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Removal of fluoride and pathogens from water using the combined electrocoagulation-inline-electrolytic disinfection process

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ABSTRACT

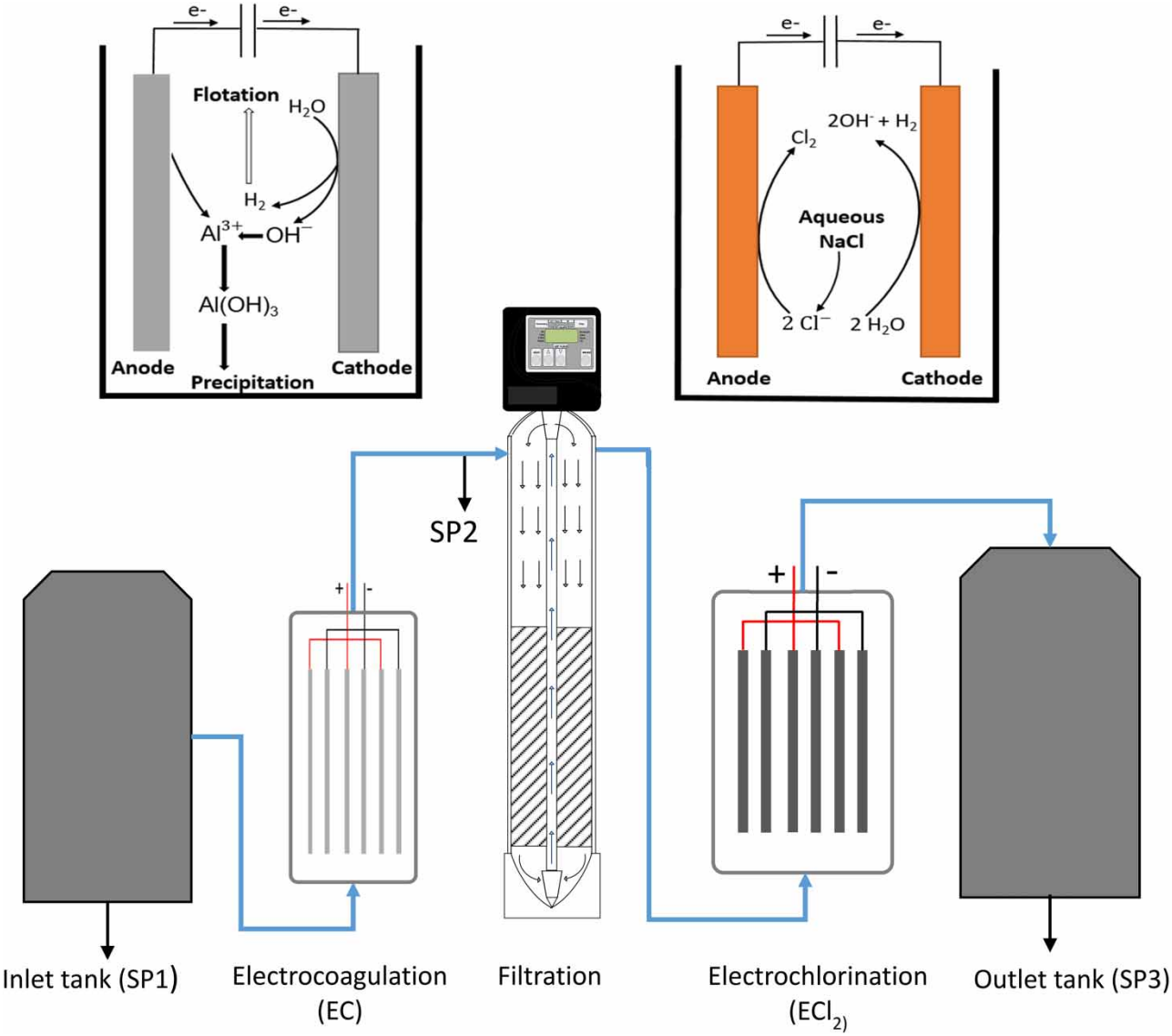
The consecutive removal of fluoride (defluoridation) and pathogens (disinfection) in drinking water through combined electrocoagulation-inline-electrolytic disinfection (EC–ECl_2) process with aluminum and dimension-stable mixed oxide electrodes was reported in this study. Laboratory trials were conducted on the effects of flow rate, initial pH, current density, and supporting electrolytes for defluoridation and disinfection processes. The results have shown that with a flow rate of 10 L/h, initial pH of 6, the current density of 9.4 mA/cm^2 (EC cell) and 3.1 mA/cm^2 (ECl_2 cell), supporting electrolyte concentration of 165 mg/L, and electrolysis time of 50 min, a defluoridation rate of 88% (initial concentration of 12.3 mg/L) and complete disinfection (initial fecal coliforms of 19,700 colony-forming units per 100 mL (CFU/100 mL)) can be reached. The final concentration of fluoride and pathogens in treated water was 1.44 mg/L and 0 CFU/100 mL, which are within the acceptable limit of the World Health Organization and the Tanzania Bureau of Standards of 1.5 mg/L and 0 CFU/100 mL, respectively. The EC–ECl_2 system is a promising approach for consecutive defluoridation and disinfection of water to save millions from fluorosis and waterborne diseases. However, optimization potential with regard to energetic efficiency and system complexity was identified.

Key words: defluoridation, disinfection, drinking water, electrocoagulation, inline-electrolysis, supporting electrolyte

HIGHLIGHTS

- Consecutive defluoridation and disinfection of water by combined electrocoagulation-inline-electrolytic disinfection (EC-ECl2).
- Aluminum and titanium mixed oxide as sacrificial anodes.
- Electrolyzed at various current density, flowrates, and supporting electrolyte.
- 88% defluoridation (12.3 mg/L) and complete disinfection (19700 CFU/100 mL) meet the WHO standards (<1.5 mg/L and 0 CFU/100 mL, respectively).
- Chloride ion break passivation in EC cell and enhance chlorine production in the EC-ECl2 system.

