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# Fluoride and pathogens removal from water using combined electrocoagulation-inline-electrolytic disinfection process

Njau, Oscar

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# FLUORIDE AND PATHOGENS REMOVAL FROM WATER USING COMBINED ELECTROCOAGULATION-INLINE-ELECTROLYTIC DISINFECTION PROCESS

**Oscar Ezekiel Njau**

**A Dissertation Submitted in Partial Fulfilment of the Requirements for the Degree of Master's in Environmental Science and Engineering of the Nelson Mandela African Institution of Science and Technology**

**Arusha, Tanzania**

**August, 2024**

## 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 have been 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/cm2 (EC cell) and 3.1 mA/cm2 ($ECl_2$ 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 energy efficiency and system complexity were identified.

## DECLARATION

I, Oscar Ezekiel Njau do hereby declare to the Senate of the Nelson Mandela African Institution of Science and Technology that this dissertation is my original work and has neither been submitted nor being concurrently submitted for degree award in any other institution.

13/08/2024

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13/08/2024

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Prof. Revocatus Machunda — Date

13/08/2024

Dr. Anita Rugaika — Date

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## CERTIFICATION

The undersigned certify that have read the dissertation titled "*Fluoride and Pathogens Removal from Water Using Combined Electrocoagulation-inline-Electrolytic disinfection Process*" and recommend for acceptance by the Senate of the Nelson Mandela African Institution of Science and Technology for the degree award of Master's in Environmental Science and Engineering of the Nelson Mandela African Institution of Science and Technology.

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## DEDICATION

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## LIST OF ABBREVIATIONS AND SYMBOLS

| | |
|---|---|
| °C | Degree Celsius |
| µS/cm | Micro Siemens per Centimeter |
| CFU/100 mL | Colony Forming Unit per 100 milliliters |
| DBPs | Disinfection Byproducts |
| DC | Direct Current |
| DNA | Deoxyribonucleic Acid |
| DO | Dissolved Oxygen |
| EC | Electrocoagulation |
| $ECl_2$ | Electrolytic disinfection |
| FruVaSe | Fruits and Vegetation for all Seasons |
| GSM | Global System for Mobile Communication |
| L/h | Litre per Hour |
| MF | Membrane Filtration |
| mg/L | Milligram per Liter |
| mL | Milliliter |
| NM-AIST | The Nelson Mandela African Institution of Science and Technology |
| pH | Potentiometric Hydrogen Ion Concentration |
| SDGs | Sustainable Development Goals |
| SuMeWa | Sun Meets Water |
| TBS | Tanzania Bureau of Standards |
| TDS | Total Dissolved Substance |
| UN | United Nations |
| UV | Ultraviolet |
| WHO | World Health Organization |

# CHAPTER ONE

# INTRODUCTION

## 1.1 Background of the problem

In many developing countries around the world, access to safe drinking water is still a major concern (Baker, 2018). Drinking water is polluted from anthropogenic sources such as the dumping of untreated waste in the environment and natural 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). In Tanzania, Lake Momella is reported to have the highest fluoride level of about 690 mg/L compared to other water sources in Tanzania (Kitalika *et al*., 2018). The possible sources of high fluoride in Lake Momella are connected to volcanic activities on 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, co-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 the adsorbent's active sites (Akafu *et al*., 2019). Co-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 sulphate (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, efficiency is limited by clogging and requires continual maintenance and monitoring (Thakur & Mondal, 2017; Baker, 2018; Kumar *et al*., 2019; Giwa *et al*., 2021). The 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. 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 ($Cl_2$), ozone($O_3$), 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 and 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 electrolytic disinfection ($ECl_2$) process in one unit system SuMeWa (Sun Meets Water).

## 1.2 Statement of the problem

An electrochemical coagulation technology developed by Autarcon Company has been used for water disinfection in Tanzania, Egypt, Cameroon, India, Gambia, and Costa Rica. Because of the high fluoride in some water sources in Tanzania and the problem of water contamination by microbes, the system has been redesigned to achieve both defluoridation and disinfection of water in one unit system. However, the electrochemical (SuMeWa) system has not been optimized and the operating conditions for the combined (electrocoagulation-inline-electrolyticdisinfection) process have not been established. It is known that one of the most difficult aspects of the electrocoagulation process is passivation. This work aimed to solve the passivation problem and optimize the operating conditions of the SuMeWa system for combined defluoridation and disinfection of drinking water.

## 1.3 Rationale of the study

In Tanzania, bone char technique has been widely employed for defluoridation; however, disinfection remains a great challenge. The EC-$ECl_2$ system could be used as an alternative treatment unit, the two processes of defluoridation and disinfection of drinking water can happen in one combined unit. Therefore, it is worth doing this study in order to save and protect millions of people from endemic fluorosis and water bone diseases especially in developing countries where electrical power and safe drinking water is a challenge. The SuMeWa system could address these challenges because is a solar-powered water pumping and filtration unit.

## 1.4 Research objectives

### 1.4.1 General objective

To assess fluoride and pathogens removal from water using combined electrocoagulation-inline-electrolytic disinfection process.

### 1.4.2 Specific objective

(i) To assess the defluoridation capacity of electrocoagulation process at various operational conditions.

(ii) To evaluate the disinfection capacity of electrolytic disinfection process at various operational conditions.

(iii) To establish the optimal operating conditions of SuMeWa (EC-$ECl_2$) system for the removal of fluoride and disinfection.

## 1.5 Research questions

(i) How do the operating conditions (residence time, current density) and physical characteristics of water (pH, water conductivity) affect the defluoridation process during electrocoagulation?

(ii) How do the operating conditions (current density) and physical characteristics of water (pH, water conductivity) affect the disinfection process during electrolytic disinfection?

(iii) What are the optimal conditions of SuMeWa (EC-ECl2) system for consecutive removal of fluoride and pathogens from water?

## 1.6 Significance of the study

The results of this study contribute to Tanzania's achieving the sixth ($6^{th}$) goal of the United Nation (UN) sustainable development goals (SDGs) "ensure access to water and sanitation for all" that means, increasing access to clean drinking water and sanitation services by 2030. The presented system can provide sustainable water solution that is accessible and affordable to the local communities.

## 1.7 Delineation of the Study

The present study optimized an already developed system and establishes the optimal operating conditions that improve the system's performance in both laboratory and field conditions under the continuous flow mode. Further, the study assessed fluoride and pathogens removal from water using combined electrocoagulation-inline-electrolytic disinfection process.

