

An Experimental Study of Electrocoagulation Process Applied For Influence of Fluoride Ions on Hardness Removal

P.Mahammedrafi,

Asst. Professor, Department of Civil Engg, Mallareddy Institute of Technology and Sciences, Hyderabad, Telangana.

ABSTRACT:

This paper demonstrated influence of fluoride ions on hardness removal and simultaneous removal efficiency from water by Electrocoagulation process. The process parameters such as initial pH, applied voltage, DO level and Electrolysis time were studied. Electrocoagulation test results revealed that a maximum hardness removal 60-70% for the concentrations of hardness of 800, and 1000 mgL-1 and a removal of 91% for fluoride concentration of 5 mgL-1 investigated at an applied voltage of 15 V and an electrolysis time of 120 minutes. Hardness removal by Electrocoagulation majorly involves adsorption on to iron flocs, while fluoride replaces a hydroxyl group from iron aggregates. In this paper, influence of fluoride ions on hardness removal was investigated and gave a marginal increase in hardness removal. This paper also include the important parameter DO level that plays a key role in the electrocoagulation process.

Keywords: Hardness and fluoride removal, DO level, Influence of fluoride ions, electrocoagulation.

I. INTRODUCTION

Water as a precious resource is being used for several purposes by humans but the purity of water being consumed is very important since it has a direct effect on health. To get such a purity at which no contaminants were able to found find the ways to develop simple, effective, low-cost and easy to use technologies able to reduce organic, inorganic and microbiological water contamination. Nowhere on the Globe pure water is found. Therefore water in its life gets contacted with other impurities like hardness agents, fluorides, bacteria, other toxic substances among the impurities.

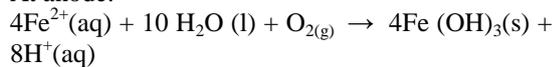
Hardness in water caused by existing divalent ions, such as salts of calcium and magnesium. However, calcium and magnesium are known as the dominant species for water hardening⁽¹⁾. Although it has been seen that water hardness doesn't have serious health impact, it has been demonstrated that hard water is responsible for scale deposits. In general, water sources with hardness higher than 200 mg/L are unaccepted by consumers but are considered as poor resources. WHO Recommendation for drinking water's hardness is based on maximum 500 mg/l calcium carbonate⁽¹⁾.

Fluoride in water is usually one of the elements essential for humans. Naturally it occurs in geochemical deposits, minerals, and natural water systems and gets involved in food chains with drinking water, plants and cereals. Although the fluoride present in drinking water is essential for human health, an excessive intake of fluoride causes severe dental or skeletal fluorosis⁽²⁾. The suitable level of fluoride in drinking water specified by the World Health Organization (WHO) is 1.5 mg L-1⁽³⁾.

Electro-coagulation is a simple and effective method to remove the flocculating agent generated by electro-oxidation of a sacrificial anode and generally made up of iron or aluminum. In this process, the treatment is performed without adding any chemical coagulant or flocculants. Thus, reducing the amount of sludge which must be disposed. Electrocoagulation is based on the in situ formation of the coagulant as the sacrificial anode corrodes due to an applied current, while the simultaneous evolution of hydrogen at the cathode allows for pollutant removal. This process combines three main interdependent processes, operating synergistically to remove pollutants namely, electrochemistry, coagulation and

hydrodynamics. Chemical reactions occurring at the electrodes are:

At anode:



At cathode:



The iron hydroxide flocs normally act as adsorbents and/or traps for removal of hardness and fluoride ions from the solution. The electrocoagulation process can be described as follows: at the anode, the dissolved metallic ionic species destabilize the colloids; besides that, this contributes further to their precipitation and at the cathode, the obtained hydrogen bubbles (resulted as a product of water electrolyses) capture the dispersed particles that are then separated by flotation (Bebeşelea et al., 2006; Miron et al., 2010).

Many investigators were studied hardness removal from various sources with electrocoagulation process and the results they found were tabulated below (Table 1). Fluoride removal in electrocoagulation process was also studied and results were tabulated. With a focus of influence of fluoride ions on hardness removal, the present study is investigated.