GRAPHICAL ABSTRACT



INTRODUCTION

In many developing countries around the world, access to safe drinking water is still a major concern (Baker 2018). Water pollution originates from anthropogenic sources such as the dumping of untreated waste in the environment and geogenic sources such as fluoride and arsenic contamination of groundwater sources (Kihupi *et al.* 2016; Dhadge *et al.* 2018). The presence of fluoride in drinking water is a serious problem in some parts of the world. High fluoride in drinking water is known to cause dental fluorosis and skeletal defects when consumed (Mureth *et al.* 2021). The possible sources of high fluoride in Lake Momella are connected to volcanic activities in rift valley zones of Mt Meru (Malago *et al.* 2017; Kitalika *et al.* 2018). Thus, for human health, defluoridation is very important in drinking water.

Several defluoridation methods are reported, including adsorption, coagulation–precipitation, ion exchange, reverse osmosis, membrane separation, and electrocoagulation (EC) to maintain permissible fluoride levels (Dubey *et al.* 2018). Adsorption is performed with great efficiency and low cost using locally accessible adsorbent materials. Its efficiency decreases with an initial concentration greater than 5 mg/L because of faster saturation of adsorbent's active sites (Akafu *et al.* 2019). Coagulation–precipitation is a well-known approach, but it requires a large chemical dosage resulting in a

large amount of sludge production and negative health consequences (Akafu *et al.* 2019; Alkurdi *et al.* 2019). Ion exchange resins have a high ability to remove fluoride, but their application is limited by high resin costs, high chloride levels, low pH in treated water, and reduced effectiveness due to ionic competition with phosphate, carbonate, and sulfate (Mobeen & Kumar 2017; Chatterjee *et al.* 2020). Membrane techniques, including microfiltration, nanofiltration, and reverse osmosis, are efficient and chemical-free. The drawbacks include the high cost of specialized labor, not ideal for high-salinity water, and the efficiency is limited by clogging and requires continual maintenance and monitoring (Thakur & Mondal 2017; Baker 2018; Kumar *et al.* 2019; Giwa *et al.* 2021). EC is a fast way of removing pollutants such as fluoride from drinking water. The advantages of EC include less sludge generation, simplicity of operation, and no chemical additives (Ndjomgoue-Yossa *et al.* 2015; Guzmán *et al.* 2016). The EC method is reported to have high deflouridation efficiency when iron (Fe) or aluminum (Al) electrodes are used (Aoudj *et al.* 2017).

On the other hand, physical and chemical disinfection processes are utilized to make water safe for drinking purposes (Baker 2018). Microorganisms are destroyed or removed in physical disinfection approaches by using ultraviolet (UV) or ionizing radiation, heat, ultrasound, and membrane filtration (MF). The lack of a reservoir effect is the major drawback of physical disinfection techniques (Baker 2018; Ghernaout 2019). Chemical processes include adding chemicals to the water to be treated, such as chlorine (Cl_2), chlorine dioxide (ClO_2), or sodium hypochlorite (NaOCl, i.e., bleach) (Baker 2018). These processes destroy pathogens, and chlorine-based reagents create a residual effect that protects the water for a period of time against recontamination. Unwanted disinfection side reactions with chemicals in the water are a common drawback of chemical processes (WHO 2017; Baker 2018).

Electrochemical water disinfection uses appropriate electrodes, such as dimension-stable titanium electrodes coated with oxides of ruthenium and iridium (mixed oxide electrodes – MOX), to produce chlorine gas (Cl_2), which hydrolyzes to hypochlorous acid inactivating a broad range of microbes (Otter *et al.* 2017; Ghernaout 2019). This method has several advantages over conventional chemical disinfection methods, including the elimination of disinfectant supply, storage, or dosage. It has a residual effect, is typically more cost-effective, and requires less maintenance compared to other disinfection methods (Ndjomgoue-Yossa *et al.* 2015).

In this study, EC was combined with inline-electrolysis (ECl_2). This study optimized the conditions of EC for fluoride removal and chlorine production from sodium chloride (NaCl) to disinfect waters with low geogenic chloride concentration.

Principle of the EC–ECl_2 process

The EC–ECl_2 process involves the following overall equations:

$$Al + (3 - x)OH^- + xF^- \rightarrow Al(OH)_{3-x}F_x \quad (1)$$

$$Cl^- + H_2O \rightarrow HClO + H^+ \leftrightarrow ClO^- + 2H^+ \quad (2)$$

In the EC cell (Equation (1)), Al^{3+} formed on the Al anode reacts with F^- in the water to form fluoride aluminum complexes $Al(OH)_{3-x}F_x$, thus precipitating F^- out of the water. In the ECl_2 cell, chloride (Cl^-) ions dissolved in the water are electrochemically oxidized to form chlorine gas that hydrolyzes to hypochlorous acid (HClO).

MATERIALS AND METHODS

Solution chemistry

All of the compounds utilized in this study were analytical grade. Laboratory experiments were conducted at a room temperature of 25 ± 3 °C with 1.91 and 12.3 mg/L as initial fluoride concentrations of raw water from the NM-AIST laboratory and synthetic water, respectively. In a continuous flow reactor, the effect of operational conditions (flow rate, initial fluoride concentration, initial pH, current density, and supporting electrolyte) on the defluoridation process was examined using synthetic water (deionized water+ sodium fluoride (NaF) salt + NaCl salt). Sodium fluoride (12.3 mgF^-/L) and sodium chloride (up to 165 mg/L) were added to the aqueous solution to promote the required tested fluoride concentrations and conductivity (for breaking and preventing anodic passivation), respectively, in the EC–ECl_2 cells; 6 N sodium hydroxide and 2 N hydrochloric acid solutions were added for pH adjustment (values 5.0–7.5). Fifty-two grams of the M-FC Agar Base powder were suspended in 1 L of purified water and mixed thoroughly for media preparation. With frequent agitation, the mixture was heated and boiled for 1 min to completely dissolve the powder. Ten milliliters of a 1% solution of rosolic acid were added

in 0.2 N NaOH and continuously heated for 1 min but not autoclaved. Stable, typical control cultures were used to test the performance of finished product samples.