# CHAPTER TWO

# LITERATURE REVIEW

## 2.1 Introduction

Fluorine ($F_2$) is a highly reactive electronegative element that can be found in trace amounts in soil, water, plants, and animals (Kumar *et al*., 2019). The $F_2$ is never found free in nature due to its reactivity. Fluorides ($F^-$), can be found in a variety of minerals, including fluorapatite ($Ca_5(PO_4)_3F$), fluorspar ($CaF_2$), and cryolite ($Na_3AlF_6$) (WHO, 2017; Alkurdi *et al*., 2019). The oxidation state of fluoride determines it's toxicological and physiological behaviors. Elemental fluoride is the most toxic compared to its salts (Singh *et al*., 2016).

Leaching and dissolution of fluorine into groundwater occur due to weathering processes and water circulation within soil and rocks. Alkaline environment favors fluoride dissolution from rocks into groundwater (Kitalika *et al.,* 2018; Alkurdi *et al.*, 2019). Excess aqueous ionic concentrations in groundwater, such as sodium bicarbonates or sodium carbonates, also enhance fluoride dissociation activity. Fluoride ions are released when fluorite ($CaF_2$) reacts with bicarbonates ($HCO_3^-$) and precipitate calcium carbonate ($CaCO_3$) as indicated in Equation 1 (Malago *et al*., 2017; Kitalika *et al.*, 2018; Alkurdi *et al*., 2019).

$$CaF_2 + 2HCO_3^- \rightarrow CaCO_3 + 2F^- + CO_2 + H_2O \quad (1)$$

Higher fluoride levels in the aquatic environment can also be attributed to anthropogenic activities such as discharge from fertilizer industry, phosphate fertilizer application, agricultural runoff, and byproduct released from aluminium smelters (Mobeen & Kumar, 2017).

According to WHO guidelines, the maximum allowable fluoride level in drinking water is 1.5 mg/L (WHO, 2017). Studies have reported that the consumption of water containing high fluoride cause diseases including fluorosis (Fig. 2), hip fractures, arthritis, osteoporosis, polydipsia, and infertility (Singh *et al*., 2016; WHO, 2017; Kumar *et al*., 2019). The prolonged accumulation of fluoride can induce changes in the deoxyribonucleic acid (DNA) structure of an individual (Kumar *et al.,* 2019). Thus, fluoride determination is important due to its implications on health as well as in pharmaceutical formulations (Singh *et al*., 2016).

Fecal coliform bacteria are a group of thermotolerant bacteria, the most common member being *Escherichia coli*, that are associated only with fecal material originating in the intestinal tract of warm-blooded animals (Mohamed *et al.,* 2016). The existence of fecal coliforms in the water sources indicates that the water has been polluted by human or other warm-blooded animals' fecal

matter (Kihupi *et al.*, 2016; WHO, 2017; EWURA, 2020). Fecal contamination is an indicator that a possible health risk occurs for individuals exposed to that water, some waterborne pathogenic diseases including dysentery, typhoid, viral and bacterial gastroenteritis, hepatitis A, and fever are expected (Kihupi *et al.*, 2016; WHO, 2017). Tanzania Bureau of Standards (TBS) (TZS 789:2016) and WHO (2017) recommend drinking water to have fecal coliform (FC) of 0 Colony Forming Unit per 100 ml (CFU/100 mL) at 44°C (Kihupi *et al.*, 2016; WHO, 2017; EWURA, 2020).

## 2.2 Fluoride occurrences in Tanzania

Tanzania and some rift valley countries are affected by high fluoride concentration in both surface and groundwater (Istituto-Oikos, 2011; Mwakabona *et al*., 2014; Chacha *et al*., 2018). Studies reported that, about 30% of drinking water in Tanzania has more than 1.5 mg/L fluoride (Fawell *et al.*, 2006; Kitalika *et al*., 2018). There is no uniform distribution of fluoride concentration throughout the country-in fluoride prone areas (Malago *et al*., 2017; Chacha *et al.*, 2018). In the northern part (Arusha, Manyara and Kilimanjaro) of Tanzania, fluoride pollution is a serious problem (Ghiglieri *et al.*, 2010; Mwakabona *et al.*, 2014; Masawe *et al.*, 2019). The typical high fluoride in water sources has been associated with volcanic activities on rift valley zones of Mount Kilimanjaro and Meru with the most affected areas being Arusha, Moshi, Manyara, Singida and Shinyanga (Malago *et al*., 2017; Kitalika *et al*., 2018; Wagutu *et al.*, 2018).

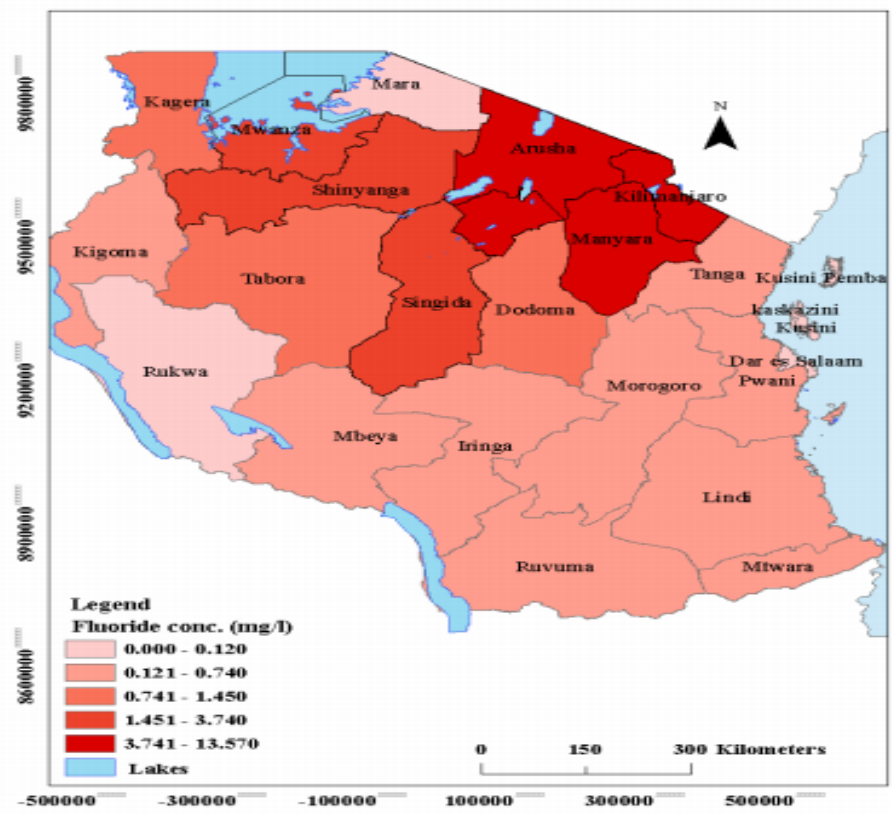


**Figure 1: A map of Tanzania showing fluoride distribution by regions (Malago *et al*., 2017)**

**Figure 2: Dental fluorosis in children (Kumar *et al.*, 2019)**

### 2.3 Estimation of health risk of excess fluoride intakes

Continuous ingestion of potable water with average fluoride concentration around 1.0 mg/L has been shown to strengthen teeth and bones. However, concentrations ranging from 1.5 to 4 mg/L are undesirable since prolonged consumption will cause dental fluorosis which is characterized by mottling and browning of teeth (Ahada & Suthar, 2019; Kumar *et al.*, 2019).

