye solution. The removal of a reactive textile dye (Remazol Red RB 133) to investigate the effects of wastewater conductivity, initial pH, current density, dye concentration, treatment time, and energy consumption in batch electrocoagulation reactor was carried out. The results showed that decolourisation decreased steadily when the conductivity in the solution was increased. The experimental results showed that the colour removal decreased when the initial pH increased from 5 to 9. Decolourisation decreased from 90 to 70% when the initial concentration of dye was increased from 100 to 1000 mg L^{-1}. This means that removal is a function of the initial concentration of dye. Aluminium hydroxy-polymeric species remove dye molecules by precipitation and flotation. The results showed that electrocoagulation is an efficient process for removing dye from wastewater.

In 2004, the use of batch and continuous electrocoagulation reactors were investigated for decolourising dye solution (Daneshvar et al., 2004; Mollah et al., 2004a; Fan et al., 2004; Den and Huang, 2005). The researchers focused on the technical performance of this process while the effects of various parameters including wastewater conductivity, pH solution, current density, and operating time have been discussed. Daneshvar et al. (2004) showed that more colour was removed in the EC cell with monopolar electrode connections than those with bipolar electrode connections. However, there was missing information for its reason in this paper. To increase the removal of dye the optimum conditions for operating the EC process were experimentally determined by Mollah et al. (2004b), ever since an EC process using sacrificial iron electrodes was used to treat a solution of orange II dye. It was found that 98.5% of dye was removed under parameters including, current value of 4 A, voltage value of 42 V, initial pH of 7.3, conductivity of 381 mS m^{-1}, flow rate of 350 mL min^{-1}, and an initial 10 mg L^{-1} concentration of dye.

Dyeing is the most important process usually applied in almost all textile-manufacturing industries. In the developed countries these wastewaters are treated by traditional methods like, physical and chemical processes. The biological methods are cheap and simple to apply, but can not be applied to most textile wastewaters because most commercial dyes are toxic to the organisms used in the process. The electrocoagulation technology is considered to be potentially an effective tool for treatment of textile wastewaters with high removal efficiency. The removal efficiency was found to be dependent on the initial pH, the dye concentration, the applied current density, the electrolysis time in batch model, and the flow rate in continuous flow reactor. It was found that maximum 98.5% of the dye was removed from the solution by EC technology.

3.3. Organic removal

3.3.1. Recovery of phenolic compounds

In Thailand two aluminium plates placed inside in an electrocoagulation reactor acting as reactors were applied to recover phenolic compounds by Phutdhawong et al. (2000). Sodium chloride (2 g) was added as an electrolyte and current (0.5 A, 22 V) from a DC power supplier was passed through the solution. It was previously proposed that main reactions produce aluminium ions at the sacrificial anode, and hydroxide ions and hydrogen at the cathode. The aluminium ion liberated from the anode may also interact directly with an anion or a phenol which then precipitates out of the solution in the form of an insoluble salt, e.g. aluminium triphenolate [$Al(OAr)_3$]. The experimental results showed that this method can also be applied to some phenolic compounds and can eliminate unwanted particles or chemical species from an aqueous solution.

3.3.2. Municipal wastewater treatment

In France an electrocoagulation and flotation system was joined to study the membrane process (microfiltration) on the flux of municipal wastewater permeates (Pouet and Persin, 1992; Pouet and Grasmick, 1995). The experiments were conducted in a continuous mode with a 71 L electrolytic cell and 15 aluminium electrodes, as is shown schematically in Fig. 3. The experimental results for removal of COD, SS, and turbidity from the municipal wastewater permeate using the hybrid process has been studied (results not shown). This study showed that a combination (hybrid process) of an electrocoagulation/flotation system with microfiltration could increase the removal efficiency. As seen, due to combination of electrocoagulation, flotation and microfiltration, it is possible to increase by more than 30% the turbidity removal yield, by more than 20% that of the COD and by more than 65% that of the SS in comparison with electrocoagulation alone. So the association of these three processes seems to be a good alternative to an extensive treatment because of its effectiveness.

Traditional methods for treating of laundry wastewater consist of various combinations of biological, physical and chemical methods. Due to the large variability of the composition of this kind of wastewater, most of these traditional methods are becoming inadequate for the simultaneous removal of high content of pollutants. A new bipolar EC and EF process was developed to treat laundry wastewater (Ge et al., 2004). The operating parameters including initial pH, hydraulic residence time (HRT), and current density were investigated. The laboratory results showed that the COD removal was greater than 70%. The removal of phosphate, surfactant, and turbidity could be above 90% in a wide pH range of 5–9.

These studies showed that the ECF technique can be effectively used for organic removal from wastewater. The pollutant removal processes were reported to be to neutralize pollutant charge, generate ultra-fine bubbles and separate the coagulated flocs from water or wastewater.

3.3.3. Industrial wastewater treatment

An electrocoagulation process was conducted for treatment of different soluble oily wastes with very high COD by Calvo et al. (2003). Electrocoagulation tests were conducted initially in a continuously operated laboratory scale unit to reduce COD from 70 to 90%. For different concentrations of soluble oils, removing of the COD was between 49.5 and 77.8% when the current rate was a constant 7 A. However, the removal of COD increased to 90% when the current rate was increased to 10 A. As seen, the removal efficiency is increased when the current input increases. The capacity and efficiency of an electrocoagulation process depends on the nature of the liquid waste.

The removal of COD, turbidity, phenol, hydrocarbon, and grease from petrochemical wastewater was studied by Dimoglo et al. (2004) using the electrocoagulation (EC) and electroflotation (EF) process. Two laboratory scale reactors were used for the EF, EC, and EF processes, as shown schematically in Fig. 4(a and b).

Iron and aluminium electrodes were used in the EC unit because a graphite anode and stainless steel electrode were used in the EF unit. The experimental results showed that using Al and Fe electrodes in EC process effectively treats wastewater. A comparison between Al and Fe electrodes at different electrolysis times was used in the EC process to remove pollutants, and the results are shown in Fig. 5(a and b). The results showed that more phenol and hydrocarbons were removed with Al electrodes and more turbidity and grease was removed with Fe electrodes. It may be because of low adsorption capacity of hydrous ferric oxides. There is no significant different between Al and Fe electrodes for COD removal.

Treatment of chemical mechanical polishing (CMP) wastewater from a semiconductor plant by electrocoagulation was investigated by Lai and Lin (2004) and Belongia et al. (1999). The experimental results under different operating conditions showed that the rate aluminium dissolved into the aqueous solution was calculated as 10.4 mg min^{-1} when the potential and electrolysis time respectively were found to be 20 V and 30 min in the electrocoagulator (Lai and Lin, 2004). The experimental results showed that electrocoagulation of the CMP wastewater with an Al/Fe electrode removed 96.5% of turbidity and 88.7% of COD in less than 30 min of electrolysis time in the electrocoagulator (Belongia et al., 1999).

The research studies showed that electrocoagulation is an efficient technology for treatment of industrial wastewaters. However, the capacity and efficiency of an electrocoagulation process depends on the nature of the liquid waste and initial pollutant concentrations.

3.4. Heavy metals removal

Removing arsenic from industrial effluent using an EC batch reactor was investigated by Balasubramanian and Madhavan (2001). It was found that the rate of removal depends on the different operational parameters including initial concentration of arsenic, current density, the influence of pH, and electrolysis time. The generation of ferric ions can easily be controlled during electrocoagulation by adjusting the operating conditions. The results showed that arsenic can be removed through electrocoagulation. However, the arsenic removal mechanism was not considered by the authors in the study.