Experimental setup

The SuMeWa|SYSTEM (from Sun Meets Water) is a solar-powered water pumping and filtration unit that has been developed to consecutively defluoridate and disinfect drinking water. As seen in Figure 1, the experimental setup was designed and constructed with inlet and outlet/reservoir tanks of 500 L each, electrocoagulation cell (EC), filter, and ECl_2 cell. The filter shown in Figure 1 contains AFM® 0 with particle size of 0.025–0.05 mm to retain mainly aluminum fluoride particles. Based on the differential pressure within the filter bed, the filter was automatically backwashed and rinsed once a differential pressure of 0.4 bar was reached. No media exchange was required.

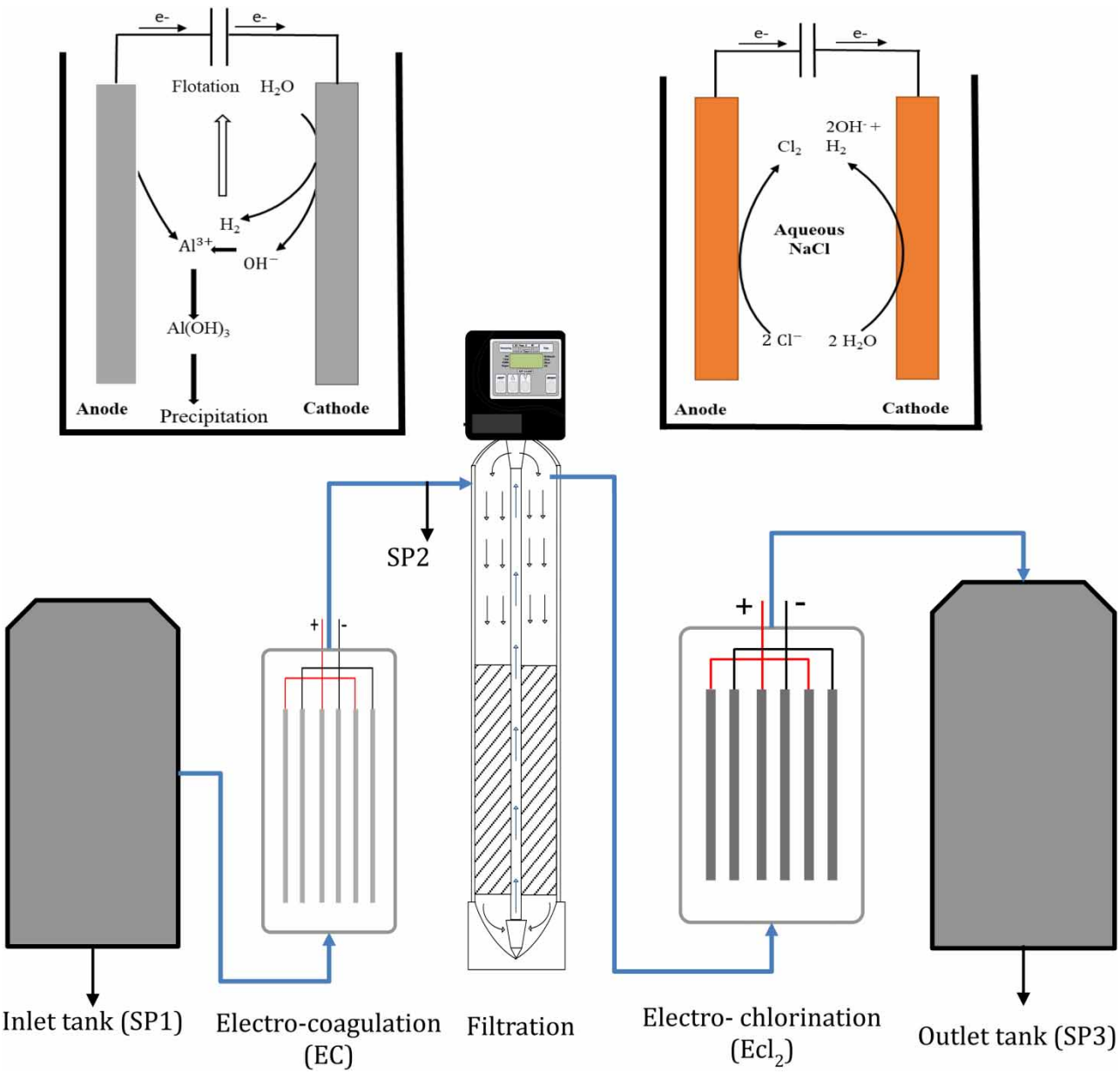


Figure 1 | Schematic diagram of water treatment (SuMeWa) system.

Water flows from the inlet tank through the electrocoagulation cell (size, 1 L) made up of six aluminum plates each (purity of Al, 99.7%). Figure 2(a) shows the configuration of the aluminum electrode. The area of each plate was 85 cm², with a gap of 5 mm. The total surface area of the stack was 510 cm^2, and the effective surface area of the stack was 425 cm^2. As seen in Figure 2(b), an electrical connection was made in a parallel monopolar connection. Water was then allowed to flow through the filter for flocs removal and finally to the ECl_2 cell (size, 2 L). Through the applied current, aluminum (Al^{3+}) ions were produced in the electrocoagulation cell and the coagulant $Al(OH)_3$ was generated to allow defluoridation.