**Figure 3: Crippling (www.alamy.com/E8RXRP)**

In addition to that, consumption of the water with fluoride levels between 4.0 and 10 mg/L induces skeletal fluorosis while water with fluoride levels above 10 mg/L may cause crippling skeletal fluorosis which is characterized by skeleton deformity and weakening of bones (Fawell *et al.*, 2006; WHO, 2017; Ahada & Suthar, 2019).

**Table 1: Summary of health problems associated with fluoride ($F^-$) ingestion in individuals**

| $F^-$ in mg/L | Potential Health Problems |
|---|---|
| 0.0 – 0.5 | Teeth susceptible to cavities. |
| 0.5 – 1.5 | Ideal concentration for healthy teeth and bone. |
| 1.5 – 4.0 | Dental fluorosis in children: mottling of enamel. |
| 4.0 –10 | Skeletal fluorosis: pain in bones, pitting of enamel. |
| 10> | Crippling skeletal fluorosis, chronic pain, bone deformities, possibly cancer. |

**Fawell *et al.* (2006), WHO, (2017) and Ahada and Suthar (2019)**

Chemistry behind the positive and negative impacts of fluoride on the skeletal structure composition (hydroxyapatite) is based on the ion exchange reactions between $F^-$ and hydroxide ($OH^-$) ions. The substitution of $OH^-$ with $F^-$ ions, produce acid resistant structure, fluoroapatite ($Ca_5(PO_4)_3F$), Equation 2.

$$Ca_5(PO_4)_3OH + F^- \rightarrow Ca_5(PO_4)_3F + OH^- \quad (2)$$

Fluoroapatite prevents dental caries compared to hydroxyapatite ($Ca_5(PO_4)_3OH$) by offering a protective layer to the tooth enamel against acids from foods.

**Figure 4: Health and caries teeth (Charlie Sung, 2019)**

Long-term fluoride consumption will influence the reaction to go beyond phosphate replacement, as seen in Equation 3. Calcium decafluoride ($Ca_5F_{10}$), is a hard and brittle substance which is unsuitable for skeletal structural functions (Malago *et al.*, 2017).

$$Ca_5(PO_4)_3F + 9F^- \rightarrow Ca_5F_{10} + 3PO_4^{3-} \quad (3)$$

## 2.4 SuMeWa system

The SuMeWa|SYSTEM (from Sun Meets Water) developed by German (AUTARCON) company is a solar driven drinking water supply technology that produces clean and safe drinking water through EC processes. The electrolytic formation of disinfectant from raw water has been its

unique property. The residual disinfectant helps to ensure complete disinfection (reservoir effect). The disinfectant production is automatically adjusted to the appropriate level based on the water quality (Autarcon, 2012; Franklin & Livingston, 2015). Since the system is based on continuous flow processes, operational parameters such as initial fluoride concentration, flow rate, current density, initial pH, and supporting electrolyte are important as they influenced the removal efficiency of pollutants (Dura, 2013; Thakur & Mondal, 2017; Apshankar & Goel, 2018; Mureth *et al.*, 2021).

This system has been installed in Egypt, Cameroon, India, Gambia, Costa Rica, and Tanzania for the removal of iron, manganese, arsenic, turbidity, and pathogens from drinking water (Franklin & Livingston, 2015). The system has not been tested for fluoride removal, therefore, Wydra *et al.* (2019) and Mureth *et al.* (2021) proposed the optimization of the electrocoagulation process using a one dimensional electrode.

The advantages of this system compared to other water treatment systems applied in Tanzania are explained in Table 2. This includes low maintenance cost, no filtration media renewal required for up to 10 years, remote sensing via GSM chip (Global System for Mobile Communication), automatic filter backwash to remove the filtrate materials and online controlled media regeneration, chemical-free, solar operated, and reservoir effect (Autarcon, 2012). It is designed with specifications that include inline electrolytic (electrocagulation and electrochlorination) cells, power supply (120 W), maximum pumping head of 70 m, and maximum flow rate of 400 L/h (SuMeWa, 2018; Wydra *et al.*, 2019).

**Table 2: Comparison of various drinking water treatment technologies in Tanzania**

| Approaches | Defluoridation | Disinfection | Residual effect | Auto-filter backwash | Controllability of water quality | References |
|---|---|---|---|---|---|---|
| Bone char technique | + | - | - | - | - | Kaseva (2006) |
| Nalgonda technique | + | - | - | - | - | Dahi *et al.* (1996) |
| Nanotechnology | + | + | - | - | - | Alkurdi *et al.* (2019) |
| EC-$ECl_2$ process (SuMeWa) | + | + | + | + | + | Autarcon (2012) |

**Where: + = Yes, and - = No**

However, various techniques, including bone char and nanofiltration, have been developed in Tanzania to remove fluoride and pathogens from drinking water (Alkurdi *et al.*, 2019). The application of bone char adsorbent has been reported to effectively remove fluoride, while

pathogens remain a concern, and religious objections have been a major obstacle to its implementation (Kaseva, 2006; Kiagho *et al.*, 2016; Mobeen & Kumar, 2017). Nanofiltration selectively eliminate environmental contamination (chemical-contaminants, organic-matter, turbidity, bacteria) in water but it only performs the goal at household levels in impacted areas (Ndé-Tchoupé *et al.*, 2019). Therefore, this situation calls for innovative and sustainable system that can simultaneously remove fluoride and pathogens in drinking water.

