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Assessment of fluoride removal in a batch electrocoagulation process: A case study in the Mount Meru Enclave.

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ABSTRACT

The presence of excessive amounts of fluoride than prescribed standards has been reported in various sources of domestic water supply around the slopes of Mount Meru and other parts in Tanzania. Efforts to remove the excessive fluoride have been carried out using various technologies. In this study, electrocoagulation experiments were carried out to assess its efficiency on fluoride removal. The fluoride concentration tested ranged from 1.37 to 48 mg/L in both synthetic and natural waters. The voltage applied in the electrocoagulation (EC) process ranged from 0 to 50 V while maintaining pH values of 4 to 9. The representative experimental results for the Ngarenanyuki river water with initial fluoride concentration of 29.5 mg/L accomplished a removal efficiency of 90% at an optimal electrolysis time of 30 min, an applied voltage of 30 V and an optimal pH of 6. The method showed efficient fluoride removal in water to allowable limits by World Health Organization (WHO) and Tanzania Bureau of Standards (TBS) (1.5 mg/L). Despite the voltage applied (30 V), the pH at neutrality remained unchanged thus making the process more efficient. At this voltage (30 V) the process has been reported previously also to have the capability of disinfecting the water and hence rendering such water safe for use.

Introduction

Depending on its concentration, fluoride in water can have both beneficial and harmful effects on the environment and human health. The concentration of fluoride around 1 mg/L in drinking water can help in teeth decay prevention [29]. However, the long-term consumption of water containing an excess of fluoride can lead to fluorosis of teeth and bones [28,29]. Globally, fluoride has become a source of some health concern when consumed either at very low or very high concentrations. A fluoride concentration of 1.5 mg/L is the amount recommended by the World Health Organization [28] and the Tanzania Bureau of Standards [26] in drinking water. Exposure to lesser concentrations than this recommended amount can lead to tooth decay; whiles prolonged exposure to greater concentrations does have harmful effects such as dental fluorosis, retarded growth of children, and skeletal defects [17,29].

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The contamination of water with fluoride can occur naturally in regions with geological deposits of marine origin and at the foot of volcanic mountains such as the mount Meru considered in this present study [30]. Furthermore, groundwater gets polluted due to deep percolations from intensively cultivated fields due to various ecological factors either natural or anthropogenic, liquid and solid wastes from industries, disposal of hazardous wastes, sewage disposal, etc. Drinking water in Tanzania is reported to have a fluoride concentration of 20 mg/L and beyond in some areas under the Mount Meru slopes [5]. This is way above the recommended levels making defluoridation of drinking water necessary for human health.

Various methods for defluoridation from water such as adsorption, coagulation and flocculation, ion exchange, reverse osmosis, chemical sedimentation, electrodialysis, electrocoagulation (EC), and nanofiltration have been under investigation [5,11,21,27]. Chemical coagulation requires a huge amount of chemicals and produces large volumes of sludge which makes it very costly and undesirable. Other membrane filtration technologies such as electrodialysis, reverse osmosis, and nanofiltration requires energy, high maintenance costs and produces a very concentrated sludge [2,22,27]. The Bone char defluoridation technique is the method that is widely applied in Tanzania but it requires high maintenance cost and social-cultural issues which limits its application [18].

Quite recently, there have been numerous studies concentrated on electrocoagulation (EC), which is an effective process used to destabilize and remove finely dispersed particles like fluorides from water and wastewater. Electrocoagulation is a simple electrochemical process in which voltage is applied across two or more metal electrodes (*aluminum or iron*) in an EC reactor containing water and coagulants in the form of metal cations, which are generated in-situ by the dissolution of the sacrificial anode. [13]. Mixing of the coagulant leads to flocculation and precipitation that results in the removal of dissolved and suspended pollutants. During the same process (see fig.SM2a), hydrogen is generated at the cathode and oxygen at the anode, resulting in further removal of pollutants by flotation. The reactions that occur on aluminum electrodes are illustrated (*see fig.SM2a*) in equations 1–3:

The oxidation reaction that takes place at the anode,

$$\mathbf{Al{-}3e^- \rightarrow Al^{3+}} \tag{1}$$

The reduction reaction that takes place at the cathode,

$$\mathbf{2H_2{}^+ + 2e^- \rightarrow H_2} \tag{2}$$

The hydrolysis reaction,

$$\mathbf{Al^{3+} + 3H_2O \rightarrow Al\,(OH)_3 + 3H_2{}^+} \tag{3}$$

Aluminum ions react with water to generate solid aluminum hydroxides. Flocks are formed by the precipitates that combine with water contaminants, which depend on the metal hydroxides formed by hydrolysis [12]. These coagulants aggregate suspended particles and adsorb the fluorides (F) to form the aluminum complexes. $Al(OH)_3$ flocks are believed to adsorb F strongly as indicated in Eq. (4).

$$\mathbf{Al\,(OH)_3 + \mathit{x}F \rightarrow Al\,(OH)_{3-\mathit{x}}\,F_{\mathit{x}} + \mathit{x}OH} \tag{4}$$

In comparison, coagulant salt is added in conventional coagulation and pollutants are removed by flocculation, and settling only. Electrocoagulation has several other advantages, such as electro-oxidation, higher efficiency, in situ generation of coagulant ions, lower sludge production, no mechanical parts, and low operation and maintenance costs. Moreover, it is a self–sustained technique that does not require filter/ membrane exchange since it can operate with automatic backwash unlike other techniques applied in Tanzania. In conventional coagulation (CC), the pH of the water decreases, requiring post-coagulation neutralization. Conversely, in EC, the pH of the water can increase, decrease, or remain neutral depending on the nature of the contaminant and initial pH [1,6,24]. The floc formed by EC is relatively large and contains less bound water. They also are more stable and, therefore, responsive to filtration [4]. Electrocoagulation has been widely applied for the purification of wastewater and leachate, but its use in the field of drinking water is relatively limited. Electrocoagulation has been used to remove a wide variety of pollutants from water and wastewater, including suspended solids, fluoride, chromium, nitrate, phosphate, arsenic, hardness, algae, oils and greases, dissolved organic carbon, several types of dyes, and for desalination.

The Electrocoagulation technique requires good maintenance of the aluminum anodes to ensure effective fluoride removal. Furthermore, EC has also been shown to be effective in disinfection as well. According to [7] it shows that electrocoagulation using aluminum– electrodes has been effective in disinfection with a significant removal efficiency of 98% with an increase in time at an applied voltage of 100–200 mV. The *E.coli* eliminating performance of 99.8% was also observed at an electrolysis time of 2 min at an applied voltage of 4 V [15] which provide safe drinking water.

This study aimed at investigating and optimizing the Electrocoagulation process for fluoride removal from selected sources of domestic water supply around the Mount Meru areas in Arusha- Tanzania *(see fig.SM1)*.

Materials and methods

Synthetic and natural water

In this study, batch experiments were carried out on water samples by varying current, electrodes set up, and fluoride concentrations. Water samples from different sources were collected; and fluoride concentrations ranging from 1.37–48 mg/L were used. Synthetic fluoride solutions were prepared by dissolving 3, 7, and 10 mg/L of sodium fluoride to make different solutions using distilled water. Tap water with 1.37 mg/L was also used. Domestic water supply source samples from Bulebule spring (7.05 mg/L), Bulem spring (8.05 mg/L), River Ngarenanyuki (29.5 mg/L), and River Uluwile (48 mg/L) were collected around the Mount Meru areas in Arusha- Tanzania *(see fig.SM1)* and parameters such as pH, fluoride concentration, and conductivity were analyzed.