Six electrodes were used to pass direct current from a DC power supply (0–30 V, 1.0–7.8 A) to the water resulting in current densities between 2.4 and 18.4 mA/cm². The cell's current was controlled using a digital power display. For each run, water samples were collected from the drain tube parts of the system for the measurements of pH, aluminum, fluoride, chlorine, and fecal coliform. The ECl_2 cell was composed of four pairs of dimensional stable titanium electrodes coated with oxides of ruthenium and iridium (MOX-electrodes) (Otter *et al.* 2017; Kunz *et al.* 2018). Figure 2(c) shows that the surface area of each anode/cathode plate was 640 cm². In the ECl_2 cell, chlorine gas was produced inline-electrolytically from dissolved chloride ions for disinfection.

Analytical techniques

The defluoridation in the laboratory was performed in a continuous mode using fluoridated water made from a synthesized solution of NaF with tap water. The physical parameters of water, such as temperature, electrical conductivity, pH, total dissolved solids (TDS), and dissolved oxygen (DO) of water samples, were measured by using the Hanna HI 9829 multiparameter. A particular fluoride electrode (PF4L from Tacussel (Lyon)) or ion selective electrode (ISE) was used to determine the fluoride concentration. In order to prevent the influence of interfering ions, TISAB or total ionic strength adjustment buffer (58 g of NaCl, 57 mL of glacial acetic acid, 4 g of 1,2 cyclohexylenediaminetetraacetic (CDTA), and

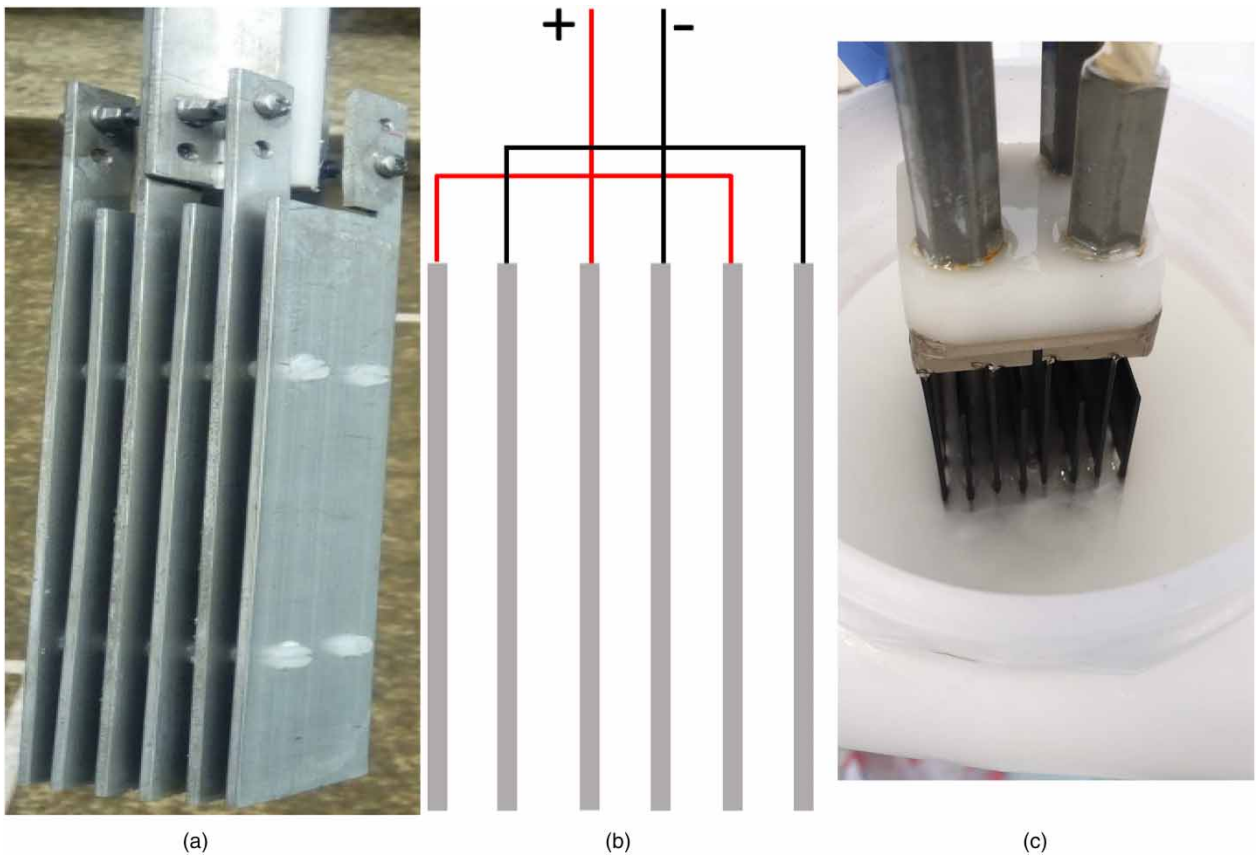


Figure 2 | (a) Al electrode configuration. (b) Power supply in a parallel monopolar connection. (c) Titanium electrode configuration.

125 mL of 6 N NaOH were dissolved in 1 L of distilled water by stirring until pH 5.3–5.5 was reached) was added to the samples in equivalent amounts.

Cell voltage and current were monitored using a digital power display. Chloride concentrations were determined by using the titration method. The free chlorine and total chlorine in water samples were analyzed using the DPD (*N*,*N*-diethyl-*p*-phenylenediamine) colorimetric method. The aluminum concentration was analyzed by using a spectrophotometer (Hach DR900) via Method 8012 in the presence of AluVer 3 Aluminum Reagent Powder Pillow.

The influent water was then prepared by mixing 400 L of synthetic water with 100 L of domestic wastewater (as a source of numerous fecal coliforms). For a lower number of fecal coliforms, 0.5 L of domestic wastewater was mixed with 500 L of synthetic water, and the water samples were analyzed using a MF technique with 0.45 μm pore size microbial filter paper. The counting of fecal coliforms before and after electrochemical treatment was conducted to determine the efficiency of the process. Following the MF procedure, an enriched lactose (M-FC Agar Base) medium and an incubation temperature of 44.5 ± 0.2 °C for 24 h were applied. Fecal coliform concentration was reported by means of a colony counter in terms of the number of bacterial colonies per 100 mL of sample water or colony-forming units per 100 mL (CFU/100 mL). The ranges of the operational conditions (such as flow rate, fluoride concentrations, pH, current, and supporting electrolyte) that have been evaluated during this study are presented in Table 1.