### 2.5 Defluoridation by electrocoagulation process

The EC process is still a simple and successful technique for eliminating the flocculating agent produced by electro-oxidation of a Fe or Al sacrificial anode. The treatment in this process is carried on without addition of any coagulant or flocculant chemicals, therefore, it reduces the amount of sludge which must be disposed off (Emamjomeh & Sivakumar, 2009; Takdastan *et al.*, 2014; Ndjomgoue-Yossa *et al.*, 2015). During EC process, direct current is passed through Al electrodes which dissolve to produce $Al^{3+}$ that combine with $OH^-$ to form metal hydroxyl in the water which is soluble under definite pH values. Multiple reactions take place simultaneously as water pass through EC cell. In anode, surface-metal ions are driven into the water (Takdastan *et al.*, 2014). Through oxidation processes, aqueous $Al^{3+}$ species will be produced by electrolytic dissolution of sacrificial Al anodes, and the anodic reaction is shown by Equation 4.

$$\text{Anode: } Al \rightarrow Al^{3+} + 3e^- \tag{4}$$

At the surface of cathode, water hydrolyzed into hydrogen gas ($H_2$) and hydroxyl ion ($OH^-$**)** by the following reaction:

$$\text{Cathode: } 2H_2O + 2e^- \rightarrow H_2 + 2OH^- \tag{5}$$

The electron from the anode is consumed by hydrogen ions ($H^+$) in the cathode during hydrolysis to reduce $H^+$ into hydrogen gas ($H_2$).

$$\text{Cathode: } 2H^+ + 2e^- \rightarrow H_2 \tag{6}$$

Electrons flow from cathode to anode to destabilize the surface charges of suspended solids and most of $H^+$. As the reaction begins metal ions, $Al^{3+}$ complex with $OH^-$ to form large flocs including metals and other water contaminants (Guzmán *et al.*, 2016).

$$Al^{3+} + 3OH^- \rightarrow Al(OH)_3 \tag{7}$$

The precipitate formed is believed to adsorb fluoride strongly to form the aluminium complexes ($Al(OH)_{3-x}F_x$.)

$$Al(OH)_3 + xF^- \rightarrow Al(OH)_{3-x}F_x + xOH^- \quad (8)$$

The hydrogen bubbles help to separate and lift the flocs, this makes EC more efficient and effective. The flocs might float or settle at the bottom of water as the reaction continues to form a large floc. Water will be filtered through media bed ultrafiltration (Emamjomeh *et al.*, 2011).

### 2.6 Disinfection by electrodisinfection processes

The removal of pathogens (disinfection) has become an important part of water treatment because water contamination by pathogens is a serious problem in developing countries (Baciu *et al.*, 2015; WHO, 2017). Chlorination is commonly used as disinfection process, however, ozonation, chloramination, ultraviolet radiation and chlorine dioxide can be also applied (WHO, 2011). These methods are effective in killing bacteria and some might inactivate many protozoa, including *Giardia, Cryptosporidium* and viruses (WHO, 2011; Ghernaout *et al*., 2019). The most practical approach for successful removal or inactivation of pathogenic microorganisms is filtration supported by coagulation and flocculation, followed by disinfection (single or combination of disinfectants) (WHO, 2011).

Chlorine damages the cell membrane of microorganisms and inactivates them. Because the cell membrane is weakened, chlorine can enter the cell and damage cell respiration and DNA function (two processes necessary for cell survival). The $ECl_2$ technique does not require additional chemicals other than common salt, but the level of chlorine can be adjusted depending on the needs and the energy required, which is not high (Ghernaout & Elboughdiri, 2019).

#### 2.6.1 Production of free chlorine from water containing chloride

Chloride ($Cl^-$) ions dissolved in raw water are electrochemically converted into chlorine gas (Equation 9).

$$2Cl^- \rightarrow Cl_2 + 2e^- \quad (9)$$

Chlorine gas rapidly hydrolyzes in water to form hypochlorous acid (HClO), which dissociates into hypochlorite ions ($ClO^-$) and hydrogen ions ($H^+$). According to health based guideline, 5 mg/L is the free chlorine dosage needed for public water supply. Moreover, to avoid detectable customer rejection, the concentration of free chlorine should be lower than 5 mg/L (WHO, 2017).

$$Cl_2 + H_2O \rightarrow HClO + H^+ + Cl^- \quad (10)$$

Hypochlorous acid dominates at pH levels below 6.5, while at pH levels above 8.5, the hypochlorite ions dominate. Water becomes more acidic as its pH decreases due to production of hydrogen ions. The more effective disinfectant form is hypochlorous acid; therefore, a lower pH is suitable for disinfection. The chemical Equation 11 is a pH-dependent equilibrium formed between hypochlorous acid and the hypochlorite anion (Kraft, 2008).

$$HClO \leftrightarrow ClO^- + H^+ \quad (11)$$

However, increasing disinfectant contact times with storage water after disinfection before supply to consumers can improve disinfection. This is the most important process particularly for more resistant microorganisms, like *Giardia* and some viruses (WHO, 2011).

### 2.6.2 Simplified reaction mechanisms for free chlorine

According to nomenclature of water disinfection, the 'free chlorine' or 'active chlorine' is referred as the total number of hypochlorous acid and hypochlorite concentrations. Consider the following equations (12) and (13) for the disinfecting effect of active chlorine which depend on the disappearance of oxygen atom.

$$HClO \rightarrow O + Cl^- + H^+ \quad (12)$$

$$ClO^- \rightarrow O + Cl^- \quad (13)$$

The production of $Cl^-$ will compensate for the chloride ions consumed during disinfection by electrochemical free chlorine production. As a result, it restores the overall change in the chemical composition of the water after electrochemical water disinfection (Kraft, 2008).

## 2.7 Anodic passivation

In EC process, an anodic passivation is regarded as the greatest operational challenge. The formation of passive oxide layer over the anode's surface tends to prevent the production of adequate coagulants. The passive layer on a metal or alloy has a good corrosion resistance. In EC processes, this layer raises the applied potential, resulting in energy loss (Mohora *et al*., 2012). The formation of an oxide or passive layer on the surface of the electrodes can further reduce the efficiency of the EC process (Mollah *et al.*, 2004).

# CHAPTER THREE

# MATERIALS AND METHODS

## 3.1 Study area

This study has been conducted in Meru district, Arusha: Tanzania and the laboratory work has also conducted at the Nelson Mandela African Institution of Science and Technology (NM-AIST). The study design is a laboratory based analytical study.