Experimental setup

The electrocoagulation (EC) experiment was accomplished in a double-walled (beaker) with 300 mL of water as shown in fig. SM2b. Two electrodes of 95% pure aluminum alloy with dimensions 1×5 cm^2 were submerged in the solution with a 2 cm electrode gap. A GW-Instek GPR-3510HD laboratory power supply system was used to provide direct current (DC) power supply to the electrodes in the range of 0–50 V and 0–5 A. The two electrodes were rinsed with deionized water before and after each experimental run. The EC process was carried out under constant magnetic stirring to enhance solution homogenization and adsorption of fluoride ions. After the treatment, the solution was passed through a filter membrane with 0.45 µm pores to remove the formed flocks. The temperature, conductivity, and pH were continuously monitored. The mass weight of the aluminum electrode was recorded at the beginning and end of the experiment. Furthermore, the cell voltage was recorded at 10 minutes' intervals.

Analytical techniques

After each experiment, about 10 mL of the solution at systematic intervals was filtered from aluminum (III) hydroxide. The fluoride concentration was analyzed by the ion-selective electrode method according to the APHA alkalinity procedures [23]. To avoid interference from other ions, TISAB buffer (solution containing acetic acid, sodium hydroxide, sodium chloride, and 1, 2–cyclohexylene diamine tetra acetic acid) was poured to each aliquot within the volume of 1:1 ratio. Aluminum concentration was analyzed by using the 2320 alkalinity standard titration method for examination of water and wastewater according to [23]. Water physical parameters such as pH, temperature, conductivity, dissolved oxygen (DO), and total dissolved solids (TDS) were measured using the Hanna HI 9829 multi-parameter meter. The concentration of magnesium and calcium in the natural and synthetic water was analyzed by titration according to standard methods; potassium and sodium contents were analyzed by flame spectrometric method; chloride species via the Mohr titration technique; and sulfate ions by the gravimetric method. Each experiment was executed three times, and the results were averaged. The removal efficiency (%) of fluoride by the EC process was deliberated using Eq. (4) below:

$$E = \frac{(C_0 - C_f)}{C_0} \times 100\% \tag{4}$$

Where; C_0 is the initial fluoride concentration, and C_f is the final fluoride concentration at time t.

Results and discussion

Characterization of water sources

In this study, the physical and chemical compositions of selected sources of water for domestic use were analyzed. Additionally, selected rivers that are not directly used for domestic water supply but rather for livestock and irrigation purposes were sampled to compare the fluoride removal efficiencies since they have been previously reported to contain very high amounts of fluoride. Physical parameters such as pH, dissolved oxygen (DO), total dissolved solids (TDS), temperature (Temp), conductivity (Cond), resistivity (Rest) as indicated in Table 1 were measured; while the chemical compositions such as magnesium (Mg^{2+}), sodium (Na^+), chlorine (Cl^-), potassium (K), Sulphate ($SO_4{}^{2-}$) calcium (Ca^{2+}), and nitrate ($NO_3{}^-$) were measured.

These parameters are very important especially for fluoride removal in that water. Total dissolved solids (TDS) is the term used to describe the inorganic salts and small amounts of organic matter present in solution in water. The principal constituents are usually calcium, magnesium, sodium, and potassium cations and carbonate, hydrogencarbonate, chloride, sulfate, and nitrate anions. Even if the high amount is due to the presence of beneficial minerals, increased levels of TDS can give water a bitter, metallic, or salty taste, along with discoloring the water and creating an unpleasant odor. Total Dissolved Solids (TDS) correlates positively with conductivity and affects pH that means the higher the TDS, the higher the conductivity and the lower the pH, towards acidity. The presence of high TDS in water for instance, might affect its taste, hinder the removal of fluoride while the resultant changes in pH might affect the state of existence of coagulant $Al(OH)_3$.

Table 1
Physical characteristics of the selected water sources.

Parameters	Coordinates		pH	TDS (ppt)	Cond (µS/cm)	Temp (C)	DO (mg/L)	Rest (Ω)
	South	East						
Local tap water	3° 25′ 1″	36° 49′ 7″	6.82	1.63	846	26.6	2.46	232.2
Bulebule spring	3° 11′38″	36° 51′ 19″	7.2	738.8	731.3	19.4	4.33	678.8
Belem spring	3° 11′ 37″	36° 51′ 26″	7.09	1.082	1.077	21.8	2.27	462.5
River Ngarenanyuki	3° 52′ 39″	36° 51′ 26″	8.8	1.777	1.766	20.4	6.72	284.4
River Uluwile	3° 12′ 39″	36° 51′ 52″	6.67	2.151	2.133	20.7	3.46	232

Table 2
Composition of ions in water sources in mg/L.