Previous works

Table 1 summarizes literature hardness and fluoride removed by electro coagulation in water sources.

Reference	Pollutants	Current or Current density	Cell voltage	Electrode material, Electrode connection	Treatment efficiency, %	Mode
Malakootian, M. and N. Yousefi,	Tap water		20 V	Al/Al plate	80.6 %	Continuous
Sanfan and Qinlai	Brackish water hardness	2–30 A/m ²	1.3 – 3.6	Al/Fe plate	60 - 80%	Continuous
Daneşhvar et al.	Brackish water	1–22 A/m	1.1-3.1	Al/Fe plate	40 – 76%	Continuous
Saravanan et al.	Groundwater		5 - 20 V	Al plate	60 %	Batch
Khairi et al.	Tap water		10 - 28.5V	Al and graphite plate	85%	batch process

M.M. Emamjomeh et.al.	synthetic water	6.25 A/m	0-30 V	Aluminum plate	92.7 %	batch process
-----------------------	-----------------	----------	--------	----------------	--------	---------------

The contribution of this paper was to investigate the influence of fluoride ions on hardness removal and also fluoride removal employing Electrocoagulation process and also other parameters such as DO level, applied voltage, electrolysis time and pH.

II. MATERIALS AND METHODOLOGY

Sodium Fluoride (NaF), Calcium chloride fused (CaCl₂), Magnesium chloride hexahydrate (MgCl₂.6H₂O), Calcium sulphate dehydrate (CaSO₄) and Magnesium sulphate heptahydrate (MgSO₄.7H₂O) chemicals are analytical grade (Merck). Specifications of iron metal plate electrode are 5 cm x 10 cm and thick 3 mm.

In Electrocoagulation test, Iron electrodes were used as the anode and cathode. Both electrodes were square in shape (100 mm side), each with a geometric area of 50 cm² and with an electrode gap of 12 mm. The electrical current flow of 1 A was applied using a DC power supply having a variable output of 0–32V (10 and 15 V for this study) as direct current source. Electrodes were connected as monopolar with the help of a piece of wood. Test solutions of desired concentrations of hardness and fluoride with pH adjusted to required values were injected into the reactor. Total hardness and fluoride by EDTA Titrimetric method and SPADNS method using UV-VIS Spectrophotometer at 570nm (APHA/AWWA/WEF, 1999).

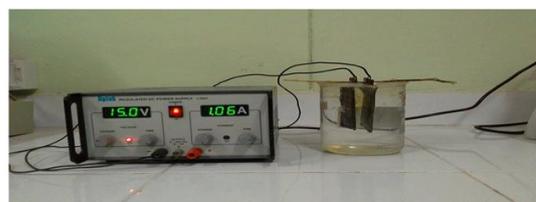


Fig 1. Electrocoagulation Experimental Setup

III. RESULTS AND DISCUSSIONS

Optimization of process parameters

This section includes information related to effects of various parameters such as electrolysis time, initial fluoride and Hardness concentration,

pH of solution and applied voltage which will be studied in the desired range as shown in Table 2 to evaluate their effect on simultaneous removal efficiency and influence of fluoride ions as well.

Table 2: Parameter optimization specifications

Sl No.	Parameters	Remarks
1	Initial fluoride concentration	5 mg/l
2	Hardness concentration	800 & 1000 mg/l
3	Electrolysis time(ET),	120 minutes
4	pH	6,7,8
5	Voltage	5-15 V

Electrolysis time is one of the important parameters of Optimization of EC runs, which It primarily involves determining the optimum residence time for removal of hardness. Electrolysis time along with applied voltage decides the coagulant dosage (Fe³⁺ concentration) and rate of bubble production. In the study Electrolysis time for various Hardness concentrations (800 and 1000 mg/l) was studied. This is because as the time progresses the dissolved coagulants from the iron electrode increased according to Faraday's law. A sufficient amount of coagulant dissolved from iron electrode adsorbed Hardness ions and thus higher removal efficiency for 120 minutes duration was observed as shown in Fig.1 and Fig.2. This is because In dilute solutions, formation of the diffusion layer has no effect on rate of diffusion or migration of metal ions to the electrode surface⁽⁴⁾. Most of 30 - 50% of Hardness was removed in the first 30 - 60 minutes and was observed that hardness removal efficiency increases with increasing concentration to an electrolysis time of 120 minutes as shown in Fig.1 and Fig.2.

