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A Review on “Fluorides Removal by Electro-coagulation Method Using AC and DC Current in Water Treatment”

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ABSTRACT: Fluoride ions present in drinking water are beneficial to human health when the current enforceable drinking water standard for fluoride is 4.0 mg/L. This is the maximum amount that is allowed in water from public water systems, also called the Maximum Contaminant Level (MCL). An excess intake of fluoride ($>1.5 \text{ mg L}^{-1}$) may pose several health problems like Fluoride not only affects bone and teeth, but it also impacts young children in the development of the brain. Exposure to fluoride before birth could lead to poorer cognitive outcomes in the future. Higher levels of fluoride lead to low scores in IQ tests. In this context, reducing high fluoride concentrations in water is a major worldwide challenge. According to WHO 1984 and Indian standard drinking water specification 1991 the maximum permissible limit of fluoride in drinking water is 1.5 ppm and highest desirable limit is 1.0 ppm.. The application of electro-coagulation (EC) processes using AC and DC Current has received widespread and increasing attention as a promising treatment technology and a competitive treatment for fluoride control. EC technology has been favourably applied due to its economic effectiveness, environmental versatility, amenability of automation, and low sludge production. This research paper provides more detailed information on fluoride removal from water by the EC process using AC and DC Current, including operating parameters, removal mechanisms, energy consumption, and operating costs.

KEYWORDS- Electro-coagulation, Water Treatment, Fluorides, Human Health, WHO

I. INTRODUCTION

1.1 BACKGROUND OF THE STUDY

Fluoride is one of the most essential elements to the human body. It has been observed the consumption of the fluoride in the water should not exceed above 1.5mg/L. This leads to fluorosis problem in the human being. It has undertaken the range of the semi-arid area and source where the ground water is the only source for drinking water. Fluoride is encountered as important geochemical deposits in Indian sub-continent. There are different channels through which natural fluoride pollution occurs in the environment. It has been examined that the discharge of the wastewater from the industries would lead to heavy contribution in the water reserves especially in ground water. According to Central Pollution Control Board of India, maximum number of the fluoride constituent is directed from industrial wastewater which is around 15mg/l. Fluoride removal from the contamination from the groundwater has been reluctantly done by using various processes such as adsorption, reverse osmosis, chemical precipitation and Nalgonda technique, ion-exchange and electro-coagulation. Each and every method has their operational importance and constraints. Electrocoagulation is the method which utilizes wash water treatment and other processed water through radio frequency diathermy or electrolysis of short wave. This method has the ability to remove the contaminants from the water which are more difficult to remove by filtration or chemical treatment systems. A



typical electro-coagulation unit consists of electrochemical cell which has been connected metal electrodes and gaining power from DC supply. The only difference observed in the electro-coagulation method is that they generate coagulant from the electrochemical dissolution at anode. Further, the sacrificial anode is however made up of aluminum metal where the aluminum hydroxide flocs are produced. However, the negative charged fluoride ions and precipitated out in the form of sludge. The classical physico-chemical treatment processes that are used for the wastewater treatment is filtration, air stripping, ion-exchange, chemical precipitation, chemical oxidation, carbon adsorption, ultrafiltration, reverse osmosis, electrodialysis, volatilization and gas stripping. One of the advanced electrochemical technology based technique is the electrocoagulation process. Electrocoagulation (EC), the passing of electric current through water, has proven very effective in the removal of contaminants from water. Electrocoagulation systems have been in existence for many years using a variety of anode and cathode geometries, including plates, balls, fluidized bed spheres, wire mesh, rods and tubes. In the past few decades it has been used for the treatment for the water containing foodstuff wastes, oil wastes, dyes, suspended particles, chemical and mechanical polishing waste, organic matter from landfill leachates, defluorination of water, synthetic detergent effluents, mine wastes and heavy metal-containing solution. EC has become one of the affordable wastewater treatment processes around the world by reducing electricity consumption and miniaturization of the needed power supplies.

II. LITERATURE REVIEW

2.1 Introduction

The world population will exceed 8 billion in 2025, double the number it was in 1975 (United Nations, 2018). This will lead to rapid increase of water consumption in different sectors, i.e., domestic, agriculture and industry. While water scarcity is already a key concern in several countries, with the future water demand, industries need to search for alternative sources of water supplies other than surface and ground water. Industries use large quantities of water that mostly ends up as wastewater. Thus, recovered industrial wastewater is an important water resource. Industrial effluents are treated for meeting wastewater discharge standards, recovery of valuable constituents, and reuse. Treatment methods that can satisfy these objectives are becoming progressively attractive. This study aims on applications of electroflotation in treatment of industrial wastewaters, with detailed discussions on fundamentals of electroflotation process, electrode materials and arrangements, design aspects of electroflotation reactor and influential process variables. In this chapter, a comprehensive review on related subjects is presented.