	F^-	Cl^-	SO_4^{2-}	Ca^{2+}	Mg^{2+}	Na^+	K	NO_3^-
Local tap water	1.37	0.02	8	18.4	6.8	19.8	10.7	0.6
Bulebule spring	7.05	0.02	16	10.2	3.9	42.4	22.3	0.3
Belem spring	8.45	0.01	29	17.8	4.7	54.7	31	0.1
River Ngarenanyuki	29.5	0.03	57	6.09	1.89	68.7	45.6	0.1
River Uluwile	48	0.03	88	5.81	1.62	96.4	53.2	0.1

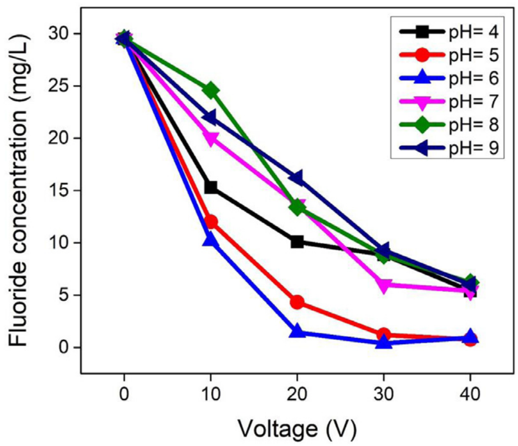

Fig. 1. Effect of pH on efficiency of fluoride removal for initial fluoride concentration of 29.5 mg/L over 30 mins.

[8,28]. The temperature (t) of a solution can control the EC process during treatment. An increase in temperature generates an increase in treatment efficiency due to the increased movement of ions. This movement promotes collision of ions with the coagulated material, thereby facilitating the removal process. [14]. A high dissolved oxygen (DO) level in a community water supply is good because it makes drinking water taste better. Adequate dissolved oxygen is necessary for good water quality. However, high DO levels speed up corrosion in water pipes. A minor increase in pH levels can cause an oligotrophic (rich in dissolved oxygen) lake to become eutrophic (lacking dissolved oxygen). Even minor pH changes can have long-term effects. Environmental impact of total dissolved gas concentration in water should not exceed 110% (above 13–14 mg/l). Concentration above this level can be harmful to aquatic life [28].

The chemical compositions shown in Table 2, do have a significant influence on the efficient removal of the fluoride. For instance, the various water sources show variation in the amount of fluoride and other ions. The local tap water, Bulebule, and Belem springs are the three (3) selected domestic water supply sources with less than 10 mg/L fluoride content. The Ngarenanyuki and Uluwile river bodies which have relatively high fluoride contents are used for irrigation and livestock purposes. This has a potential of indirectly affecting human beings through the food chain.

To understand the fluoride removal efficiency, the experimental setup was designed to investigate the influence of the initial fluoride concentration in the water source, the applied voltage, the electrolysis time, and the pH of the water.

Effect of pH

The pH is a very crucial parameter in the electrocoagulation process since it results in different hydroxides formation. Fig. 1 illustrates a representative trend of the effect of voltage variations on fluoride concentration at different pH values for river Ngarenanyuki with initial fluoride concentration of 29.5 mg/L within 30 mins. The fluoride removal efficiency by EC was shown to depend highly on the pH over the voltage ranges. Generally, at an optimal pH of 6, voltage of 30 V and