Studies on removing arsenic by the EC process indicated that hydrous metal oxides such as ferric or aluminium hydroxides

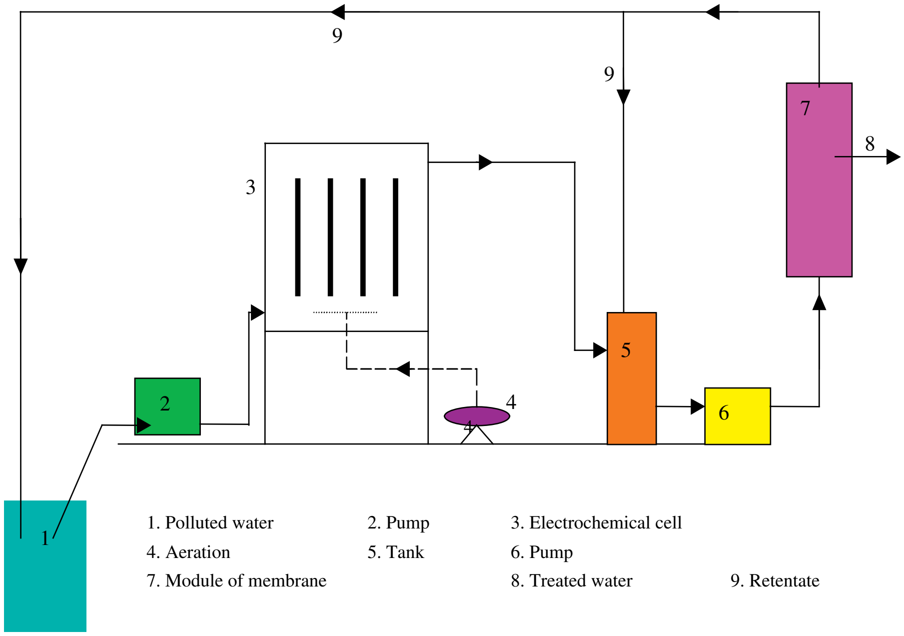
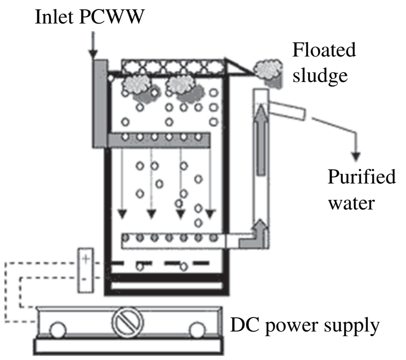


Fig. 3. Flow diagram of a hybrid process including electrocoagulation, flotation by aeration, and microfiltration processes for the municipal wastewater permeates removal (adapted from Pouet and Grasmick, 1995).

adsorb arsenic strongly. Laboratory scale experiments to remove arsenic by the EC process were conducted with three types of electrodes, namely iron, aluminium, and titanium by Kumar et al. (2004). The highest removal of arsenic (99%) was obtained by using iron electrodes at a pH range of 6–8. It may be because of high adsorption capacity of hydrous ferric oxides for arsenic removal. It was noted that As (III) removal mechanism in EC process seems to be oxidation of As (III) to As (V) and subsequence adsorption on to hydrous ferric oxides.

a Electro-flotation Cell



b EC and EF cells

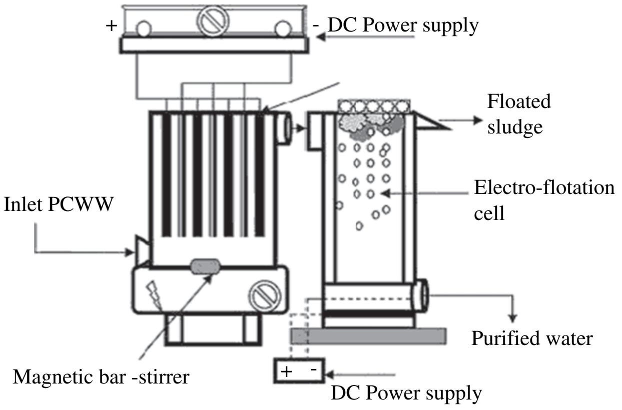


Fig. 4. Laboratory scale processes: (a) EF process, (b) EC and EF processes (adapted from Dimoglo et al., 2004).

A comparison between a bench scale electroflotation (EF) system and dissolved air flotation (DAF) from soil washing water was undertaken by Park et al. (2002) to remove cadmium ions. The results showed that much more cadmium (100%) was removed by electroflotation using aluminium electrodes. The results also showed an unsatisfactory removal of cadmium removal by the DAF process. It can be concluded that electroflotation was considered to be an effective technology to treat cadmium from contaminated soil washing water.

The results on the investigation of the electrocoagulation process for treatment of heavy metals indicated that using an EC reactor successfully removes arsenic and cadmium from water or wastewater. The highest removal of arsenic was obtained by using iron electrodes when the removal mechanism was considered to be adsorption of arsenic on hydrous ferric oxides. However, no evidence was reported to support the mechanism.

3.5. *Potable water treatment by EC*

Coagulation using chemical coagulants is one of the most essential processes in the conventional treatment of drinking water. However chemical coagulation (CC) has some inherent problems in cost, maintenance, and sludge production. Thus electrocoagulation (EC) has recently been suggested as an alternative to conventional coagulation. Several reports have been published in scientific journals on electrocoagulation combined with electroflotation for treating potable water (Nikolaev et al., 1983; Musquere and Ellingsen, 1983; Vik et al., 1984; Mills, 2000; Ciorba et al., 2002; Pulido et al., 2001; Jiang et al., 2002b; Holt et al., 2002; Han and Kwon, 2002; Abuzaid et al., 2002).

An electrocoagulation process for potable water treatment was studied in USA by Vik et al. (1984). Some laboratory experiments including a DC power supply (6–12 V), resistance box to regulate current intensity, and a multi-meter to read the values were

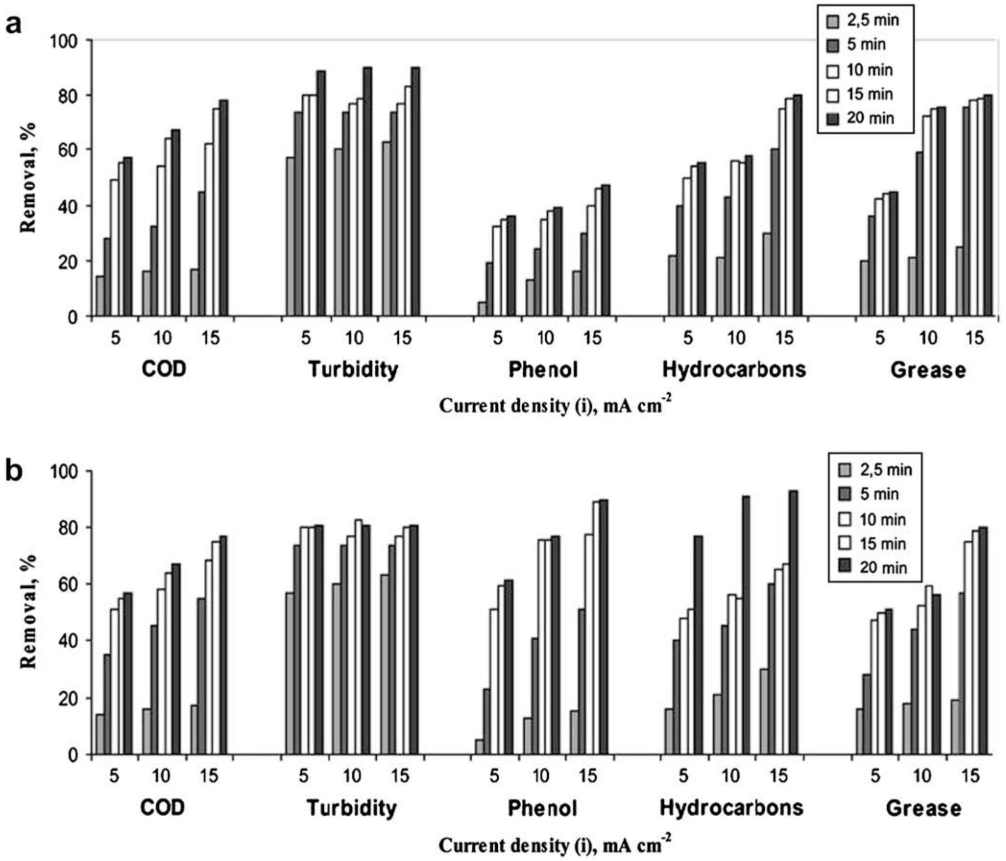


Fig. 5. Summary of EC performance for removal of the pollutants in different current densities and various electrolysis times [a] Fe electrode, [b] Al electrode (quoted in Dimoglo et al., 2004).

performed. In the electrochemical cell four plate aluminium anodes and cathodes (dimension $14 \times 20 \times 0.25$ cm) were used as electrodes, at a flow rate of 0.171 L min^{-1}. This process was compared with a conventional water treatment process and was an effective process for use in small water treatment plants because of the following:

- The amount of chemicals having to be transported to the solution is lower than for chemical treatment (approx. 1/10 of the amount).
- The electrocoagulator unit will be made with enough electrodes for at least one year of treatment (long lifetime).
- The maintenance and operation of the EC system will be simple.
- A lesser amount of sludge is formed in an EC system.