Data analysis and interpretation

The efficiency of defluoridation was determined via Equation (3):

$$E = \frac{C_0 - C_f}{C_0} \times 100\% \tag{3}$$

where E is the fluoride removal efficiency, C_0 is the initial fluoride concentration in the water sample (mg/L), and C_f is the residual fluoride concentration at equilibrium time (mg/L).

RESULTS AND DISCUSSION

Water quality of synthetic and treated water

The characteristics of synthetic and treated water used in this study before and after treatment, respectively, were analyzed as shown in Table 2. Data values for treated water were presented, the final conditions of the water after both EC and ECl_2 processes.

Table 1 | Ranges of the experimental parameters

Common EC–ECl_2 parameters	
Electrolytic time (min)	10–90
Flow rate of water (L/h)	10–40
Supporting electrolyte, NaCl (mg/L)	99–165
Electrocoagulation parameters	
Fluoride concentration (mg/L)	1.9–12.3
pH	5.0–7.5
Current density (mA/cm^2)	9.4–18.4
Anode area, electrocoagulation cell (cm^2)	425
Volume of electrocoagulation cell (L)	1
Electrodisinfection parameters	
Current density (mA/cm^2)	1.6–9.4
Anode area, electrodisinfection cell (cm^2)	640
Volume of electrodisinfection cell (L)	2

Table 2 | Characteristics of synthetic and treated water

Parameter	Synthetic water	Treated water
pH	6.8 ± 0.08	7.26 ± 0.16
Temperature (°C)	25 ± 3	25 ± 3
Conductivity (μS/cm)	954 ± 3.43	814.38 ± 3.71
Total dissolved solids (mg/L)	441 ± 0.78	409 ± 1.09
Dissolved oxygen (mg/L)	6.34 ± 0.37	5.95 ± 0.14
Fluoride (mg/L)	12.3 ± 0.08	0.16–1.44
Chloride (mg/L)	240.11 ± 0.42	165.96 ± 1.02
Aluminum (mg/L)	23.32 ± 0.83	0.055 ± 0.0056
Free Cl_2 (mg/L)	0.00	0.3–1.5
Total Cl_2 (mg/L)	0.00	0.7–2.2
Fecal coliform (CFU/100 mL)	36–19,700	0

Effect of current on coagulant formation

The most important operating parameter in the electrocoagulation process was current (Emamjomeh & Sivakumar 2009; Takdastan *et al.* 2014). It influences the liberation of free aluminum concentration (Al^{3+} ions) from the anode needed to combine with hydroxide (OH^-) ions from the cathode to produce the required coagulant, $Al(OH)_3$, responsible for defluoridation. In this study, current density values applied were 9.4, 11.8, 14.1, 16.5, and 18.4 mA/cm^2 to achieve currents of 4, 5, 6, 7, and 7.8 A to investigate its influence on fluoride removal at constant initial fluoride concentration (12.3 mg/L), varied flow rates (10–40 L/h), and electrolysis time (10–90 min). According to Faraday's first law of electrolysis, the amount of aluminum ions produced from aluminum electrodes during electrolysis is proportional to the quantity of electricity (product of current and electrolysis time) passed through the solution. Figure 3 shows that the released concentrations of aluminum ions into the solution were increased with current density and eventually reduced the fluoride concentrations. Figure 4 shows that at a minimum current density of 9.4 mA/cm^2, initial pH of 6.8, flow rate of 10 L/h, and an electrolysis time of 50 min, a residual fluoride concentration of 1.5 mg/L was achieved. Aluminum concentration reached 13.97 ± 2.98 mg/L (measured after the EC cell). At a flow rate of 20 L/h, electrolytic time of 50 min and a pH range of 5.0–7.5, the concentration of aluminum ions increased with applied current density (18.4 mA/cm^2) up to 23.32 ± 0.83 mg/L. As a result, at 10 L/h, the optimum current

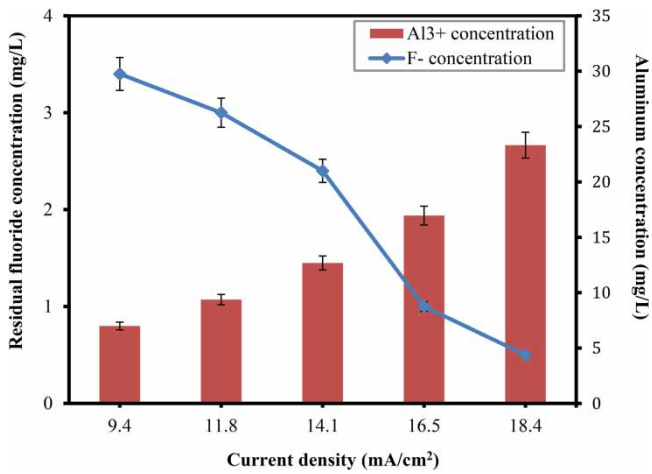


Figure 3 | Effect of current density (9.4–18.4 mA/cm^2) on the production of aluminum (Al^{3+}) at a constant flow rate (20 L/h) and initial fluoride concentration (12.3 mg/L).