2.2 Fundamentals

Electroflotation/ Electrocoagulation is separation of suspended particles from water by means of gas bubble generated at electrodes during electrolysis of water. The process was first employed in the mineral industry and later was implemented in water and wastewater treatment (Bhaskar Raju and Khangaonkar, 1984). Throughout the process, fine bubbles nucleate at electrodes, detach and while rising to the water surface, collide with solid or liquid particles suspended in water. Some of these collisions lead to attachment of particles and bubbles and formation of bubble-particle aggregates. Then, the aggregates ascend to the water surface and are collected by mechanical skimming.

Electroflotation has been receiving increasing attention by researchers for treatment of different effluents including industrial wastewaters in recent years. By generating fine bubbles, e.g., 1-30 μm compared to 50-100 μm in pressurized air flotation (Il'in and Sedashova, 1999) the process efficiency is higher than conventional flotation methods because of larger number and surface area of bubbles. The greater density of bubbles increases the chance of bubble-particle collision, aggregation and removal. Furthermore, fine bubbles have greater surface to volume ratio, and therefore, greater overall surface interactions between bubbles and particles occur. Electroflotation systems do not usually include mechanically-moving parts, making the installation, retrofit and maintenance simpler and more convenient. Also, process adjustments can be readily accomplished by changing applied electric potential/current.



Electroflotation can be an alternative to chemical treatment methods. The chemical methods involve addition of coagulant and flocculant chemicals to wastewater, which can be expensive and also produce large quantities of non-recoverable sludge. In comparison electroflotation can be implemented without adding chemicals.

2.3 Electrodes

2.3.1 Electrode Material

Electrodes are the heart of electroflotation reactors and therefore, their material and design are crucial for the performance of the system. Electrode materials are divided into two categories, i.e., inert and active. This is the case especially for anode electrodes.

Since the cathode does not corrode in electroflotation, stainless steel and aluminium are the most common and inexpensive materials used for the cathodes in the treatment of industrial wastewaters (Mostefa and Tir, 2004; Mansour and Chalbi, 2006; Kobya et al., 2006; Ezechi et al., 2014; Aoudj et al., 2017).

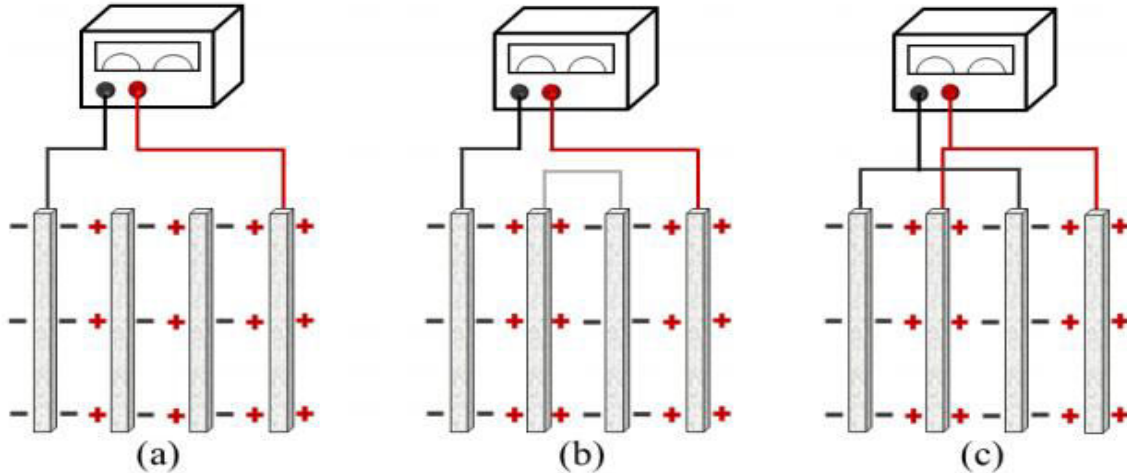
Inert anodes are used for production of oxygen gas bubbles in electroflotation. Graphite is used as an anode in electrochemical processes and electroflotation treatment of wastewater (Muruganathan et al., 2004; Yang, 2007; Zaidi et al., 2016). It has a relatively stable condition; however, studies show that graphite deteriorated rather quickly (e.g., Hernlem and Tsai, 2000), and lost its smooth surface resulting in the production of coarse-sized bubbles and reduction of separation efficiency.