An innovative electrocoagulation process has been shown to affectively treat raw water sources to produce potable water by Mills (2000). In this process coagulant formation and particle destabilisation occurred through electrolytic cells in the turbulent flow in which iron or ferric hydroxide was continuously produced. Aggregation occurred in the flocculation stage with the formation of large particles that could readily be separated from the water. The experimental results on the capability of the EC process for removing heavy metals from contaminated streams, leachate from mining, and land fill sites showed that the molybdenum was reduced from 9.95 to 0.006 mg L^{-1} in leachate at a mining site, and iron was reduced from 130 to 0.015 mg L^{-1} in leachate from a land fill site (Mills, 2000).

An electrocoagulation/flotation process has been developed by Jiang et al. (2002a) in the UK for treating water. An electrocoagulator reactor with a 500 W DC power supply, a peristaltic pump, a flow meter, a reservoir for raw water and an outlet reservoir, a separation/flotation tank, and plate aluminium (maximum Al 97%, minimum Al 95%) anodes and cathodes was used in this research. The electrocoagulator operated under the following conditions:

- Water flow rate of 0.01 $\text{m}^3\,\text{h}^{-1}$
- Two electrode arrangements, bipolar electrodes connected in series via the water and monopolar electrodes connected in parallel.
- The quality of the treated water was measured in terms of dissolved organic carbon (DOC), ultra-violet absorbance at 420 nm, pH and conductivity.
- Operating electrical current densities of 3–25 A m^{-2}, which produced Al (III) concentrations of 2.4–14 g m^{-3}, determined as the sum of the concentrations in the treated effluent and floc sludge.
- pH values of 6.5 and 7.8 ± 0.2

Electrocoagulation experiments were performed by two laboratory arrangements, up-flow electrocoagulator configuration and horizontal flow configuration, as shown schematically in Fig. 6 (a and b).

Regarding the specific electrical energy consumed using monopolar and bipolar connections (different electrode arrangements); the experimental results showed that the energy consumed by electrocoagulation was very low. For example at current densities of 10–20 A m^{-2} it was 20 and 20–30 kWh $(\text{kg Al})^{-1}$ for the monopolar and bipolar electrode arrangements. Over wider current densities of 10–60 A m^{-2} the electrical energy consumed was

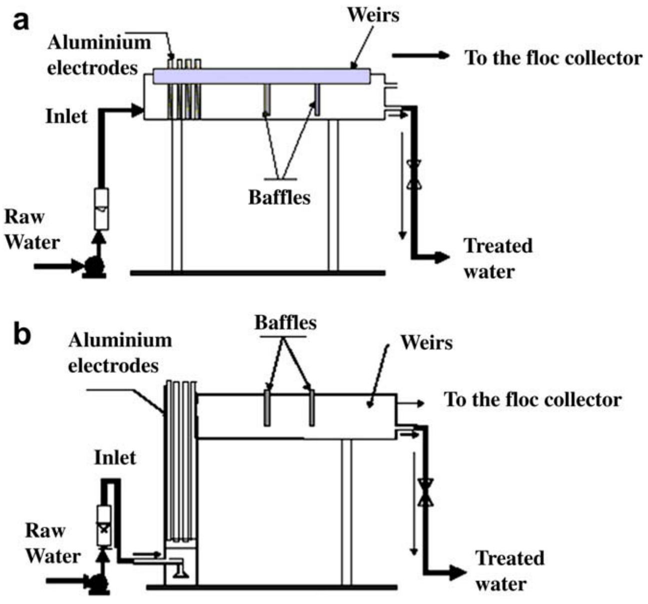


Fig. 6. Schematics of EC process [a] horizontal-flow electrocoagulation reactor and [b] Up-flow electrocoagulation reactor (adapted from Jiang et al., 2002a).

20–80 kWh $(\mathrm{kg\,Al})^{-1}$. The results showed that an up-flow electrocoagulator configuration performed better than a horizontal flow configuration when both bipolar and monopolar electrode arrangements were used. In an up-flow reactor, an average of 5–8% more DOC, colour, and UV 254 removal were achieved than in a horizontal flow reactor. The up-flow reactor arrangement allows more collisions to occur between the gas bubbles and the resulting Al-contaminant flocs which enhance the overall flotation performance.

A laboratory batch electrocoagulation reactor on removing turbidity was performed by Han and Kwon (2002) to compare with conventional chemical coagulation (CC). The experimental results showed that current efficiency (ε_C), which is defined as a fraction of the current passing through an electrolytic cell, was more than 100% for the batch EC system. Based on the initial pH of removing turbidity in the electrocoagulation process, the experimental results showed that the highest efficiency was obtained between pH 6 and 8 during experiments. It may be because of minimum solubility of aluminium hydroxide in this pH ranges. It is clear that most of the particles are negatively charged in water while similar charges prevent aggregation through electrostatic repulsion. Thus, the characteristics of particles and zeta potential, which represents the electric condition of particles, are important factors in the EC operating process. It was recommended that the particle charge should be maintained in the range of -10 mV to $+10$ mV for providing proper conditions for coagulation (Han and Kwon, 2002). For the same removal a lower dosage of aluminium was needed for the EC process than the CC process. In addition the EC process was less sensitive to pH than the CC process. The experimental results showed that the EC process was more efficient than the CC process at removing turbidity.

A relationship between electrolysis voltage and other operational parameters on the electrocoagulation process was analysed by Chen et al. (2000). The electrolysis voltage is strongly dependent on the current density, the conductivity of water and wastewater, the interelectrode distance, and the state of the electrode surface. One of the objectives was to find theoretical models for the voltage of electrolysis required in the electrocoagulation process. Regarding any theoretical analysis, when current passes through an electro-chemical reactor it must overcome the potential equilibrium difference, anode over-potential, cathode over-potential, and ohmic potential drop of solution (Scott, 1995). The total electrolysis voltage (U_T) of the monopolar connection was the same as the electrolysis voltage between electrodes, as follows:

$$U_T = U_0 \tag{1}$$

The total electrolysis voltage for the bipolar connection can be expressed as follows:

$$U_T = (N-1)U_0 \tag{2}$$

where N is the number of electrodes. A quantitative comparison between chemical dosing and electrocoagulation was investigated by Holt et al. (2002). Chemical coagulation was evaluated using an adaptation of the standard jar testing technique, with aluminium sulphate as the chemical coagulant. Sodium hydroxide and hydrochloric acid solution were added for final pH adjustment. The pollutant used was “potter clay” comprising kaolinite (67%), quartz (25%), illite/micas (3%), feldspar (3%), and other trace elements (2%), as characterised by X-ray diffraction analysis. A batch electrocoagulation reactor was conducted for a quantitative comparison on experiments such as turbidity, zeta potential, and pH solution. Four aluminium anodes with five stainless steel cathodes were used in the electrocoagulator. Current over a 0.25–2 A range was investigated when it held constant for each run. The results showed that the mechanism for delivering coagulant was a key difference between chemical dosing and electrocoagulation performance. In chemical coagulation, coagulant addition is a discrete (shot-fed) event with equilibrium determining aluminium speciation and pH. By contrast, in electrocoagulation the continuous addition of aluminium and hydroxyl ions results in a non-equilibrium state and is a function of current and time (Holt et al., 2002). Another search by the same authors (Holt et al., 2005) showed that three operating stages (lag, reactive and stable) were identified in a batch electrocoagulation reactor. Little or no turbidity change is observed in the lag stage with the majority (95%) of turbidity removal occurring during the reactive stage. All experiments had an initial lag stage in their turbidity response, before a rapid decrease. At the reactive stage, the greatest reduction in turbidity occurred which showed aggregation by an adsorption mechanism. Continued precipitation of aluminium hydroxide during the stable stage, and a decrease in turbidity, indicated a sweep coagulation mechanism. In this stage the highest current (2 A) reduced the pollutant level in the short term.