Effect of pH:

The Electrocoagulation process depends on pH of solution. It has been established from previous studies that pH has a considerable effect on efficiency of EC process as the rate of coagulation depends on pH of solution. In this study, pH is varied in the range 7 - 8 in an attempt to investigate the effect of pH on Hardness and

fluoride removal individually as well as simultaneously with increasing Electrolysis time as shown in Fig.1 and Fig.2. It was clearly found that an initial pH 8 has got better hardness removal as compared to an initial pH of 7.

During the process it was observed that there was a considerable change in pH post treatment i.e. an increase in pH was identified and was being an uncontrolled factor⁽⁵⁾. This is because the fact that production of hydrogen bubbles and pH shift occurs towards alkaline conditions and for higher initial pH of 8, the final pH reaches a value of 11 which is way beyond the limit for drinking water⁽⁵⁾.

Effect of Applied Voltage:

The voltage applied has a direct effect on bubble production rate and floc growth because of its strong influence on current density or applied voltage. Therefore it has a significant influence on hardness and fluoride removal efficiency. It was found that for a given time, the removal efficiency increased significantly with the increase in applied voltage as shown in Fig.1 and Fig.2.

About 25 - 60 % of the hardness removal was observed in 120 minutes of Electrolysis time at 10 and 15V for different initial pH of 7 and 8 respectively as shown in Fig.1 and Fig.2. This is because at higher voltage the amount of metal hydroxides increases, thus therefore greater amount of coagulant is released for precipitation of hardness and/or fluoride.

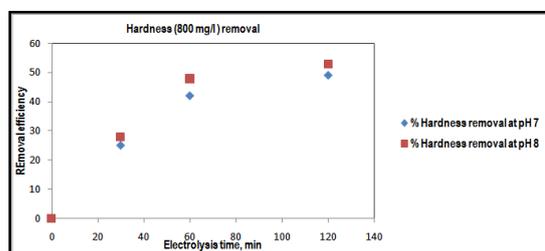


Fig 1. Hardness removal at applied voltage of 10 V

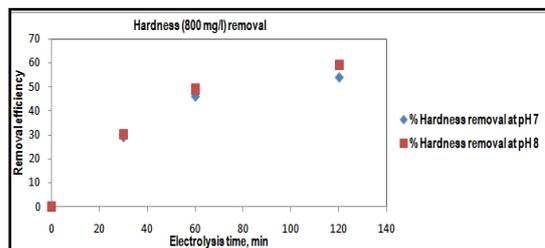


Fig 2. Hardness removal at applied voltage of 15 V

From the study it was then noted that The EC process efficiency was gradually followed reduction after electrolysis time reached to 120 minutes. The process parameters such as initial pH 8, applied voltage 15 V and electrolysis time of 120 minutes were optimized for further studies.

Effect of Dissolved Oxygen

It was observed that Dissolved oxygen has a considerable effect on bubble production and floc distribution of ferric ions and also determines the oxidation of ferrous ions to ferric ions. From the study carried out, as the electrolysis time increases, Dissolved oxygen required for oxidation of iron was decreased gradually. This however, indicated that the simultaneous removal of hardness and fluoride was rapid for an electrolysis time of 120 minutes due to the fact that the test solution has a reasonable Dissolved oxygen level and then the simultaneous removal follows a constant or declined path as shown in the Fig.3 and Fig.4.

It was concluded from the previous studies that In electrocoagulation, If iron electrodes undergoes oxidation and however the generated Ferrous ions will immediately undergo further spontaneous reactions to produce corresponding hydroxides and/or polyhydroxides⁽⁶⁾. These metal hydroxides remove hardness and fluoride ions by the mechanism of rapid adsorption of hardness and fluoride ions.