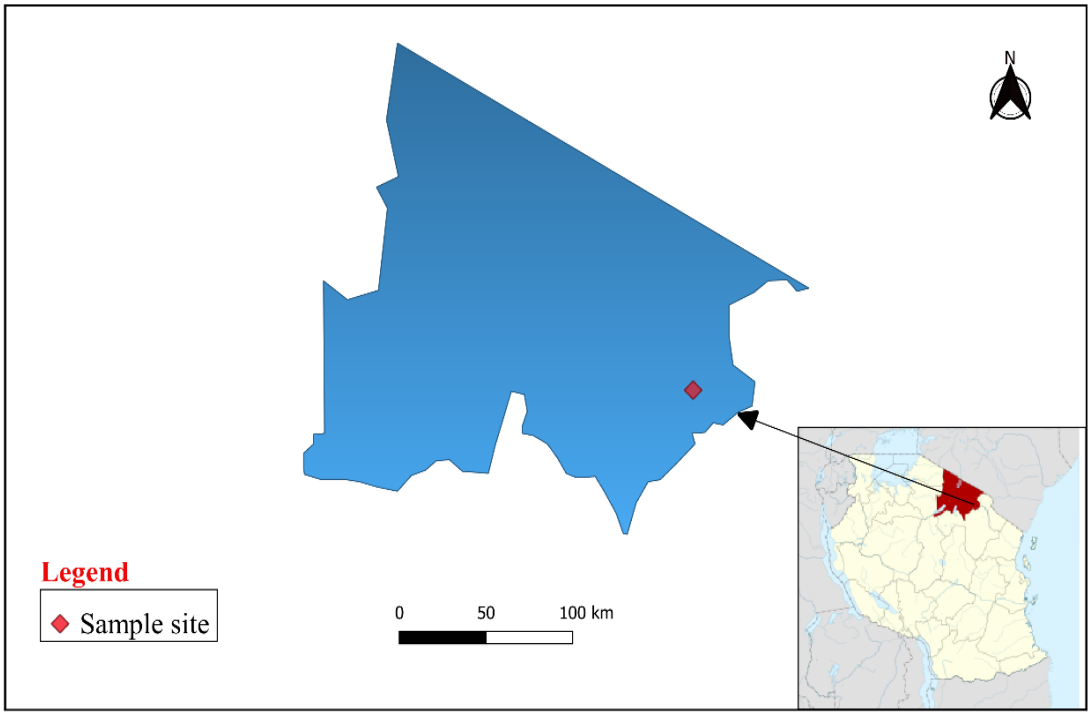


**Figure 5: Arusha Map showing the study area**

## 3.2 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 mg/L and 12.3 mg/L as initial fluoride concentrations of raw water from 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 + sodium chloride (NaCl) salt). The NaF (12.3 mg$F^-$/L) and NaCl (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. In an additional laboratory experiment the effect of resident time on treated water before filtration was tested with 36.5 mg/L initial fluoride concentration. The 6 N sodium hydroxide and 2 N hydrochloric acid

solutions were added for pH adjustment (values 5.0 to 7.5). The 52 g of the M-FC Agar base powder is suspended in 1 L of purified water and mixed thoroughly for media preparation. With frequent agitation, the mixture was heated and boiled for 1 minute to completely dissolve the powder. The 10 mL of a 1% solution of Rosolic Acid was added in 0.2 N NaOH and continuously heated for 1 minute but not autoclaved. Stable, typical control cultures were used to test the performance of finished product samples.

### 3.3 Experimental setup

The SuMeWa system is a solar-powered water pumping and filtration unit that has been developed to defluoridate and disinfect drinking water in a one-unit using the EC-$ECl_2$ process. As seen in Fig. 6, the experimental setup was designed and constructed with inlet and outlet/reservoir tanks of 500 L each, electrocoagulation (EC) cell, filter, and electrochlorination ($ECl_2$) cell. The defluoridation and disinfection of water were the two steps of EC-$ECl_2$ process that performed at EC (aluminum electrodes) and $ECl_2$ (mixed oxide electrodes) cells, respectively. The filter shown in Fig. 6 contains AFM® 0 with particles size of 0.025-0.05 mm. Particles, here mainly aluminum fluoride particles, are retained in the media. 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.