A laboratory batch electrocoagulation reactor was designed and constructed in Australia by Sivakumar et al. (2004) to remove turbidity. This research was directed towards comparing the efficient removal of turbidity and colour, the size of flocs formed, and volume of sludge generated by the electrocoagulation (EC) and chemical coagulation (CC) processes. The operational parameters, including pH, electrolysis time, and total concentration of aluminium were investigated in this research. In the electrochemical cell, five plate aluminium (purity of Al 95–97% Ullrich Aluminium Company Ltd, Sydney) anodes and cathodes (dimensions 250 × 100 × 3 mm) were used as electrodes. The electrodes were connected in a monopolar configuration, as shown schematically in Fig. 7. The experimental results showed that the concentration of aluminium required in EC process was much less than the CC process to achieve the same performance. When turbidity could be removed at a rate of 87% the concentration of aluminium needed by the EC process was only 2.2 $\mathrm{mg\,L^{-1}}$ but around 6 $\mathrm{mg\,L^{-1}}$ was needed in CC process. The results obtained also showed that floc formed by the EC process was larger than by

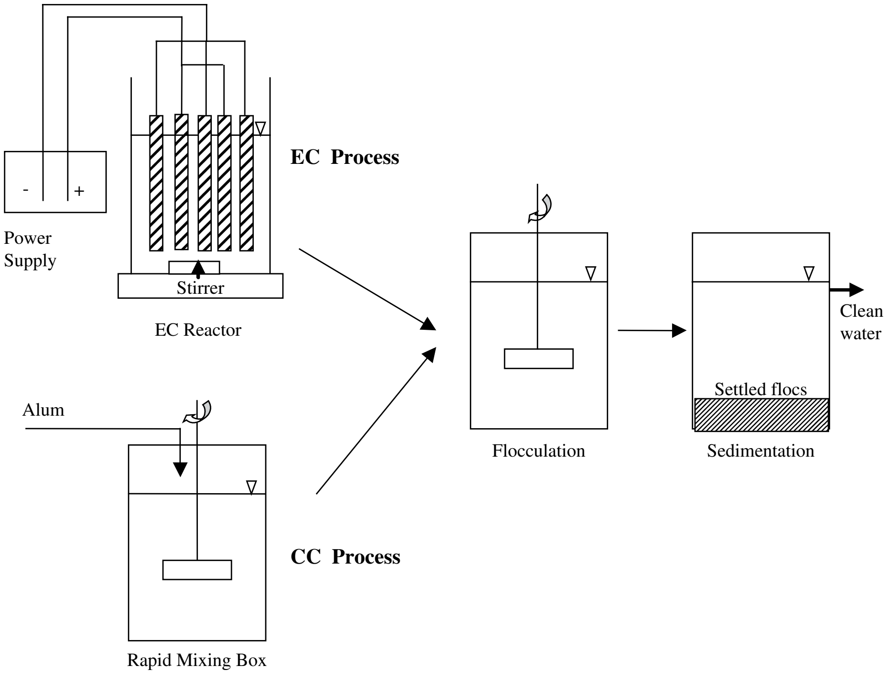


Fig. 7. Experimental set up for EC and CC processes (after Sivakumar et al., 2004).

the CC process. It was reported that more turbidity was removed and much better treatment could be achieved in the EC process. The volumes of sludge produced on the EC process were much less than the CC process at two detention times of 90 and 180 min. At an operating value of 1 A and 1 min electrolysis time, the volumes of sludge were 2.6 and 3.8 mL L^{-1} in EC process when the detention time increased from 90 to 180 min in the sedimentation tank. The volume of sludge in the sedimentation tank increased from 8.7 to 10.6 mL L^{-1} in the CC process when the detention time increased from 90 to 180 min (results not shown). Because more sludge is settled when the detention time increases in the sedimentation tank.

The research studies showed that electrocoagulation process is more efficient than coagulation process for potable water treatment. Less aluminium dosage was needed to get the same removal efficiency in EC process, which is related to chemical cost and sludge production.

3.6. Nitrate, nitrite and ammonia removal

The electrochemical method was used by Lin and Wu (1996) to investigate nitrite and ammonia removal from aqueous solution. The results showed that removal was improved when electrolysis time in the cell was extended. The pH effect on nitrite removal was less significant than conductivity and current input. The iron electrode was unsuitable for removing nitrite due to its low removal efficiency. However insoluble electrodes such as a graphite anode and titanium dioxide cathode were appropriate for removing nitrite and ammonia by electrochemical cells. It was thought there are two possible mechanisms involved in the process, electroflotation and electro-oxidation. More investigations need to be done.

Electrochemical reduction of nitrate ions was studied in synthetic solutions after ion exchange column regeneration by Paidar et al. (1999). A copper cathode could remove nitrate ions from the solution. At a current density of 38 A m^{-2}, the concentration of nitrate was reduced from 600 to 50 mg L^{-1}. The mechanism of denitrification was related to an electro-reduction process, as nitrate ions were reduced to nitrite, ammonia, and nitrogen gas. However, no evidence was noted to present the removal mechanism in the paper.

A comparison of electrocoagulation (EC) and electro-reduction for removing nitrate from water was investigated by Koparal and Ogutveren (2002). A batch reactor (160 cm^3) consisting of a graphite rod anode and carbon cathode was used for electro-reduction. Electrocoagulation experiments have been also carried out by a bipolar packed bed reactor filled with iron rings. The optimum pH was between 9 and 11. The reduction of nitrate to N_2 gas can also be possible in this process and nitrate removal has been accomplished with the precipitation of $Fe(OH)_3$. However, no evidence was presented to support it by the authors.

The removal of some inorganic compounds such as nitrate, nitrite, and ammonium ions from paper mill effluents by electrocoagulation was studied by Ugurlu (2004). In these experiments iron and aluminium plates were used as electrodes. Nitrate removal at same electrolysis time (7.5 min) reached 95 and 65% when Fe and Al electrodes were used. It was found that 40–80 mA current value was sufficient to remove some inorganic compounds by electrocoagulation on the large scale, from paper mill effluents. The removal mechanisms were not considered by the author.

Batch experiments were designed to investigate the nitrate removal efficiency by ECF process (Emamjomeh and Sivakumar, 2005). Nitrate removal efficiency depends on electrolysis time and current values. At both low current and electrolysis time, the nitrate removal efficiency was lower. A minimum of 55 min electrolysis time is required to reduce nitrate concentration from 45 mg L^{-1}–N to maximum acceptable level at a current value of 2.5 A. At an operating current of 2.5 A, the nitrate removal efficiency can reach up to 90%. The experimental result showed that the rate of change of nitrate concentration can be expressed as a first order process. Maintaining high pH in the range 9–11 was favourable for this process. The results obtained indicate that the ammonia concentration increased when nitrate concentration was decreased in the solution. Although ECF process can be used as a method for denitrification, it should be kept in mind that for the process to work satisfactorily, high pH is to be maintained and the ammonia generated may have to be removed.

The results on the investigation of the electrocoagulation process for treatment of nitrate, nitrite, and ammonia indicated that using an EC reactor removes the pollutants from aqueous solution. However, further investigations need to be done when the removal mechanisms were not considered in the literatures.

3.7. *Fluoride removal*

An electrochemical method to remove fluorine from drinking water was studied by Li-Cheng (1985). The main aim was to describe a mechanism for removing fluorine by electric coagulation. The experiments were conducted at a 4 L h^{-1} flow rate with an initial F^- concentration 4.5 mg L^{-1}. When aluminium electrodes are used as electrodes for removing fluoride, the following processes are summarised:

3.7.1. *Electrochemical process*

It contains the reaction on the anode (reduction of aluminium) and cathode electrode, and formation of aluminium hydroxide. As a side reaction on the cathode, hydrogen gas can be taken as minute bubbles and floc floating up together.

3.7.2. *Adsorption of $Al(OH)_3$ towards fluorine*

After electrolysing the formation of $Al(OH)_3$ flocs is possible. The fluorine can be adsorbed by these flocs and then the aluminium fluoride complex will be settled.

3.7.3. *Moving process of fluorine in the electric field*

In the electrocoagulation process the fluoride ions move towards the anode. The following reactions may occur by a combination of F^- with Al^{3+}, Na^+ and subsequent precipitation of Cryolite (Na_3AlF_6).

The experimental results showed that the removal of fluoride reached 78% when the initial concentration of fluoride and current density 4.5 mg L^{-1} and 21 A m^{-2}. The consumption of aluminium in the electrocoagulator was 15.1 g for every gram of fluoride ion removed. The experimental results indicated that not only can the concentrations of fluoride ion be reduced by electrocoagulation process but also the total hardness, ammonia (as N), chloride, and ferrous contents. However, the effects of two important parameters including electrolysis time and initial pH on the fluoride removal were not considered by the author.