Dimensionally stable anodes (DSA) are made of coated metals such as titanium. They were first patented in the US in 1966 in form of TiO₂-RuO₂-coated titanium for chlorine evolution. Oxides of other metals, e.g., Pb, Sb, Zr and Sn, have been used as coating as well (Chen and Chen, 2005). Ho and Chan (1986) employed PbO₂-Ti anode for electroflotation treatment of palm oil mill effluents. Also, treatment of radioactive wastewater using titanium anode with coating of isomorphous titanium and ruthenium oxides was reported by Il'in and Kolesnikov (2001). These electrodes are expensive and have short service life. In recent years, studies have been performed on production of cheaper DSA electrodes with longer service lives. Ti/IrO₂-Ta₂O₅ anodes were used for treatment of synthetic wastewater containing oil and peptides (Mraz and Krýsa, 1994). More complex electrodes made of Ti/IrO_x-Sb₂O₅-SnO₂ and Ti/RuO₂-Sb₂O₅-SnO₂ have demonstrated significantly longer service lives (Chen et al., 2002; Chen and Chen, 2005).

Active anodes made of metals such as Al and Fe have also been employed for treatment of industrial wastewaters, e.g., urban wastewater (Pouet and Grasmick, 1995), slaughterhouse wastewater (Bayramoglu et al., 2007), textile wastewater (Aouni et al., 2009), leachate of oil-drilling (Ighilahriz et al., 2014) and dairy effluents (Bassala et al., 2017). During the process, metal ions are released from the 'sacrificial anodes' and react with hydroxide ions, forming metal hydroxides such as Al(OH)₃, Al(OH)₄⁻, Fe(OH)₃ and polymeric species such as Al₂(OH)₂⁴⁺, Al₆(OH)₁₅³⁺, Fe(H₂O)₆³⁺ and Fe(H₂O)₅OH₂⁺ (Aouni et al., 2009; Kim et al., 2002). These metal hydroxides act as coagulant and adsorb colloidal particles, form bonds and create aggregates, rise to surface and remove the particles from water.

2.3.2 Electrode Arrangement

Simple electroflotation systems comprise a reactor and electrodes (anode and cathode) connected to a DC power supply. The connection mode of electrodes to DC power supply can be monopolar (parallel or series) or bipolar. While in monopolar-connection mode all electrodes are connected to each other or to DC power supply, only outmost electrodes connect to power supply in bipolar connection mode. Schematic of different electrode connection modes are depicted in Figure 2.1.

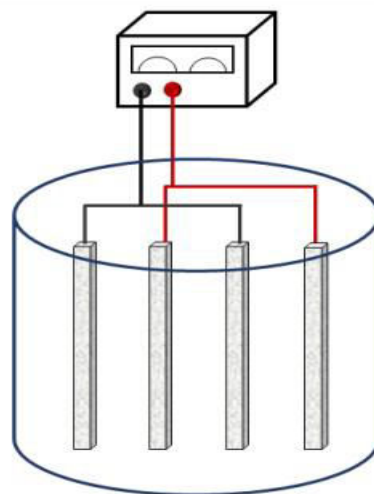


[Fig.2.1: Schematics of electrodes connection modes: (a) Bipolar, (b) Monopolar series, (c) Monopolar parallel]

Comparative studies of electrodes connection modes have been conducted by different researchers. The monopolar connection mode is considered more advantageous than the bipolar mode, in terms of separation efficiency and power consumption (Daneshvar et al., 2004; Golder et al., 2007; Modirshahla et al., 2007; Kobya et al., 2007; Ghosh et al., 2008; Solak et al., 2009).

2.4 Reactor Design

A design of electroflotation reactor includes the electrodes arrangement, reactor shape and flow regime inside the reactor. Most lab/bench-scale electroflotation experiments have been performed in batch-flow processing regime. They usually consist of a small cell as the reactor and a few electrodes. Figure 2.2 presents a basic and simple design of a batch electroflotation reactor with vertically-oriented monopolar electrodes. Other batch reactors have been presented by researchers as well.