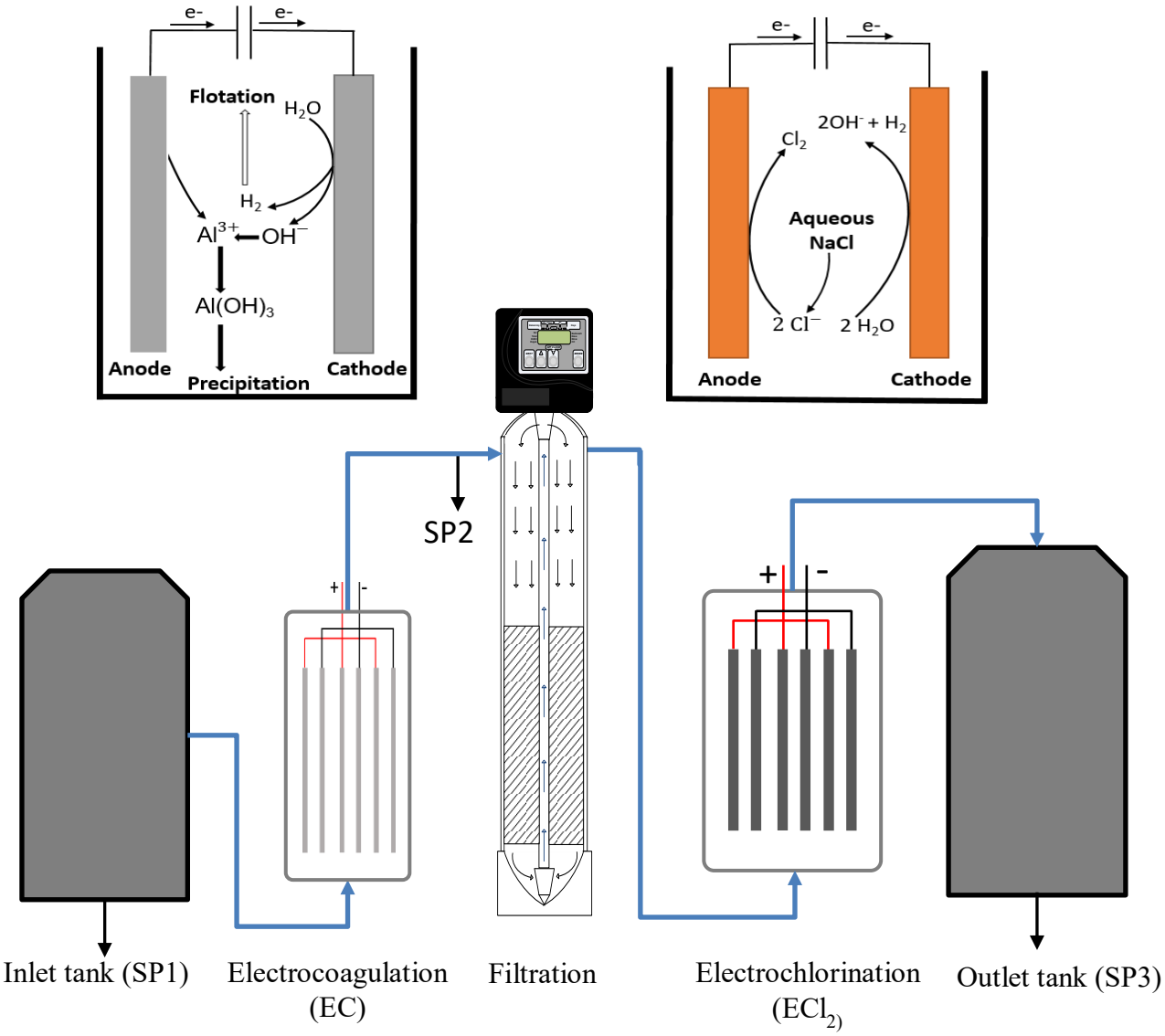


**Figure 6: Schematic diagram of water treatment (SuMeWa) system**

Water flows from the inlet tank through electrocoagulation cell (size, 1 L) made up of 3 aluminum plates each (purity of Al, 99.7%). The configuration of the aluminium electrode is shown in Fig. 7. The area of each plate was 85 $cm^2$, with a gap of 5 mm. The total surface area of the stack was 510 $cm^2$ and the effective surface area of the electrode was 425 $cm^2$ each. Electrical connection was made in a parallel monopolar connection (Fig. 8). 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 EC cell and the coagulant $Al(OH)_3$ was generated to allow defluoridation.

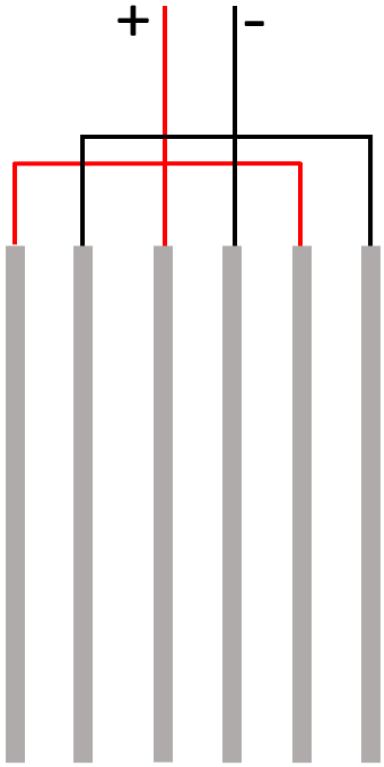


**Figure 8: Al electrode configuration**

**Figure 7: Power supply in a parallel monopolar connection**

Six electrodes were used to pass Direct Current (DC) from a DC power supply (0–30 V, 1.0-7.8 A) to the water resulting in current densities between 9.4 mA/cm² 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 by four pairs of dimensional stable titanium electrodes coated with oxides of ruthenium and iridium (MOX-electrodes) (Otter *et al.*, 2017; Kunz *et al.*, 2018). The surface area of each anode/cathode plate was 640 $cm^2$, consider Fig. 9. In the $ECl_2$ cell, chlorine gas was produced electrolytically from dissolved chloride ions for disinfection.

**Figure 9: Titanium electrode configuration**

### 3.4 Analytical techniques

The defluoridation in the laboratory was performed in continuous mode using fluorinated water made from a synthesized solution of Sodium Fluoride (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 multi-parameter. 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 NaCl, 57 mL glacial acetic acid, 4 g 1,2 cyclohexylenediaminetetraacetic (CDTA), and 125 mL 6 N NaOH were dissolved in 1L of distilled water by stirring until pH 5.3–5.5 was reached) and 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. 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 membrane filtration (MF) technique with 0.45 µm pore size microbial filter paper. The counting of fecal coliforms before and after treatment was conducted to determine the efficiency of the process. Following the MF procedure, enriched lactose (M-FC Agar Base) medium and an incubation temperature of 44.5 $\pm$ 0.2°C for 24 hours were applied. Fecal coliform concentration was reported in terms of the number of bacterial colonies per 100 mL of water sample 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 used this study are presented in Table 3.

**Table 3: Ranges of the experimental parameters**

| | |
|---|---|
| **Common EC-E$Cl_2$ 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 electro-coagulation cell (L) | 1 |
| EC residence time (min) | 1.5–6.0 |
| **Electrodisinfection parameters** | |
| Current density (mA/$cm^2$) | 1.6–9.4 |
| Anode area, electrochlorination cell ($cm^2$) | 640 |
| Volume of electrochlorination cell (L) | 2 |
| $ECl_2$ residence time (min) | 3-12 |

### 3.5 Data analysis and interpretation

Statistical data analysis such as mean and graphics were done by using Excel software. The efficiency of defluoridation was determined via Equation 11.

$$E = \frac{C_o - C_f}{C_o} x\ 100\% \quad (14)$$

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

### 3.6 Specific energy consumption

The total specific energy consumption (SEC) of the EC cell, $ECl_2$ cell and pump for a given flow rate was calculated by Equation (15).

$$\text{SEC}\left(\frac{\text{kWh}}{\text{m}^3}\right) = \frac{\text{current} \times \text{time} \times \text{voltage}}{\text{Flow rate}} \quad (15)$$

# CHAPTER FOUR

# RESULTS AND DISCUSSION

## 4.1 Water quality of the 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 4. Data values for treated water at flow rate (10-40 L/h), current density (9.4-18.4 mA/cm$^2$), pH (5.0-7.5) and initial fluoride concentration (1.91-12.3 mg/L) were presented the final conditions of the water after both EC and $ECl_2$ processes.

**Table 4: Characteristics of synthetic and treated water**

| Parameter | Synthetic water | Treated water |
|---|---|---|
| pH | 6.8 | 7.26 ± 0.16 |
| Temperature ($^0$C) | 25 ± 3 | 25 ± 3 |
| Conductivity (µS/cm) | 954 | 814.38 ± 3.71 |
| Total dissolved solids (mg/L) | 441 | 409 ± 1.09 |
| Dissolved oxygen (mg/L) | 6.34 | 5.95 ± 0.14 |
| Fluoride (mg/L) | 12.3 | 0.164-1.44 |
| Chloride (mg/L) | 240.11 | 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 | 0.7-2.23 |
| Fecal coliform (CFU/100mL) | 36-19 700 | 0 |

## 4.2 Passivation of the EC electrode

Electrode passivation is a limiting factor for the usage of EC in this system. A study was conducted to optimize the conditions of EC for fluoride removal while at the same time using the chloride ions from NaCl to break passivation in the EC cell and enhance chlorine production in the electrochlorination process. The results shows that the passivation has been prevented on aluminium electrodes and the sufficient amount of chlorine gas was produced for water disinfection as explained in Section 4.6.2 and 4.7.1, respectively.