An electrocoagulation technique was applied by Ming et al. (1987) to remove excess fluoride in potable water. Different parameters were investigated in this study by the EC process including pH, current density, and stirring rate on defluoridation. It was found that fluoride can be reduced from 5 to 1 mg L^{-1} by precipitation at a pH range of 5.5–7. In the electrocoagulation process, hydrogen is released at the cathode and micro-bubbles that are adsorbed into flocs produced by electrolysis are floated up to the surface. The experimental results showed that when the speed of stirring is increased in the electrocoagulation reactor defluoridation decreased in the EC process. This is because some micro-bubble cut off the adsorption layer between the fluoride and colloids and a shearing stress will be produced on the colloidal clots. In this study, the Al^{3+}/F^- ratio was found to be between 10 and 15 to ensure a satisfactory result. In summary, the results indicated that the electrocoagulation method is an efficient fluoride treatment for potable water. However, the mechanism of fluoride removal was not considered by the authors in the paper.

Removal of fluorine and chromium from industrial wastewaters and underground sources using electrochemical technology was investigated by Drondina and Drako (1994). The process consisted of two stages. First iron (II) and calcium hydroxide solutions were added and the sediment was separated. In the second stage, electrocoagulation and electroflotation processes were applied after acidification. Electrocoagulation and electroflotation processes were conducted with different electrode types, including aluminium (D-16), and an insoluble anode (titanium coated with manganese oxides) or a soluble anode (grid of stainless steel). The experiments were conducted at initial concentration of F^- from 8 to 14 mg L^{-1}. The main aims of this research are as follows:

- To study the effect of the initial pH on purification by EC process.
- To study the efficiency of applying aluminium hydroxide. This is produced by dissolution of the aluminium anode under current values.
- To review the possibility of using electroflotation for separation

The experimental results of applying aluminium hydroxide generated by hydrolysis showed that electrocoagulation is an efficient process for removing fluoride because of its higher adsorption capability. The results showed that defluoridation was more efficient for a pH ranging from 6.35 to 6.65 when the initial fluoride concentration decreased from 14 to 1 mg L^{-1}. In this research, the effects of the important parameters including current and electrolysis time were not considered.

An efficient EC process for defluoridation of Sahara water (Ain Boukhial region) in North Africa that does not require a large investment was investigated by Mameri et al. (1998). The influence of some experimental parameters including electrolysis time, initial concentration of fluoride, distance between electrodes, current density, temperature solution, initial pH, electrode area to cell volume ratio (A/V), were investigated on the defluoridation using bipolar aluminium electrodes. Effect of the initial fluoride concentration on the EC process was studied in this research. The experimental result, results not shown, illustrated that the treatment time is needed to increase when initial concentration in the solution is increased. The kinetic of defluoridation of the drinking water followed an exponential law with electrolysis time. The results showed that defluoridation is more efficient for a pH ranging from 5 to 7.6, where strong fluoro–aluminium complexes such as: AlF_3, $AlOHF_3^-$ and $Al(OH)_2F_2^-$ are present, inducing an efficient aluminium complexion by fluoride. In this process the experimental Al^{3+}/F^- mass ratio was found to be 16–17.5. The experimental results showed when the distance between electrodes is increased; the less fluoride is removed due to an increasing ohmic resistance between electrodes. The determination of the characteristics of water before and after the treatment has confirmed no contamination of the natural water by aluminium during experiments was observed. The results obtained showed that the aluminium bipolar electrodes system is an effective process for the defluoridation. However, no evidence was presented in this paper to support the mechanism of defluoridation.

Defluoridation by aluminium sorbent (Al-sorbent), which was produced in a parallel-plate electrochemical reactor by anodic dissolution of aluminium electrodes in a dilute sodium chloride (NaCl) aqueous solution, was studied by Yang and Dluhy (2002). A trace amount of chloride ions in the solution is enough to penetrate the oxide file on the aluminium electrodes for dissolving to continue. Tap water was used as the experimental design parameters for defluoridation. Based on the aluminium hydroxide solubility diagram, the principal soluble specie is the monomeric anion of $Al(OH)_4^-$ when $pH > 8$. At lower pH, $pH < 6$, the dominant soluble species are cationic monomers such as Al^{3+}. The experimental results indicated that the freshly generated Al-sorbent is able to reduce fluoride from 16 to 2 mg L^{-1} in 6 min. However, the effluent from the reactor also needs a pH adjustment. The review shows that measurement of fluoride concentration was done by SPADNS method. A buffer must be added to the samples to prevent the interference of the Al^{3+} ions, which was not considered by the authors.

The combined EC and EF process was successfully used to remove fluoride ions from industrial wastewater (Shen et al., 2003). In this combined process the EC unit was used to produce aluminium hydroxide flocs. The EF unit was used to separate the flocs formed from water, by floating them to the surface of the cell. The research investigated that aluminium hydroxide floc is believed to strongly adsorb fluoride ions. The experimental result, results not shown, illustrated that the residual fluoride decreased when charge loading was increased in the EC reactor. The experimental result, results not shown, illustrated that at the same charge loading, the residual fluoride decreased when the initial concentration decreased. In order to explain the mechanism for removing fluoride, the composition of the sludge was investigated using X-ray Photoelectron Spectroscopy (XPS). The results showed that at high initial concentration, according to the equilibrium, more fluoride was found in the sludge. The negative ToF-SIMS[1] spectra of the dried sludge showed that the strongest peaks are 61, 62, 63, 77, 79 and 81 that represent $Al(OH)_2$, AlFO, Al(OH)F, $Al(OH)_2O$, $AlFO_2H$ and AlF_2, respectively (results not shown). The results indicated that defluoridation is a chemical adsorption process with F^- replacing the –OH group from $Al_n(OH)_{3n}$ floc. At high initial concentration, according to the equilibrium, more fluoride was found in the sludge. However, no information regarding the sludge volume measurement was presented for EC process.

A laboratory batch EC reactor was used to investigate the effect of some anions including Cl^-, NO_3^-, and SO_4^{2-} on defluoridation by the ECF system (Hu et al., 2003). Defluoridation in solutions containing F^-, SO_4^{2-} and Cl^- ions was higher than in solutions containing F^-, SO_4^{2-} and NO_3^-ions because the lyotropic[2] series of anions for Al^{3+} are $F^- > SO_4^{2-} > Cl^- > NO_3^-$. The experimental results showed that defluoridation did not change very much with concentrations of Cl^- and NO_3^- but decreased when the concentration of SO_4^{2-} ions is increased. It may be due to competition effect stated by Hu et al. (2003) that increase of the concentration of sulphate and other anions, which have a strong affinity with Al^{3+}, decreases the adsorption capacity of fluoride onto hydrous alumina. Further research is needed to yield a more complete understanding of the mechanism of the EC process. The effect of pH was not considered by the authors.

A laboratory batch electrocoagulation/flotation reactor was used for defluoridation and SS removal of wastewater from semiconductor manufacturers that contains large amounts of fluoride by Hu et al. (2005b). Experiments for the ECF process with seven aluminium electrodes were undertaken in a bipolar batch reactor. An anodic surfactant, sodium dodecyl sulphate (SDS), was also applied to improve the flotation performance of the ECF process to remove the dissolved fluoride ions and CaF_2 particles in the semiconductor wastewater following calcium precipitation. The semiconductor wastewater was first treated by the conventional precipitation process with calcium salts and then removed by the proposed ECF process. The dissolved fluoride ions after calcium precipitation were effectively removed (more than 90%) in the ECF process. The defluoridation process was found to be efficient for a pH ranging from 6.5 to 7.5 during experiments. The experimental results showed that the dosage of SDS in ECF process is much lower than in the dissolved air flotation (DAF) or dispersed air flotation (DiAF) processes, because the CaF_2 particles can be collected by hydro-fluoro-aluminium flocs in ECF process. However, no information regarding cost estimation was presented for the ECF and DAF processes.