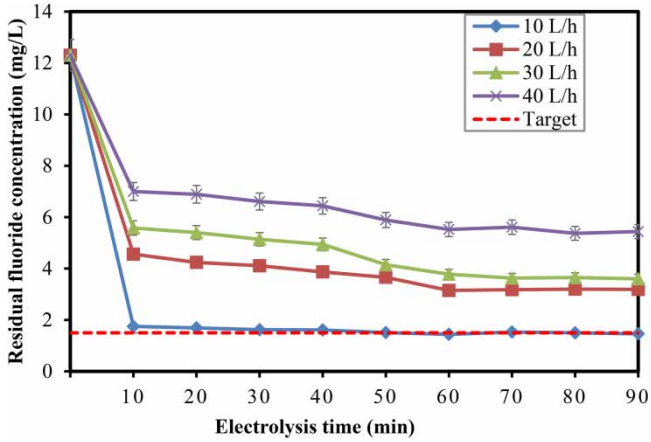


Figure 4 | Effect of flow rates (10–40 L/h) on residual fluoride concentrations at constant current density (9.4 mA/cm^2), pH (6.8), initial fluoride concentration (12.3 mg/L), and varied electrolysis time (10–90 min).

for defluoridation was 9.4 mA/cm^2, whereas, at 20 L/h, it ranged between 16.5 and 18.4 mA/cm^2. The findings of this study verified that the increase in current density improved the defluoridation. This agrees with the findings reported by Ghosh *et al.* (2008).

Residual aluminum concentration was the amount of uncombined Al^{3+} ions that remained in the treated water after the filtration step. The measured residual Al concentrations after the filtration step ranged from 0.07 to 0.03 mg/L at flow rates ranging from 10 to 40 L/h, respectively, which were less than the acceptable limit of the World Health Organization (WHO) and the Tanzania Bureau of Standards (TBS) (0.2 mg/L).

Effect of flow rate on defluoridation

The influence of flow rates on defluoridation was investigated in this study by altering the flow rates from 10 to 40 L/h at various initial fluoride concentrations and a fixed current density of 9.4 mA/cm^2. For a fixed current density of 9.4 mA/cm^2 and a pH of 6.8, with initial fluoride concentrations of 1.91 mg/L (raw water) and 12.3 mg/L (synthetic water), the residual fluoride concentrations were increased from 0.19 to 0.46 mg/L and from 1.44 to 5.37 mg/L as the flow rate increased from 10 to 40 L/h, respectively. The flow rates have been found to influence the defluoridation by defining the residence time. The flow rates of 10, 20, 30, and 40 L/h in an electrocoagulation cell with 1 L volume (size) have the residence time of 6, 3, 2, and 1.5 min, respectively. Figure 4 shows that the higher the flow rate, the lower the time the water spends within the reactor, resulting in a reasonable reduction in defluoridation efficiency. Similar results for low retention time at a high flow rate were confirmed by Emamjomeh & Sivakumar (2009). Fluoride concentrations decreased with the increase in electrolysis time as more coagulants were formed.

Effect of initial fluoride concentration

Initial fluoride concentration also influences its removal efficiency (Takdastan *et al.* 2014). In this study, raw and synthetic water with initial fluoride concentrations of 1.9 and 12.3 mg/L, respectively, were tested to evaluate the fluoride removal efficiency of the EC process at various flow rates (10–40 L/h), a pH of 6.8, and a current density of 9.4 mA/cm^2. During electrocoagulation experiments, an increase in initial fluoride concentration was found to increase residual fluoride concentrations [Appendix (2c)]. This can be explained by an increase in the ratio of fluoride ions to available coagulant complexation sites at a constant current density of 9.4 mA/cm^2. Figure 5 shows reduced removal efficiency as the fluoride concentration was increased from 1.9 to 12.3 mg/L. Despite that the total amount of fluoride removed was substantially larger at higher fluoride concentrations, the system can be applied and the maximum efficiency can be achieved.

Effect of initial pH

The initial pH of the feed water has been found to influence the EC–ECl_2 process removal efficiency for fluoride. H^+ ions react with F^- ions to produce hydrogen fluoride (HF) at acidic pH (4.5), but they can also react with OH^- and HCO_3^- ions at alkaline pH (Apshankar & Goel 2018). For effective disinfection with chlorine, the pH should preferably be less than 8 (Kihupi *et al.*

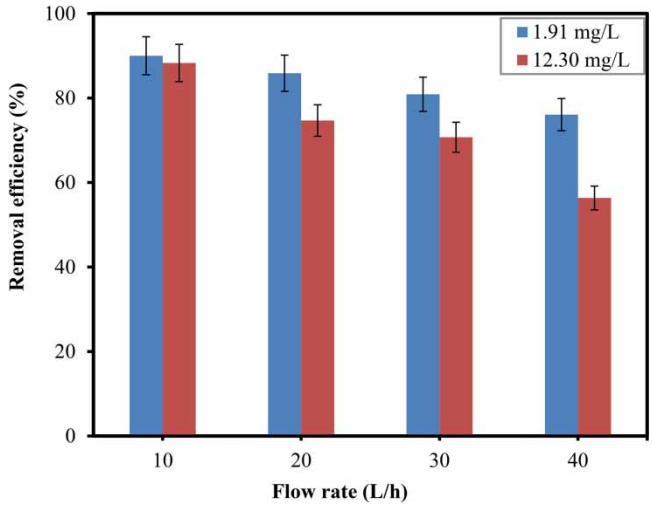


Figure 5 | Effect of initial fluoride concentrations (mg/L) on defluoridation efficiency after EC cell at constant pH (6.8), current density (9.4 mA/cm^2), and varied flow rates (10–40 L/h).

2016). The pH of the solution favored the stability of $Al(OH)_3$, resulting in the formation of the complex. In this study, the effect of the initial pH of the feed water range of 5.0–7.5 was used to evaluate the defluoridation capacity of the EC process at a constant flow rate (20 L/h), initial fluoride concentration (12.3 mg/L), and varied current density range from 9.4 to 18.4 mA/cm^2 at the EC cell. The lowest residual fluoride of 0.16 mg/L (98.7%) has been found at a pH of 6 and the current density of 18.4 mA/cm^2. This was attributed to the formation of sufficient $Al(OH)_3$. This study agreed with the batch findings reported previously by Mureth *et al.* (2021) that the optimum pH for defluoridation was 6. Figure 6 shows that, at both tested initial pH (5.0–7.5), the target residual fluoride (1.5 mg/L) was achieved at the current density range of 16.5–18.4 mA/cm^2. This verified that the EC–ECl_2 system was capable of removing high fluoride concentrations from drinking water at a wide range of the initial pH of the feed water. As seen in Figure 4, a residual fluoride concentration of 1.44 mg/L (88%) was achieved at a constant initial pH of 6.8 of raw water, the current density of 9.4 mA/cm^2, the flow rate of 10 L/h, and initial fluoride concentration of 12.3 mg/L.