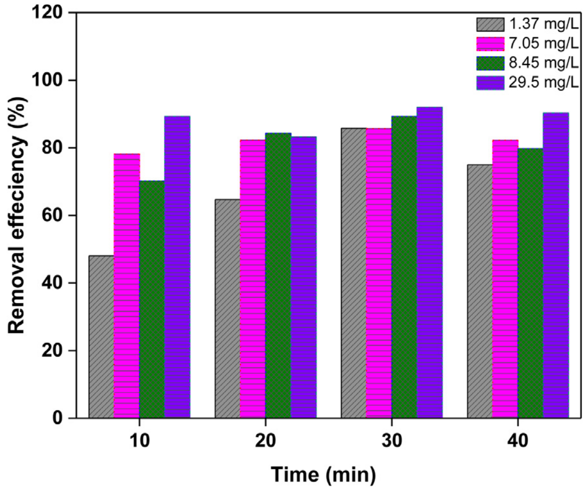

Fig. 2. Removal efficiency (%) as a function of time of experiment for various levels of initial concentration at a constant applied voltage of 30 V.

reaction time of 30 mins, the removal of fluoride was effective in all the water types considered. At a voltage of 40 V, the concentration however tends show to an increasing tendency. This could be attributed to the fact that the formed $Al(OH)_3$ gets polymerized to $Al_n(OH)_{3n}$ due to low solubility. This results in the formation of dense flocks with a large surface area that aid the fluoride removal [16]. Above pH 6 and below pH of 5 fluoride removal efficiency was found to decrease due to the evolution of $Al(OH)_4^-$ and AlO_2^- which are inefficient in fluoride removal process [2]. The study thus showed that the highest fluoride removal efficiency was at a pH of 6 attributed to the formation of stable aluminum hydroxides as earlier reported by [22]. According to a study conducted by [9], the uptake of fluoride in acidic medium was higher than in media with basic pH values; suggesting that apart from adsorption, sweep coagulation and co-precipitation are also responsible for the removal of fluoride.

The stability of aluminum hydroxide ($AlOH_3$) (coagulant) forming the complex was mostly favored by the pH of the solution. Nevertheless, the defluoridation process was found to be effective within a pH range of 6.

Effect of initial fluoride concentration

The initially dissolved fluoride concentration in water has been reported to greatly influence the defluoridation efficiency [3]. The water samples used in this study, had initial fluoride concentration values ranging 1.37 mg/L to 48 mg/L. From Fig. 2, which shows the fluoride removal efficiency over electrolysis time; it was observed that the water with the highest fluoride concentration required more $Al^{3+}$to be dissolved as compared to the others having lower fluoride concentrations. For instance, tap water with an initial fluoride concentration of 1.37 mg/L attained an 85% fluoride removal with 0.6 mg/L of Al^{3+}; whiles water from river Ngarenanyuki with an initial fluoride concentration of 29.5 mg/L obtained the same removal efficiency with 7.2 mg/L of Al^{3+}within the optimal electrolysis time of 30 mins. Similarly, Bulebule spring water with an initial fluoride concentration of 7.05 mg/L attained the same removal efficiency of 85% with Al^{3+} concentration of·4.3 mg/L. The highest fluoride concentrations showed an effective removal as compared with the lowest concentrations. This is because in more dilute solutions there is an evolution of the diffusion boundary that causes a slower rate of reaction in the vicinity of the electrode; hence, the diffusion layer has no effect on the rate of diffusion of metal ions to the electrode surface [3]. The electrocoagulation process was observed to decrease the fluoride concentrations to accepted standards (1.5 mg/L) in the drinking water.

Various studies have reported varied removal efficiencies at various applied voltages. Table SM1 compares the various studies that have been done using the similar EC process as used in this present study.

As can be seen from Table SM1, the study carried by [2,5,9] showed the highest removal at lower voltage compared to other studies and this is due to the wide range of electrolysis time (40–90 min). This had a very high affinity for the fluoride ions at pH values of 4–8.5 due to the evolution of huge amounts of aluminum hydroxide. According to [3], the fluoride removal factors such as pH (5–7), and initial fluoride concentration (10 mg/L), were similarly related to parameters applied in this study; however, voltages beyond 40 V resulted in high amounts of aluminum residues in treated water.