A laboratory batch electrocoagulation reactor was designed to investigate the effects of parameters such as, current density, electrolysis time, electrolyte pH, and using electrical conductivity to remove fluoride of the cell (Emamjomeh et al., 2003). At a constant current of 1.5 A the fluoride removed was increased from 22 to 93% by increasing the electrolysis time from 5 to 60 min. In most electrochemical processes current density is the critical parameter for controlling the reaction rate in the reactor. The highest current (2.5 A) removed the most fluoride. However, more coagulant (aluminium) is available per unit time at higher currents and this may be unnecessary because excess residual aluminium is unsafe for drinking water and high current is uneconomic in terms of energy consumption. The $Al(OH)_{3(s)}$ floc is believed to adsorb F^- strongly.

The results of batch experiments using Al electrodes in monopolar configuration showed that the residual concentration of fluoride is decreased when the initial concentration of calcium in the solution is increased (Emamjomeh and Sivakumar, 2004). By adding 250 mg L^{-1} of Ca^{2+} ions into the solution, defluoridation is increased by approximately 13%. However, increasing Ca^{2+} in the solution will cause increasing hardness in drinking water. Therefore, regarding an optimum residual calcium concentration in the effluent, the best current was 1.5 A when the initial concentration of fluoride and calcium respectively were 15 and 100 mg L^{-1} in ECF process. One of the main advantages of Ca^{2+} in the solution for defluoridation by ECF is reduced electrolysis time. The XRD traces also showed that the main reason for increasing defluoridation by ECF process is formation of CaF_2 granule and alumino-fluoro-hydroxyl complexes $[Al(OH)_{3-x}F_x]$ in the solution.

A clear understanding of various interconnected processes including measurement, speciation, and mechanisms was explained for successful scale-up of defluoridation process by electrochemical method by Sivakumar and Emamjomeh (2005a). The experimental results showed that the pH of solution is an important factor for removing of fluoride. When $pH < 5$, dissolved aluminium (Al^{3+}) is predominant in the acidic condition and aluminium hydroxide is tended soluble. In the pH range of 5–6 the predominant hydrolysis products are found to be $Al(OH)^{2+}$ and $Al(OH)_2^+$ when the solid $Al(OH)_3$ is most prevalent in the pH range of 6–8.5. When pH is >9, the soluble $Al(OH)_4^-$ is the predominant species. The results showed that the fluoride removal efficiency remains highest and remain unchanged between the adjusted pH range of 6–7.5 and then decreased when adjusted pH value is increased from 8 to 10. Thus weakly acidic condition is favoured in this treatment. The mechanism of the removal process was confirmed to be a competitive adsorption between OH^- and F^- at adjusted pH range of 6 and 7.5. The XRD analysis of the composition of the dried sludge obtained by electrocoagulation process shows the formation of $Al(OH)_{3-x}F_x$ and provides confirmation for the main mechanism for fluoride removal (Sivakumar and Emamjomeh, 2006).

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 (Sivakumar and Emamjomeh, 2005b). 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

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[1] Time-of-Flight Secondary Ion Mass Spectrometry (ToF-SIMS) uses a pulsed primary ion beam to desorbs and ionize species from a sample surface.

[2] Materials in which liquid crystalline properties appear induced by the presence of a solvent, with mesophases depending on solvent concentration, as well as temperature.

Table 1
Summary of pollutants removed by electrocoagulation in water and wastewater sources.

Reference	Pollutants	Current or current density	Cell voltage (V)	Electrode materials, electrode Connections	Removal path	Flow rates	Treatment efficiency (%)	Reactor
OIL, GREASE								
Kaliniichuk et al. (1976)	Oil- refinery	2–35 A m^{-2}	1.2–1.7	Al- Packed sheets	Floated and settled	–	60–99.5	Batch
Clemens (1981)	Oil- wastewater	300–2500 A	8–12	–	Floated and settled	–	–	–[a]
Weintraub et al. (1983)	Oil	100 A m^{-2}	5–30	Fe-mesh	Flotation in separate cell	50–300 mL min^{-1}	70–99	Continuous
Balmer and Foulds (1986)	Oil	200–781 mA	3–39	Fe/Pt- mesh	Flotation then filtration	–	70–100	Continuous
Ibanez et al. (1995)	Oil -wastewater	–	1.5	Fe/stainless steel	Floated and settled	–	60–99	A 250 mL beaker
Hosny (1996)	Oil	1.2 A, 100 A m^{-2}	8	A lead anode and stainless steel cathode	Floated	10–50 mL min^{-1}	65–92	Batch and continuous
Rubach and Saur (1997)	Oil	40–220 A	4–45	Corrugated Al plate	Floated	0.25–1.5 m^3 h^{-1}	95–99	Continuous
Chen et al. (2000)	Restaurant oil, COD	0.2–0.6 A	1.3–15.5	Fe and Al- Bipolar	Floated and settled	6–9 L h^{-1}	95–99	Continuous
Mostefa and Tir (2004)	Oil	60–140 A m^{-2}	–	Flat sheets of steel/stainless steel	Flotation in separate cell	–	85–99	Batch
Carmona et al. (2006)	Oil suspension from water	100–300 A m^{-2}	–	Al electrodes-Monopolar	Floated and settled	370 mL min^{-1}	80–99	Continuous
COLOUR								
Ogutveren et al. (1992)	Dye	10–17.5 mA	0.86–1.3	Graphite rods- Bipolar packed bed	Settled	–	98–100	Batch
Lin and Peng (1994)	Textile wastewater-Dye	1–3.5 A	–	Fe- Monopolar	Floated and settled	0.75 L min^{-1}	50–61	Batch and continuous
Ibanez et al. (1998)	Dye	–	9	Fe/Steel	Settled	–	–	A 10 mL beaker
Gurses et al. (2002)	Dye	1–5 mA cm^{-2}	5–15	Fe and Al plates, and cylindrical graphite	Settled	–	85–99	Batch
Kim et al. (2002)	Dye	1–4.5 A m^{-2}	–	Al, Fe, and stainless steel-Monopolar	Settled	50–200 mL min^{-1}	95–99.6	Continuous
Can et al. (2003)	Dye	2.5–25 mA cm^{-2}	30	Al/Al- Monopolar	Floated and settled	–	70–92.5	batch
Daneshvar et al. (2004)	Dye	60–80 A m^{-2}	–	Iron (ST 37-2) and steel (grade 304)- Monopolar	Settled	–	32–95	Batch
Mollah et al. (2004b)	Dye	4A	42	Carbon steel plates- Bipolar	Settled	350–600 mL min^{-1}	97–99	Continuous
Bayramoglu et al. (2004)	Textile wastewater-Dye-COD	45–200 A m^{-2}, 6 A	30	Al/Fe- Monopolar	Settled	–	85–99	Batch
Fan et al. (2004)	Dye	–	25–150	Graphite plate- Bipolar packed bed	Settled	1-5 L h^{-1}	90–96	Continuous
Kobya et al. (2005)	Dye	25–250 A m^{-2}	4–12	Al/Al -Monopolar	Settled	–	60-95	Batch
Golder et al. (2005)	Dye, COD	1–30 mA cm^{-2}	–	Mild Steel electrode pair	Settled	–	Dye 80–99, COD 74–89	Batch
Can et al. (2006)	Textile wastewater- Dye	100 A m^{-2}	<30	Al/Al- Monopolar	Floated and settled	–	70–80	Batch
HEAVY METALS								
Osipenko and Pogorelyi (1977)	Cr^{6+}	8.33 A m^{-2}	–	Fe- Monopolar	Settled	–	70–85	Continuous
Gnusin et al.(1985)	Cd^{2+}	10–30 A m^{-2}	–	Steel- Monopolar	Settled	–	65–91	Continuous
Drondina et al. (1985)	Se and St	10–20 A m^{-2}	–	Fe/Fe	Floated	–	55–83	Continuous
Pozhidaeva et al. (1989)	Ni, Cr	2000 A m^{-2}	–	Steel	Settled	–	60–85	Continuous
Poon (1997)	Groundwater contaminants: Ni, Zn, Pb, Cu, CN	110–220 A m^{-2}	3–5	A platinum-clad columbium screen/ a stainless steel screen cathode	Floated and settled	19 L min^{-1}	75–96	Batch and continuous
Balasubramanian and Madhavan (2001)	Arsenic	50–125 A m^{-2}	–	Two Fe/stainless steel electrodes	Settled	–	75–95	Glass beaker of 500 ml (Batch)
Park et al. (2002)	Cadmium	420 mA	24	Al/Al- Monopolar	Floated and settled	–	80–90	Batch
Ninova (2003)	Cu, Zn	100–500 A m^{-2}	–	Two Fe electrodes	Settled	–	65–88	Batch
Kumar et al. (2004)	Arsenic	15–22 A m^{-2}	5–15	Two Fe, or Al or Titanium electrodes	Settled	–	Max 99	Batch