Effect of current density on disinfection

During an experiment using the ECl_2 cell (current range of 1.6–9.4 mA/cm^2, pH of 6, flow rate of 20 L/h), water containing 19,700 CFU/100 mL was disinfected. Figure 7 shows the concentration of free and total chlorine with increasing current

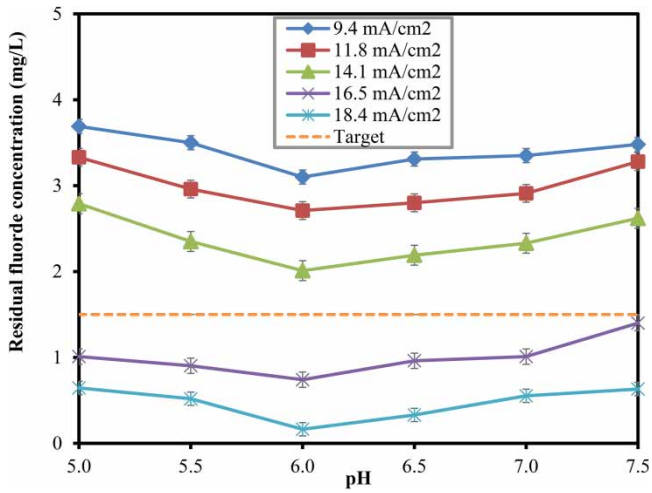


Figure 6 | Effect of initial pH (5.0–7.5) on residual fluoride concentration at constant flow rates (20 L/h), initial fluoride concentration (12.3 mg/L), and varied current density (9.4–18.4 mA/cm^2).

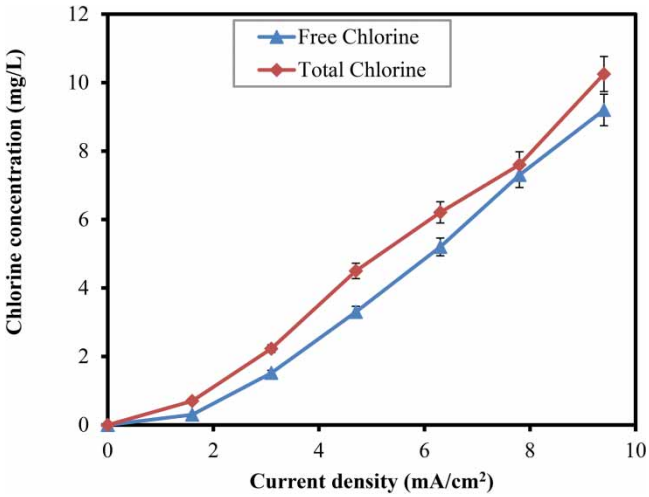


Figure 7 | Effect of current density (1.6–9.4 mA/cm^2) on disinfectant production at constant pH (6.0) and flow rate (20 L/h).

density. Moreover, even at the low range of current density (1.6–3.1 mA/cm^2), a positive impact on water disinfection efficiency was observed. With a current density of 1.6 mA/cm^2, 0.3 mg/L of free chlorine could be produced. This was sufficient to disinfect raw and synthetic water with a lower number of fecal coliforms about 8 and 36 CFU/100 mL. At a higher number of fecal coliforms, 19,700 CFU/100 mL, 1.6 mA/cm^2 was found to disinfect water to 134 CFU/100 mL (2-log reduction); 1.5 mg/L of free chlorine concentration produced at 2 A (3.1 mA/cm^2) was sufficient to disinfect the water with 19,700 CFU/100 mL to the permissible limit set by the WHO and the TBS for treated drinking water of 0 CFU/100 mL (4-log reduction).

Effect of the supporting electrolyte

NaCl was chosen as a supporting electrolyte because of its effect in causing a significant increase of electrical conductivity, formation of chlorine gas, cost-effective, and low toxicity (Ndjomgoue-Yossa *et al.* 2015; Thakur & Mondal 2017). NaCl can significantly reduce the effects of anionic species such as sulfate (SO_4^{2-}) and bicarbonate (HCO_3^-). In fact, SO_4^{2-} ions were recognized as one of the agents of passivation, whereas Cl^- causes the passive layer to decompose (Dura 2013).

Breaking of the passive layer

The effect of the NaCl-supporting electrolyte on electrode passivation, defluoridation, and disinfection has been investigated in this study. During the defluoridation experiment, the current density at the EC cell in water without additional supporting electrolytes was found to decrease as the treatment continued and this was due to the formation of the passive layer on aluminum electrodes (Appendix 11). Mohora *et al.* (2012) suggested 60 mg/L of Cl^- as a minimum electrolyte concentration for breaking and preventing anodic passivation on the Al electrode. In this study, 99 and 165 mg/L of NaCl were added into the water to be treated to promote 60 and 100 mg/L, respectively, of Cl^- ions that are needed to break the passive layer of aluminum oxide (Al_2O_3) to yield aluminum chloride ($AlCl_3$) as a soluble salt (Equation (4)):

$$2Al_2O_3 + 12Cl^- \rightarrow 4AlCl_3 + 3O_2 \quad (4)$$

The chlorides at 100 mg/L were effective in breaking down and preventing the formation of the passive layer on the aluminum electrode and improved the EC–ECl_2 defluoridation efficiency. Figure 8 shows the slight variation in the current density passing through the EC cell, which implies that the anodic passive layer has completely broken down.