[Fig.2.2: Schematics of a simple batch electroflotation system with vertical monopolar electrodes]

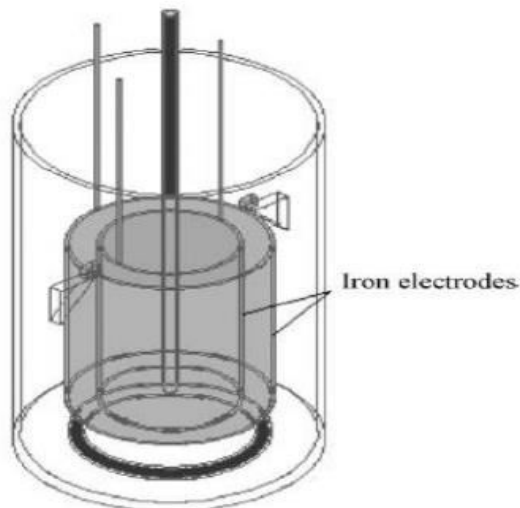


Figure 2.3 shows a cylindrical aluminum reactor which works as the anode, and an aluminium impeller cathode, used for defluoridation (Un et al. 2013).

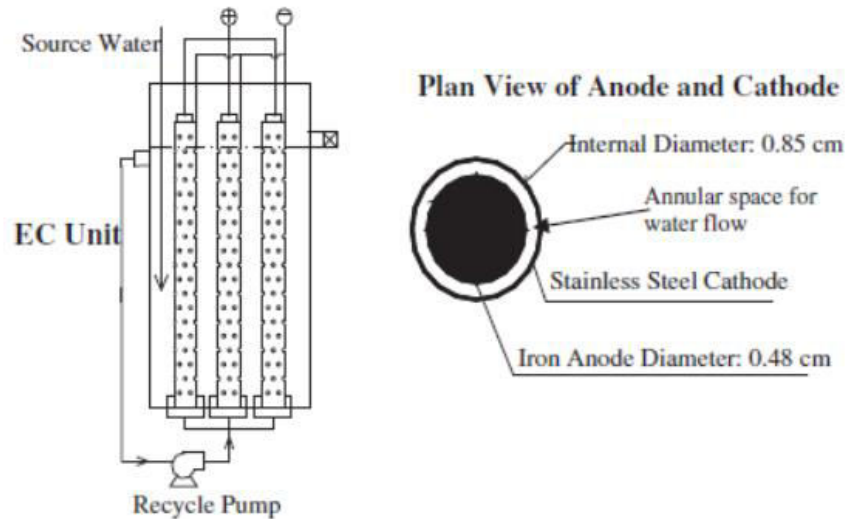


[Fig.2.3: Batch cylindrical reactor with rotating cathode electrode (Un et al., 2013)]

In another study, Nunez et al. (2011) used iron cylinders with different diameters as anode and cathode in a cylindrical acrylic cell, Figure 2.4, for arsenic removal from wastewater.

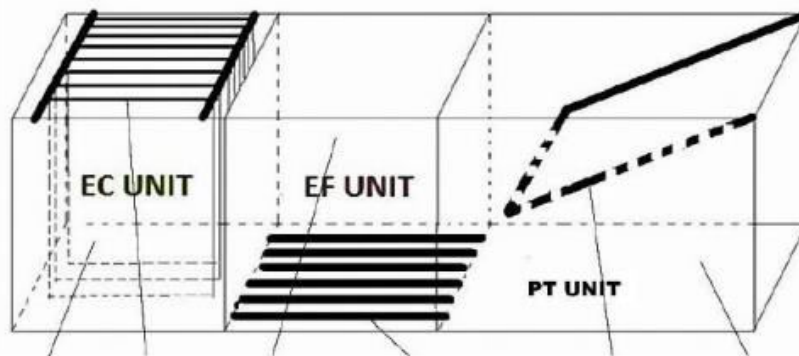


[Fig.2.4: Batch acrylic cylindrical cell with iron cylinder electrodes (Nunez et al., 2011)]



[Fig.2.5: Batch reactor with cylindrical stainless steel cathodes and rod-shaped iron anodes (Lakshmanan et al., 2010)]

Studies of continuous-flow reactors often involve larger reactors and are the next step before the full-scale design of the process. Hassani et al. (2016) designed a 3-stage continuous-flow reactor including electrocoagulation (release of metal ions from sacrificial anode) unit with Al and Fe electrode plates, and electroflotation unit with stainless steel cathodes and Ti/RuO₂ anodes followed by precipitation unit to capture the remaining solids from landfill leachate, Figure 2.6.