(*continued on next page*)

Table 1 (*continued*)

Reference	Pollutants	Current or current density	Cell voltage (V)	Electrode materials, electrode Connections	Removal path	Flow rates	Treatment efficiency (%)	Reactor
Hansen et al. (2005)	Arsenic	0.8–1.2 A dm^{-2}	–	Two Fe electrodes	Floated and settled	3 L h^{-1}	80–98	Continuous
Gao et al. (2005)	Chromium(VI)	0.1 A, 2.5 F m^{-3}	–	Fe/Fe electrodes-Monopolar	Floated and settled	50 mL min^{-1}	80–97	Continuous
Den and Huang (2005)	Silica nano- particles	0–3 A	0–200	Fe/ Fe electrodes-Monopolar	Floated and settled	80–380 mL min^{-1}	75–95	Continuous
Parga et al. (2005)	Arsenic	4–5 A	20–40	Seven parallel plates of Fe and Carbon steel	Sedimentation and flotation by air	600 mL min^{-1}	60–99	Continuous
Urban and industrial Wastewaters								
Novikova et al. (1982)	Detergents	100 A m^{-2}	–	Fe vertical plate electrodes	Flotation in cell then filtration	–	65–78	Continuous
Grechko et al. (1982)	Pesticides	150 A m^{-2}	–	Al/Al Sheets	Settled then filtered	–	–	Continuous
Vik et al. (1984)	Aquatic humus, TOC	0–2000 C	6–12	Al/Al Monopolar	Floated	170 mL min^{-1}	40–70	Continuous
Pouet and Persin (1992)	Urban wastewater: COD	5 A	40	Al/Al -	Floated and settled and microfiltration	350 L h^{-1}	69–77	Continuous
Pouet and Grasmick (1995)	Municipal wastewater: COD	0–40 A	0–80	Al/Al- Plate	Settled and floated with DAF	350 L h^{-1}	70–80	Continuous
Baklan and Kolesnikova (1996)	Organic compounds	120 A m^{-2}	3.5	Fe and Al plates	Settled	–	65–90	Batch
Hernlem and Tsai (2000)	E- coli	0.8 A	9–20	An electrode pack from Sanilec 6 electrolyzer	Chlorine generation and disinfection by electroflotation	42 mL min^{-1}	–	Continuous
Phutdhawong et al. (2000)	Phenolic compounds	0.5 A	22	Two Al plates	Settled	–	60–92	Batch
Shin et al. (2001)	Livestock wastewater	0.6–1.9 A	24–48	Al/Al- Monopolar	Floated	–	SS- 90, BOD- 41	Batch
Ciorba et al. (2002)	Nonylphenol ethoxylates	0.02 A	2	Two Al/Fe	Adsorption on floc and settled	–	30–50	Batch
Larue et al. (2003)	Latex particles	110–880 A m^{-2}	2.1–18.7	Two Fe electrodes	Settled and filtered	–	–	Batch
Jiang et al. (2002a)	DOC, colour	2.5–10 A m^{-2}	–	Al/Al Monopolar	Floated and settled	170 mL min^{-1}	DOC- 51 Colour 70	Continuous
Kobya et al. (2003)	Textile wastewater COD	5–20 mA cm^{-2}	$<$30	Fe/Al- Monopolar	Floated and settled	–	90–99.5	Batch
Ge et al. (2004)	COD, SS Laundry wastewater	30 A	32	Al/titanium- Bipolar	Floated and settled	1.5 m^3 h^{-1}	70–90	Continuous (Field test)
Kobya et al. (2005)	COD	25–200 A m^{-2}	$<$30	Al and Fe- Monopolar	Floated and settled	–	65–93	Batch
Hutnan et al. (2005)	Municipal wastewater-COD	30–80 A m^{-2}	3–18	Fe/Al electrodes-Monopolar	Floated and settled	1 m^3 h^{-1}	50–80	Continuous
Zhu et al. (2005)	Virus- wastewater	0.1–0.3 A	–	Fe/steel rod shaped electrodes	Settled	–	99.99	Batch
SS, TURBIDITY								
Stuart (1946)	Turbidity, hardness colour	5.8–28 A	2.1–5.1	Two Al sheet	Settled	–	55–74	Batch
Bonilla (1947)	Turbidity, hardness colour	2–7.3 A	8.8–28	Parallel Al sheet (Monopolar)	Settled	–	50–70	Batch
Sandbank et al. (1973)	Suspended solids	0.14–3.2 A, 22–500 A m^{-2}	9.5–11.5	Graphite and iron	Electroflotation	–	80–83	Batch and continuous (Field test)
Przhegorlinskii et al. (1987)	Suspended Solids	5–50 A m^{-2}	–	Al and Fe Plates	Settled	–	75–80	Continuous
Zolotukhin (1989)	Mine water: SS	–	9–20	Al/Fe plate	Floated and settled	0.015 m^3 h^{-1}	90–94	Continuous
Mills (2000)	Raw water and bacteria, E. coli	11–62.5 A m^{-2}	3	Fe and steel plate	Settled	4.5–23 L min^{-1}	90–99	Continuous
Matteson et al. (1995)	SS, Kaolinite	0.01 A m^{-2}	15–50	Two flat steel wire mesh	Settled	–	90–95	Batch
Abuzaid et al. (2002)	turbidity	0.2–1 A	–	Fe/Stainless steel	Settled	–	80–95	4 L reactor
Holt et al. (1999, 2002)	Potter's clay	0.25–2 A	3–24	Al/Stainless steel Monopolar	Floated and settled	–	90–95	Batch
Han and Kwon (2002)	Turbidity	–	–	Al/Al Monopolar	Settled	–	90	Batch
Larue and Vorobiev (2003)	Turbidity	1 A		Two Fe electrodes	Settled	–	90–93	Batch
Holt et al. (2004, 2005)	Turbidity Potter's clay	0.25–2 A	3–24	Al/Stainless steel Monopolar	Floated and settled	–	90–95	Batch
Sivakumar et al. (2004)	Turbidity	0.5–2.5 A, 6.25–31.5 A m^{-2}	6–25	Al/Al Monopolar	Settled	–	60–88	Batch