## 4.3 Effect of current density 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 achieved 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 10 shows that, the released concentrations of aluminum ions into solution were increased with current density and eventually reduced the fluoride concentrations. 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 minutes, a residual fluoride concentration of 1.5 mg/L was achieved. Aluminum concentration reached 13.97 ± 2.98 mg/L (measured after EC cell). At a flow rate of 20 L/h, electrolytic time of 50 minutes 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, Appendix 9 (c). As a result, at 10 L/h, the optimum current for defluoriation 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 an increased in current density improved the defluoridation. This agrees with the findings reported by Ghosh *et al*. (2008).

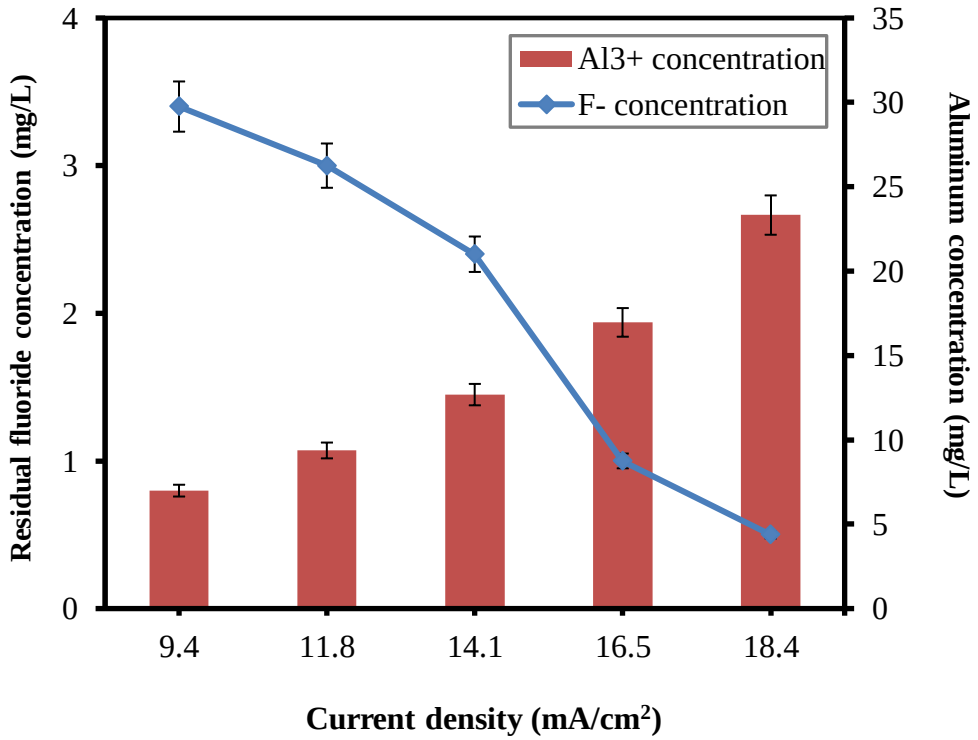


**Figure 10: Effect of current density (9.4-18.4 mA/cm$^2$) on the production of aluminum ($Al^{3+}$) concentrations at a constant flow rate (20 L/h), and initial fluoride concentration (12.3 mg/L)**

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 0.070 to 0.033 mg/L with average of 0.055 ± 0.0056 mg/L at flow rates ranging from 10 to 40 L/h respectively, which were less than the WHO and TBS acceptable limit (0.2 mg/L).

### 4.4 Effect of flow rate on defluoridation

The influence of flow rates on defluoridation was investigated in this study by altering the flow rates from 10-40 L/h equivalent to residence times of 1.5-6.0 minutes at various initial fluoride concentrations and a fixed current density of 9.4 mA/cm$^2$. As seen in Appendix 2 (c), 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 0.191 to 0.457 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 EC cell with 1 L volume (size) have the residence time of 6, 3, 2, and 1.5 minutes respectively. Figure 11 shows that the higher the flow rate, the lower the time materials spend 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 and Sivakumar (2009). However, fluoride concentrations decreased with the increase in electrolysis time as more coagulants were formed.

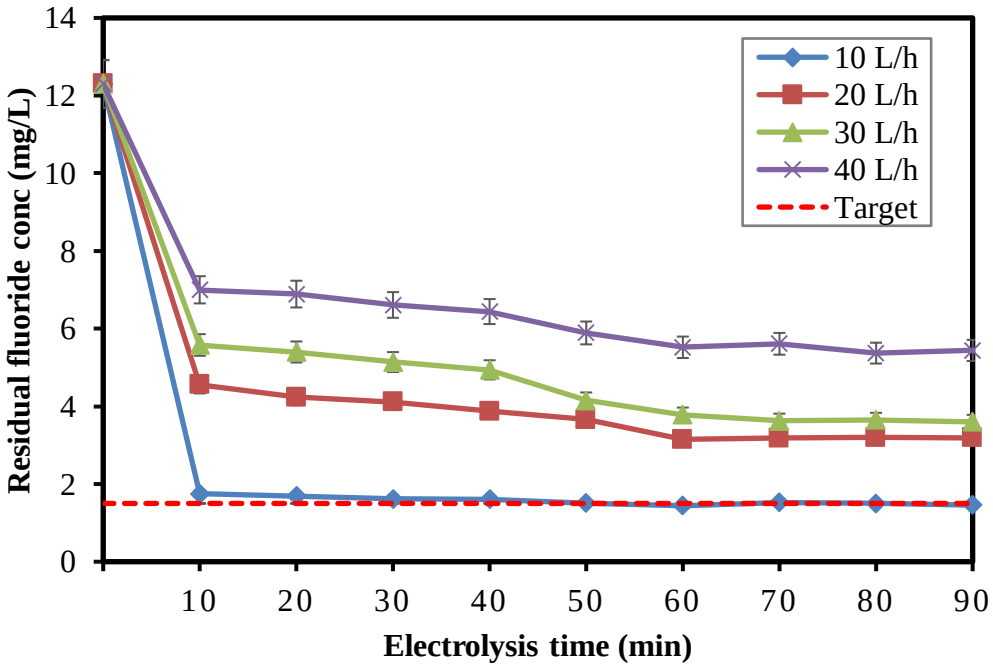


**Figure 11: Effect of flow rates (10-40 L/h) on residual fluoride concentrations at constant current density (9.4 mA/cm$^2$), pH (6.8), initial fluoride concentration (12.3 mg/L), and varied electrolysis time (10-90 min)**

### 4.5 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 1.91 mg/L 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 2 (c)). 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 12 showed reduced removal efficiency as the fluoride concentration was increased from 1.91 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 achieve the maximum efficiency.