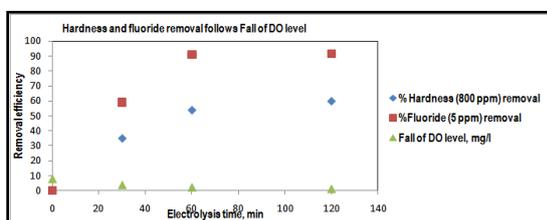


Fig.3 Simultaneous hardness and fluoride removal at applied voltage of 15 V (pH 8)

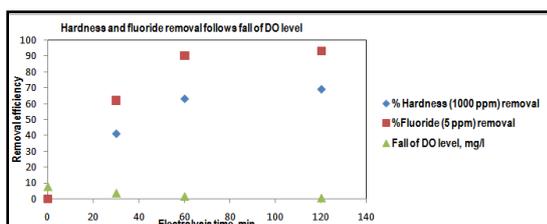


Fig.4 Simultaneous hardness and fluoride removal at applied voltage of 15 V (pH 8)

Influence of Fluoride ions

In the present study, electrocoagulation experiments were conducted to determine the influence of fluoride ions on hardness removal at an applied voltage of 15V and initial pH 8. Hardness and Fluorides concentrations of 800 mgL-1 and 5 mgL-1 simultaneously were used and Hardness and Fluorides concentrations of 1000 mgL-1 and 5 mgL-1 simultaneously were used and the results are presented in Figure.5 and Fig.6. It was followed that the hardness removal increases if the test solution contain fluorides of 5 mg/l as compared to no fluorides.

It follows from the study that fluoride removal increases if electrolysis time increases to a certain time and after which fluoride removal decreases. In the anodic adsorption layer, the concentration of hydroxyl ions is higher than the concentration of F-, so that the production of the complex Ferric hydroxide flocs becomes difficult; and this results in the reduction of fluoride removal from water.

By considering hardness, removal increases gradually in a duration of 60 minutes and no removal (can be neglected) can be expected from an electrolysis time of 120 minutes. For the study the applied voltage 15V and initial solution pH 8 was selected. The influence of pH on adsorption can explain the slower rate of pollutant removal with increasing pH. The present study confirms this fact, wherein in the beginning of the process the removal was rapid and thereafter becomes slower due to the continuous evolution of hydrogen ions from the cathode; hence pH increase over the time⁽⁶⁾.

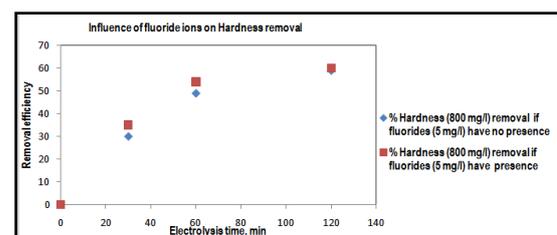


Fig.5 influence of fluoride ions on hardness removal at applied voltage of 15 V (pH 8)

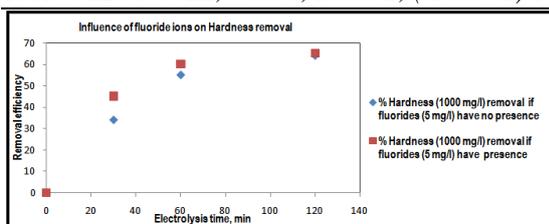


Fig.6 influence of fluoride ions on hardness removal at applied voltage of 15 V (pH 8)

Simultaneous removal by Electrocoagulation:

Electrocoagulation experiments were conducted to investigate simultaneous removal of hardness and fluorides employing different initial hardness of 800, 1000 mg L⁻¹ and fluoride concentration of 5 mgL⁻¹. Applied voltage of 15 V was applied for different contact times of 30, 60, and 120 minutes. Experimental results indicated that removal of hardness 800 mgL⁻¹ concentration at voltage 15V in a test solution of pH 8 and at different contact times of 30, 60, and 120 minutes are presented in Fig.7 and Fig.8.