IONS								
Gnusin et al. (1986)	Natural water	10–500 A m^{-2}		Steel/Al - Monopolar	Settled			Continuous
Sanfan and Qinlai (1987)	Brackish water: hardness, SO_4^-, Cl^-	2–30 A m^{-2}	1.3–3.6	Al/Fe plate	Settled and filtered	10 L h^{-1}	60–80	Continuous
Sanfan (1991)	Brackish water	1–22 A m^{-2}	1.1–3.1	Al/Fe plate	Floated and settled	18 L h^{-1}	40–76	Continuous
Daneshvar et al. (2003)	Brackish water: hardness, SO_4^-, Cl^-	22 A m^{-2}	–	Fe and steel plate	Floated and settled	10–18 L h^{-1}	40–90	Continuous
Alfafara et al. (2002)	P, Algae from eutrophied lake water	80–430 A m^{-2}	–	Al/ titanium alloy or carbon anode	Electroflocculation and electroflotation	–	40–50	Batch and continuous
Bektas et al. (2004)	Boron	0–5 A	0–30	Al/Al-Monopolar	Settled	–	92–96	Continuous
Yilmaz et al. (2005)	Boron	0–5 A	0–30	Al/Al-Monopolar	Settled	–	55–97	Continuous
Lin and Wu (1996)	Nitrite and ammonia	1–2.5 A	–	Graphite/ Titanium dioxide, Monopolar	Electro-oxidation and electroflotation	–	60–99	Batch
Paidar et al. (1999)	Nitrate	28–76 A m^{-2}	–	Ti/Pt anode and Cu cathode	Electrochemical reduction	–	60–91	Batch
Koparal and Ogutveren (2002)	Nitrate	–	2.9, 20 – 80	Graphite rod anode and carbon cloth cathode for Electro-reduction, Fe packed bed for EC, Bipolar	Electro-reduction and electrocoagulation	–	80–90	Batch
Ugurlu (2004)	Nitrite, nitrate, and ammonia	40–80 mA	12	Al and Fe electrodes	Electro-reduction and electrocoagulation	–	65–95	Batch
Emamjomeh and Sivakumar (2005)	Nitrate	0.5–2.5 A, 6.2–31.2 A m^{-2}	6–25	Al/Al Monopolar	Floated and settled	–	70–90	Batch
Li-cheng (1985)	Fluoride	0.1–3 A, 2.2–66.7 A m^{-2}	2.5–17	Al/Al plates	Adsorption, flotation, and sedimentation	4 L h^{-1}	15–92	Continuous
Ming et al. (1987)	Fluoride	14 A m^{-2}	–	Al/Al Monopolar	Adsorption, and sedimentation	–	80–88	Batch
Drondina and Drako (1994)	Fluoride	–	–	Al electrodes for EC, Stainless steel , titanium electrode for EF process	Floated and settled in separate cells	–	85–90	–
Mameri et al. (1998)	Fluoride	3.12–289 A m^{-2}	1–2.4	Al/Al Bipolar	Settled	–	70–92	Batch
Yang and Dluhy (2002)	Fluoride	1 A	14	Al/ Al plates, Parallel	Adsorption and Sedimentation	–	55–88	Continuous
Emamjomeh et al. (2003)	Fluoride	0.5–2.5 A, 6.2–31.2 A m^{-2}	6–25 V	Al/Al Monopolar	Floated and settled	–	60–99	Batch
Shen et al. (2003)	Fluoride	0–5 F m^{-3}	–	Al/Al Bipolar	Adsorption and Sedimentation	–	67–90	Continuous
Hu et al. (2003)	Fluoride	0.4 A	–	Al/Al Bipolar	Adsorption and Sedimentation	–	20–100	Batch
Emamjomeh and Sivakumar (2004)	Fluoride	0.5–2.5 A, 6.2–31.2 A m^{-2}	6–25 V	Al/Al Monopolar	Floated and settled with addition of Ca^{2+} ions	–	60–99	Batch
Hu et al. (2005a)	Fluoride	0.6A	35	Al/Al Bipolar	First calcium precipitation and then settled	–	70–95	Batch
Sivakumar and Emamjomeh (2005a)	Fluoride	0.5–2.5 A, 6.2–31.2 A m^{-2}	6–25 V	Al/Al Monopolar	Floated and settled (Speciation)	–	60–99	Batch
Sivakumar and Emamjomeh (2005b)	Fluoride	12.5–50 A m^{-2}	8–28 V	Al/Al Monopolar	Floated and settled	150–400 mL min^{-1}	89–98	Continuous
Emamjomeh and Sivakumar (2006)	Fluoride	0.5–2.5 A, 6.2–31.2 A m^{-2}	6–25 V	Al/Al Monopolar	Floated and settled (Empirical model)	–	60–99	Batch
Ma et al. (2007)	Fluoride	–	1–5	Al/Al Bipolar	adsorption	–	70–92	Batch
Hu et al. (2007)	Fluoride	0.6 A	35	Al/Al Bipolar	First calcium precipitation and then settled	–	70–95	Batch

(*continued on next page*)

Table 1 (*continued*)

Reference	Pollutants	Current or current density	Cell voltage (V)	Electrode materials, electrode Connections	Removal path	Flow rates	Treatment efficiency (%)	Reactor
Zhu et al. (2007)	Fluoride	–	–	Al/Al Bipolar	Adsorption and Sedimentation	–	60–90	Batch
Kobya and Delipinar (2008)	Baker's yeast wastewater	0–12 A	0-60	Al and Fe electrodes Monopolar	Adsorption and Sedimentation	–	70–90	Batch
Yilmaz et al. (2008)	Boron containing wastewater	0–10 A	0-30	Al/Al Monopolar	Adsorption and Sedimentation	–	99	Batch
Sayinera et al. (2008)	Boron	10–30 mA cm^{-2}	–	Al and Fe electrodes Monopolar	Adsorption and Sedimentation	–	60–90	Batch
Wang et al. (in press)	Laundry wastewater, COD	–	5	Al and Fe electrodes Monopolar	Adsorption	–	50–62	Batch

[a] Note – blank indicates information was not available.

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 18.7 A m^{-2} for a flow rate of 150 mL min^{-1} and for an initial fluoride concentration of 10 mg L^{-1}. The optimum current density can be increased to 50 A m^{-2} if flow rate is to be increased to 400 mL min^{-1}. XRD analysis of the composition of the dried sludge obtained by EC process shows the formation of $Al(OH)_{3-x}F_x$ and it is the main reason for defluoridation by electrocoagulation process.

An empirical model is developed to relate the critical parameters such as current concentration (I/V), electrode distance (d) and initial fluoride concentration (C_o) with the rate constant (K) for fluoride removal using monopolar ECF process (Emamjomeh and Sivakumar, 2006). Based on the operational parameters, an empirical equation is given to calculate the optimal detention time (d_{to}) for fluoride removal (Eq. (3)).

$$d_{to} = \frac{1}{10^{-5}\left[5.9\left(\frac{I}{V}\right) - 37.1(C_o) - 82.1(d) + 2746.4\right]}\ln(C_o) \qquad (3)$$

the optimal detention time for fluoride removal using monopolar ECF process is calculated for various operational parameters including current values (1–2.5 A), electrodes distance (5–15 mm), and initial fluoride concentrations (10–25 mg L^{-1}). The results show good agreement between the experimental data and the predictive equation. The fluoride removal efficiency increases steadily with increasing current values from 1 to 2.5 A. In the batch monopolar ECF process, the optimal detention time was found to be 55 min when the operational parameters including initial F^{-} concentration, current value, and inter electrode distance were respectively kept at 10 mg L^{-1}, 1.5 A, and 5 mm. Overall, the results showed that the electrocoagulation technology is an effective process for defluoridation of water. It is concluded that the electrocoagulation technology using aluminium electrodes is a viable process for defluoridation of water supplies that contains excess fluorides

The previous research studies on the batch and continuous flow experiments using Al electrode in monopolar and bipolar configuration showed that electrocoagulation technology is an effective process for defluoridation, but more investigation needs to be done. Diverse range explanations have been presented in the literatures for the optimization of operating condition in batch and continuous flow reactor. A number of studies have been also performed to describe the effects of the different operational parameters, yet no more attempts has been made to present an empirical model for defluoridation by ECF process. The speciation of Al and Al–F complexes were not more considered by the previous researchers when it aims to better understanding of the electrocoagulation fundamentals and investigation on the fluoride removal mechanisms in the EC process based on the solution speciation and dried sludge characteristics.

4. Conclusions

Electrocoagulation is an electrochemical technique with many applications. Oily wastes can be removed from aqueous solution by EC process. The experimental results showed that aluminium electrodes are more efficient than iron electrodes. It may be because of high adsorption capacity of hydrous aluminium oxides for oil removal. The electrocoagulation technology is also considered to be potentially an effective tool for treatment of colour from textile wastewaters with high removal efficiency. The removal efficiency was found to be dependent on the initial pH, the dye concentration, the applied current density, and the electrolysis time in batch model. It was found that maximum 98.5% of the dye was removed from the solution by EC technology. The capacity and efficiency of the EC process for treatment of industrial wastewaters was found to be dependent on the nature of the liquid waste and initial pollutant concentrations. The results on the investigation of the EC process for treatment of heavy metals indicated that using an EC reactor successfully removes arsenic and cadmium from water or wastewater. The highest removal of arsenic was obtained by using iron electrodes when the removal mechanism was considered to be adsorption of arsenic on hydrous ferric oxides. However, no evidence was reported to support it. Other research studies showed that EC process is more efficient than coagulation process for potable water treatment. Less aluminium dosage was needed to get the same removal efficiency in EC process, which is related to chemical cost and sludge production. The results on the investigation of the electrocoagulation process for treatment of nitrate, nitrite, and ammonia indicated that the pollutants can be removed from aqueous solution. However, further investigations need to be done when the removal mechanisms were not considered in the literatures. The previous research studies on the batch and continuous flow experiments using Al electrode in monopolar and bipolar configuration showed that EC method is an effective process for defluoridation, yet more investigation needs to be done. The speciation of Al and Al–F complexes was not more considered in the literatures. The fluoride removal mechanisms are investigated in the electrocoagulator based on the solution speciation and dried sludge characteristics. In summary, Table 1 presents pollutants removed by EC in water and wastewater sources and also associated references.

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