Improving the electrical conductivity

Increasing solution conductivity in water by adding salts or salt solutions improves the removal efficiency of both fluoride and pathogens. The lower initial water conductivity was found to have a negative effect on defluoridation efficiency in this study.

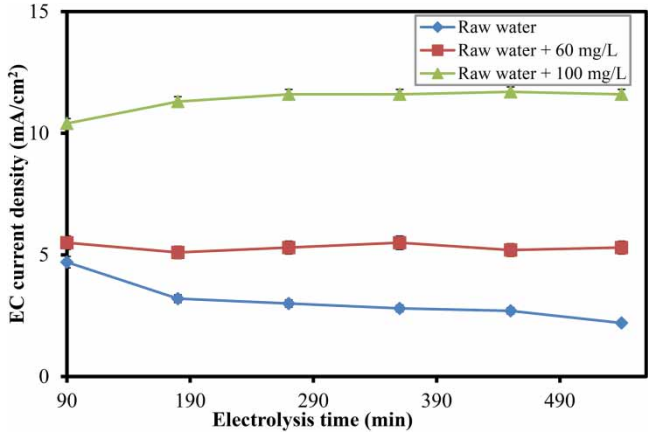


Figure 8 | Effect of supporting electrolyte, Cl^- (60–100 mg/L), on current density passed through EC cell at constant pH (6.8).

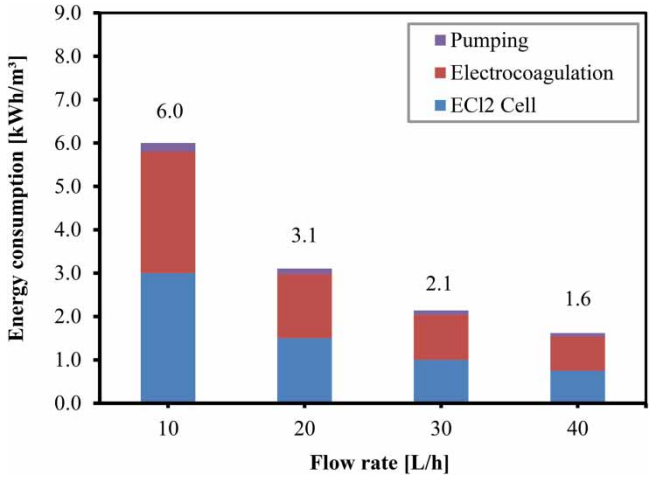


Figure 9 | Total energy consumption of the treatment process under laboratory-scale conditions.

The addition of the supporting electrolyte (99–165 mg/L) into the water to be treated increased the electrical conductivity from 331 to 954 μS/cm, reduced the applied voltage, and increased the current flowing through it.

Energy consumption

Figure 9 shows the energy consumption of the process under laboratory-scale conditions.

Only at a flow rate of 10 L/h, the treatment targets could be met. However, with energy consumption of 6 kWh/m³, the process is at this point when compared to alternative treatment technologies such as nanofiltration (~1.5 kWh/m^3, Schäfer *et al.* 2018) or reverse osmosis (3–4 kWh/m^3, Schmidt *et al.* 2016) energetically not feasible for in-field application. Optimization potential exists with regard to the alignment between the initial fluoride concentration and current applied to the EC cell. Emamjomeh & Sivakumar (2009) suggested to limiting the current density in the electrocoagulation treatment system purposely to avoid excessive energy consumption when the target fluoride concentration range was successfully reached.

CONCLUSIONS

Fluoride and pathogens in contaminated water can be consecutively removed by using the two separate processes (electrocoagulation and electrodisinfection) combined in one unit. The optimum conditions for fluoride and pathogen removal under the given test setting are found to be pH: 6, current density: 9.4 mA/cm^2 (defluoridation) and 3.1 mA/cm^2 (disinfection),

electrolysis time: 50 min, supporting electrolyte: 165 mg/L, free chlorine: 0.3–1.5 mg/L at initial fluoride concentration and pathogens of about 12.3 mg/L and 19,700 CFU/100 mL. Under the optimized conditions, the concentrations of fluoride and pathogens in treated water were found to be 1.44 mg/L (% removal: 88%) and 0 CFU/100 mL (>4 log units), respectively, below the limit set by the WHO and the TBS of 1.5 mg/L and 0 CFU/100 mL. The EC–ECl_2 system is a promising solution for the consecutive removal of fluoride and pathogens from water to save the lives of millions from fluorosis and waterborne diseases, especially in developing countries where the two challenges coexist in water sources as well as electrical power is a problem. However, before field application is technically feasible, the energy consumption must be substantially reduced in order to compete with, e.g., desalination technologies. The current study's findings are of novel significance in terms of the subsequent removal of fluoride and pathogens from drinking water, which has not been reported in previous treatment studies involving electrocoagulation processes. On a laboratory scale, the addition of electrolytes to the water was feasible. In a field application, this addition may not be feasible and the application of the presented the treatment approach limited to water with the given conductivity ranges.

RECOMMENDATIONS

The water supply is purposeful for drinking, and disinfection is of great importance. During this study, chlorination as a disinfection method was applied. The levels of disinfectant by-products (DBPs) such as trihalomethanes generated during the experiment were not determined. Because of serious health concerns, checking the levels of DBPs by considering the type of dissolved organic matter in water is recommended for future studies as well as the influence of the seasonal variations on the chlorination process since chloride in the water changes with seasons. Moreover, there is a need to include a reservoir that would allow enough time for stabilization before the filtration step. This reservoir is recommended to have a residence time of 150 min as reported in the results. Testing the system for defluoridation at higher fluoride concentrations and estimation cost per unit volume of water produced by the system are also recommended. Changing the production of chlorine from inline-electrolysis to onsite chlorine generation, where chlorine is produced from an NaCl brine, reduces the energy consumption of chlorine generation substantially.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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