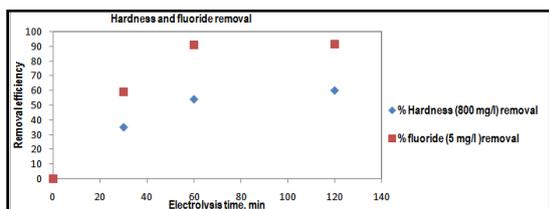


Fig.7 Hardness and fluoride removal at applied voltage of 15 V (pH 8).

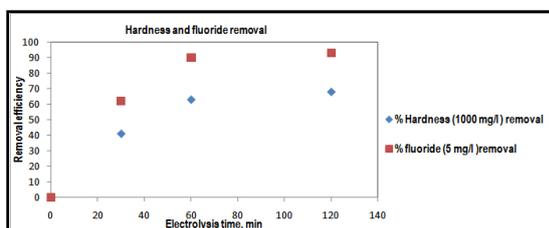


Fig.8 Hardness and fluoride removal at applied voltage of 15 V (pH 8).

It however can be seen from Fig.7 and Fig.8 that as the electrolysis time increases, hardness removal also follows a marginal increase. However by considering Fluorides, removal increases up to certain electrolysis time reaching an optimum time after which Fluoride removal decreases. As the applied voltage was being 15V, removal of both fluorides and hardness values increased at considered electrolysis times. It was

found that the removal by electrodes was primarily responsible for hardness and fluoride removal, and the adsorption by ferric hydroxide flocs gave a maximum removal.

IV. CONCLUSION

The present study was attempted to investigate influence of fluoride ions on hardness removal and simultaneous removal of hardness and fluoride from synthetic solutions by Electrocoagulation process. The optimized parameters such as initial pH, electrolysis time and applied voltage were investigated. Electro coagulation was found effective in simultaneous removal of hardness (60% and 70% for 800 and 1000 mg/l concentrations respectively) and fluorides (91% for 5 mg/l) for an electrolysis time of 120 minutes at applied voltage 15V.

Electro coagulation results also indicated that the hardness removal increases if the test solution contains fluorides of 5 mg/l as compared to no fluorides. From the study carried out, as the electrolysis time increases, Dissolved oxygen (DO level) required for oxidation was decreased (i.e 8 mg/l to 1.0 mg/l nearly) gradually. This However indicated that the simultaneous removal of hardness and fluoride was rapid for an electrolysis time of 120 minutes and then follows a constant or declined path as no a reasonable Dissolved oxygen level.

REFERENCES

- [1]. M. Yan, D. Wang, J. Ni, J. Qu, Y. Yan, C.W.K. Chow, Effect of polyaluminum chloride on enhanced softening for the typical organic-polluted high hardness North-China surface waters, Separation and Purification Technology. 62 (2008) 401–406.
- [2]. WHO (World Health Organization), "Guidelines for drinking water quality: Health criteria and supporting information," vol.3, 1984.
- [3]. Nemerow N L, Agardy F J, Sullivan P and Salvato JA. Environmental engineering, water, wastewater, soil and ground water treatment and remediation, 6th Edition, 2326, 2009. 10
- [4]. Sailaja K., Jyotsana C., Raman N., Himabindu V., Bhagwan D., " Effective

- removal of fluoride from groundwater using electrocoagulation,” International journal of engineering research and application, vol.4, pp-439-445, 2014.
- [5]. P. Mahammedrafi, J. Karthikeyan, Simultaneous removal of hardness and fluoride from water by electrocoagulation and chemical coagulation”, International journal of innovative research in science , engineering and technology, Vol. 6, Issue 10, October 2017.
- [6]. M. Yousuf A. Mollah, Robert Schennach, Jose R. Parga, David L. Cocke, “Electrocoagulation (EC)---- sciences and applications” Journal of Hazardous Materials , pp 29-41,2011.
- [7]. Kumar, P.R.; Chaudhari, S.; Khilar, K.C.; Mahajan, S.P. Removal of arsenic from water by electrocoagulation. *Chemosphere* 2004, 55, 12451252.

Marcio R. Martini Worker-Qol 76 – An Instrument For Evaluating Workers' Quality Of Life: A Description Of Its Construction And Key Psychometric Properties.” *International Journal of Engineering Research and Applications (IJERA)* , vol. 8, no. 02, 2018, pp. 77-92.