[Fig.2.6: Schematic of 3-stage electrochemical reactor. Electrocoagulation Unit, Electroflotation Unit, Precipitation Tank (Hassani et al., 2016)]

Research Summary 1

Fluorides Removal by Electro-coagulation Method Using AC and DC Current In Water Treatment; [Milad Mousazadeh , S. M. Alizadeh , Zacharias Frontistis ;2021]; Fluoride ions present in drinking water are beneficial to human health when at proper concentration levels (0.5–1.5 mg L⁻¹), but an excess intake of fluoride (>1.5 mg L⁻¹) may pose several health problems. In this context, reducing high fluoride concentrations in water is a major worldwide challenge. The World Health Organization has recommended setting a permissible limit of 1.5 mg L⁻¹.



The application of electrocoagulation (EC) processes has received widespread and increasing attention as a promising treatment technology and a competitive treatment for fluoride control. EC technology has been favourably applied due to its economic effectiveness, environmental versatility, amenability of automation, and low sludge production. This review provides more detailed information on fluoride removal from water by the EC process, including operating parameters, removal mechanisms, energy consumption, and operating costs. Additionally, it also focuses attention on future trends related to improve defluoridation efficiency.

Research Summary 2

Effects of alternating and direct current in electrocoagulation process on the removal of fluoride from water; [Subramanyan Vasudevan, Balasingam Suresh Kannan, Jothinathan Lakshmi, Subramanian Mohanraj and Ganapathy Sozhan; 2011]; When direct current (DC) is used in electrocoagulation processes, an impermeable oxide layer may form on the cathode and corrosion of the anode may occur due to oxidation. This prevents effective current transfer between the anode and cathode, so the efficiency of the electrocoagulation process declines. These disadvantages of DC have been reduced by adopting alternating current (AC). The main objective of this study is to investigate the effects of AC and DC on the removal of fluoride from water using an aluminum alloy as anode and cathode. Results showed that removal efficiencies of 93 and 91.5% with energy consumption of 1.883 and 2.541 kWh kL⁻¹ was achieved at a current density of 1.0 A dm⁻² and pH 7.0 using an aluminum alloy as electrodes using AC and DC, respectively. For both AC and DC, the adsorption of fluoride fitted the Langmuir adsorption isotherm. The adsorption process follows second-order kinetics and temperature studies showed that adsorption was exothermic and spontaneous in nature.

Research Summary 3

Effects of Alternating Current (AC) and Direct Current (DC) in Electrocoagulation Process for the Removal of iron from Water ; [Subramanyan Vasudevan; 2012]; In practice, direct current (DC) is used in an electrocoagulation processes. In this case, an impermeable oxide layer may form on the cathode as well as corrosion formation on the anode due to oxidation. This prevents the effective current transfer between the anode and cathode, so the efficiency of electrocoagulation processes declines. These disadvantages of DC have been diminished by adopting alternating current (AC) in electrocoagulation processes. The main objective of this study is to investigate the effects of AC and DC on the removal of iron from water using zinc as anode and cathode. The results showed that the optimum removal efficiency of 99.6% and 99.1% with the energy consumption of 0.625 and 0.991 kWh kL⁻¹ was achieved at a current density of 0.06 A dm⁻², at pH of 7.0 using AC and DC, respectively. For both AC and DC, the adsorption of iron was preferably fitting Langmuir adsorption isotherm, the adsorption process follows second order kinetics and the temperature studies showed that adsorption was exothermic and spontaneous in nature.

Research Summary 4

Taguchi experimental design for electrocoagulation process using alternating and direct current on fluoride removal from water; [Ghader Ghanizadeha, Gholamreza Shariati neghabb , Mohammad Salemc , Kazem Khalagid; 2015]; Taguchi experimental design with the alternating and direct current (AC and DC) electrocoagulation (EC) using initial fluoride concentration of contaminant, retention time, pH, electrode type, and voltage was used on fluoride removal. DC current is significantly more efficient than AC, and Al has a higher performance than Fe electrode ($p < 0.05$). Neutral pH is more effective than acidic and alkaline condition in which significant differences are observed in reaction time of 60 min ($p = 0.02$). Elevation of cell potential from 15 to 25 V led to enhancing efficiency, but beyond these range fluoride removal declined. Fluoride concentration increase from 3 to 8 mg/l declined the EC performance with a significant impact in reaction times of 20 and 40 min ($p = 0.004$). In similar cell potential, increasing in reaction time from 20 to 60 min led to decreasing in residual fluoride from 1.75 to 1.06 mg/l, and 1.64 to 0.76 mg/l in 25 and 40 V, respectively.