Effect of electrolysis time

Electrolysis time is an important parameter in EC since it determines how long water should be in a treatment system to reach the allowed standards. Fig. 3 indicates that the concentration of ions and their hydroxide flocs increases as the electrolysis period increases; leading to higher removal efficiencies. This is because the amount of Al^{3+}ions produced from the aluminum electrodes depend on the reaction time to scavenge the anode and resulting in the amorphous aluminum

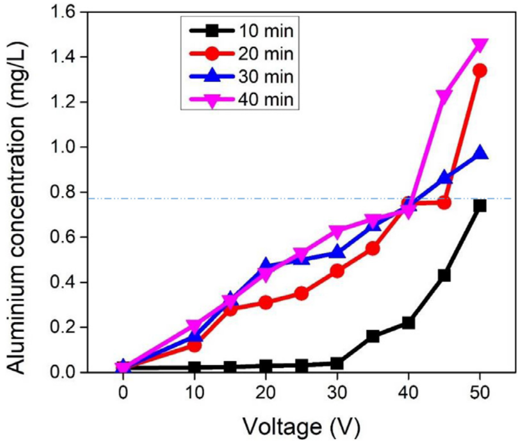

Fig. 3. Effect of applied voltage on aluminum residue concentration as a function of electrolysis time.

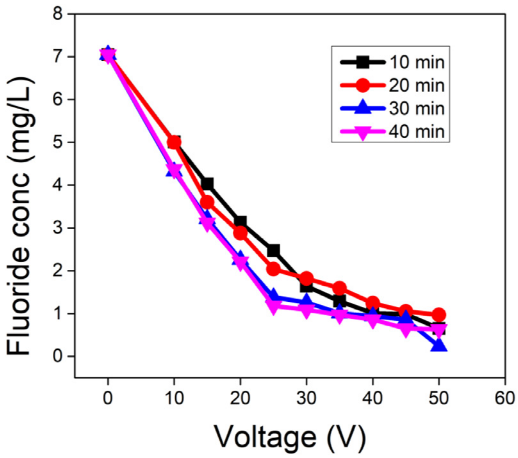

Fig. 4. Effect of applied voltage on fluoride concentration at various electrolysis times at pH of 6.

hydroxide precipitation [5]. However, as the reaction time increases, the aluminum residue content in the water also increases. From the experiments conducted in this study, quality water with fluoride concentrations below the WHO and TBS recommendations were achieved within an optimum electrolysis time of 30 min. It is worth noting however that, as the electrolysis time increased above 30 min, the aluminum residues also increased above the WHO limits of 0.7 mg/L; which can be toxic to the drinking water [19,20,25].

Effect of an applied voltage

To examine the effect of applied potential on the efficient removal of fluoride from the water, the EC experiments were executed using different voltages (0 – 50 V). From Fig. 4, it is observed that as the applied voltage increases, less time is required for the efficient fluoride removal. As previously discussed, in the electrocoagulation process, the applied cell potential contributes to the amount of coagulants ($AlOH_3$) and hence affects the growth of flocks with bubble size [5,10].

Increasing the voltage, resulted in high fluoride removal efficiency. Similarly, more electrolysis time and higher voltage resulted in more yield of the coagulant ($AlOH_3$) which subsequently increase the rate of defluoridation. The major hindrance to this phenomenon is the high Al^{3+} residues in the water which raises more toxicity concerns in the resulting water.

Conclusion

The Electrocoagulation method showed positive results in purifying water with high concentrations of fluoride. The results of the study indicate that the EC efficiency depends on the amount of the coagulant ($AlOH_3$) produced in the water, which depends on electrolysis time and applied voltage. Other parameters such as pH and initial concentration of solution also influenced the results and their effect was well determined in this study. The fluoride concentration 1.5 mg/L set by both WHO and TBS guidelines for drinking water was reached at a high removal efficiency of 90%. From the experiments, it was shown that only 30 min are required to treat water to reach the standard limits for fluoride in Tanzania. The applied voltage range of 30 – 35 V has been proposed herein as the optimum potential for a better electrocoagulation process in the treatment of fluoride contaminated water.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.sciaf.2021.e00737.

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