Research Summary 5

Effects of alternating and direct current in electrocoagulation process on the removal of cadmium from water ; [Subramanyan Vasudevan, Jothinathan Lakshmi, Ganapathy Sozhan; 2011]; In practice, direct current (DC) is used in an electrocoagulation processes. In this case, an impermeable oxide layer may form on the cathode as well as

corrosion formation on the anode due to oxidation. This prevents the effective current transfer between the anode and cathode, so the efficiency of electrocoagulation processes declines. These disadvantages of DC have been diminished by adopting alternating current (AC) in electrocoagulation processes. The main objective of this study is to investigate the effects of AC and DC on the removal of cadmium from water using aluminum alloy as anode and cathode. The results showed that the removal efficiency of 97.5 and 96.2% with the energy consumption of 0.454 and 1.002 kWh kl^{-1} was achieved at a current density of 0.2A/dm² and pH of 7.0 using aluminum alloy as electrodes using AC and DC, respectively. For both AC and DC, the adsorption of cadmium was preferably fitting Langmuir adsorption isotherm, the adsorption process follows second order kinetics and the temperature studies showed that adsorption was exothermic and spontaneous in nature.

III. PROPOSED METHODOLOGY

3.1 REAGENTS USED

- Standard fluoride solution.
- SPADNS Solution: 958mg is used in distilled water. The solution is stable for at least 1 year if protected from direct sunlight.
- Acid Zirconyl Reagent
- Sodium Arsenate
- HCL



[Fig.3.1: A- Spands Solution, B- Sodium Arsenate, C- HCL, D- Sodium Flouride]

3.2 MECHANISM OF ELECTROCOAGULATION

The electrochemical technique might provide the variety of the unwanted dissolved particles where the suspended matter can effectively managed with the aqueous solution. The process requires a steady pass through which the anode and cathode rods might have some liquidated range. The aluminum electrodes include the solution at anode and hydrogen gas at cathode. It has also dissolved the AI anodes to the aluminum species where the coagulating agent combines to provide the pollutants of the large size flocs. The suspended particles might collide as the tank might have some colloidal range to which the surface of the water can be entertained. The pollutants basically present the electro-coagulation and electro-flotation measures in order to gain the electrolytic cells over cathode and anode respective to the features. The dynamic shift in the process provides the dominant of the process. The



sacrificial electrode might include the different material where the anode can gain the interactive range to which the electro-coagulation can occurred.

3.3 METHODOLOGY

- The Conductive Metal Plates are commonly known as – sacrificial electrodes. It lower the dissolution potential of the anode and minimizes the passivation of the cathode.
- A batch pilot-scale EC reactor with electrodes connected to the EC reactor with monopolar parallel connection mode with two anodes and two cathodes electrodes was designed and constructed for fluoride removal.
- Fluoride stock solution was prepared by dissolving 221 mg anhydrous sodium fluoride (NaF) in 1,000 ml distilled water in volumetric flask and desired concentrations.
- After electrolyte preparation with the appropriate fluoride concentration and pH adjustment, the EC process with AC or DC current was started while complete mixing of the reactor constituent was done via magnetic bar stirrer.
- Fluoride measurement was conducted based on spectrophotometric technique on with SPANDS method.

IV. CONCLUSION

The electro-coagulation (EC) process proved to be an efficient method for the removal of fluoride from ground water. The experimental process that's accounted will help in providing the reduction details of the percent of the fluoride compound within the water and also provide crystal clear water for drinking. However, it's predicted that complete removal isn't possible. Electrocoagulation technology has achieved grade of applicability and hence examined as potential defluoridation technology. It is highly compared with traditional flocculation–coagulation, electrocoagulation has the advantage of removing the tiniest colloidal particles and comparatively low amount of residue generation. In spite of getting numerous advantages, EC has some drawbacks like the periodic replacement of sacrificial anodes. Also it requires a minimum conductivity reckoning on reactor design that limits its use with water containing low dissolved solids. Additionally the control of residual aluminium, technical and economical optimization of the method per the standard of water being treated must be taken into considerations. Although large numbers of studies are available in literature further studies are required to elucidate the potential and limitations of the method. so as to proportion the strategy, the longer term work should be focused on investigating the influence of assorted operational parameters including duration, energy consumption and initial fluoride concentration in addition as interference from factors like calcium ions in effluents.

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