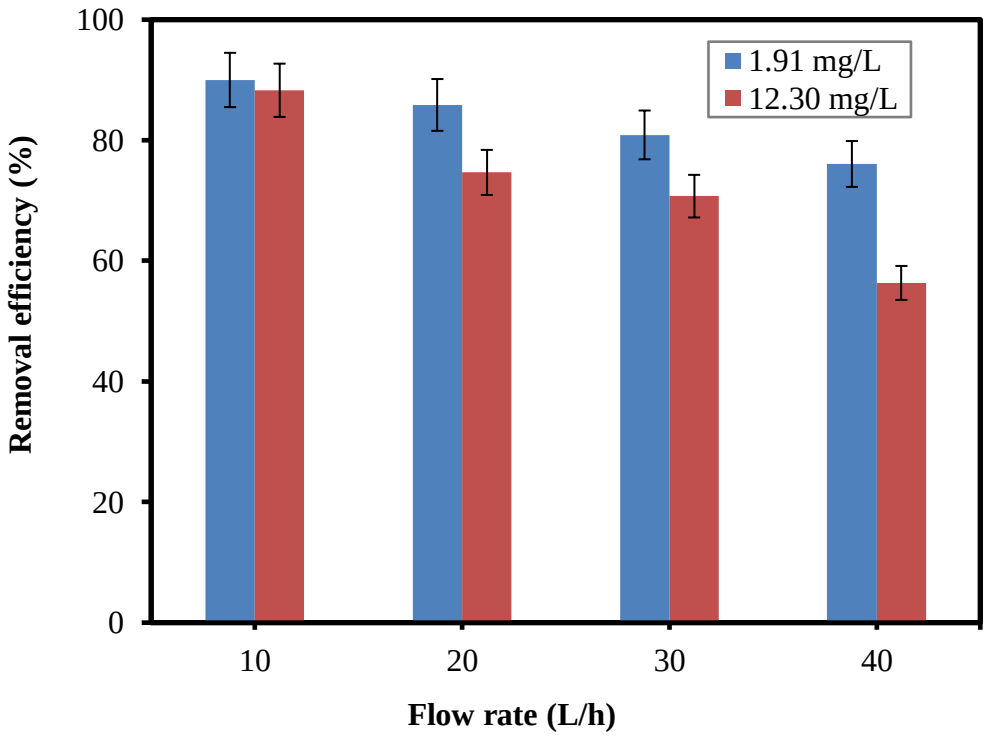


**Figure 12: Effect of initial fluoride concentrations (mg/L) on defluoridation efficiency after EC cell at constant pH (6.8), current density (9.4 mA/cm$^2$), and varied flow rates (10-40 L/h)**

Figure 13 shows that residual fluoride concentration after the EC cell ($SP_2$) was found to be lower compared to that after the filtration step ($SP_3$). This study believed that, an immediate filtration after defluoridation experiment at $SP_2$ reduced the reaction time needed by coagulants and the aqueous fluoride to stabilize the chemical bonds formed between them for enhancing successful defluoridation. There is limited information on the bond strength of aluminum complexes (particularly $Al(OH)_3$ and $F^-$) with filter media involving an EC-$ECl_2$ process. According to

Mousazadeh *et al*. (2021), fluoride is released into the solution to generate either $F^-$ free $Al(OH)_3$ or $Al(OH)^-_4$ due to the low stability of $Al_nF_m(OH)_{3n-m}$ in slightly alkaline solutions.

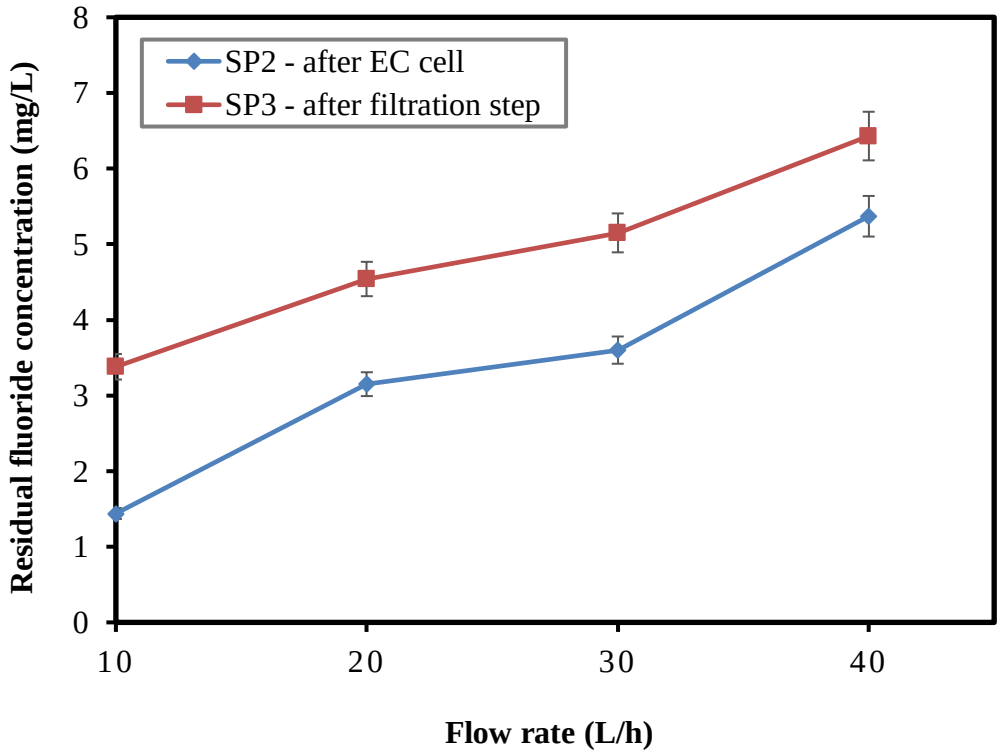


**Figure 13: Residual fluoride concentration after EC cell ($SP_2$) and filtration step ($SP_3$) at constant initial fluoride concentration (12.3 mg/L), current density (74 mA/cm$^2$), pH (6.8), and varied flow rates (10-40 L/h)**

In an additional experiment with a constant initial fluoride concentration of the synthetic water (36.5 mg/L), and the given operational parameter (current density: 9.4 mA/cm$^2$, flow rate: 10 L/h) the contact time of coagulant in treated water after EC was varied between 0 and 150 minutes in order to evaluate test the effect of residence time after the water passes through the electrocoagulation cell ($SP_2$) and before the filtration step. Figure14 shows that the residual fluoride concentration after $SP_3$ (filtered water samples) has been found to be lower compared to that of $SP_2$ (unfiltered water samples) after 150 minutes. The findings verify that the treated water by the EC must be retained for at least 150 minutes before being filtered. Increasing the residence time permits the chemical bonds between the coagulant ($Al(OH)_3$) and the aqueous fluoride ($F^-$) to stabilize, allowing for successful defluoridation.

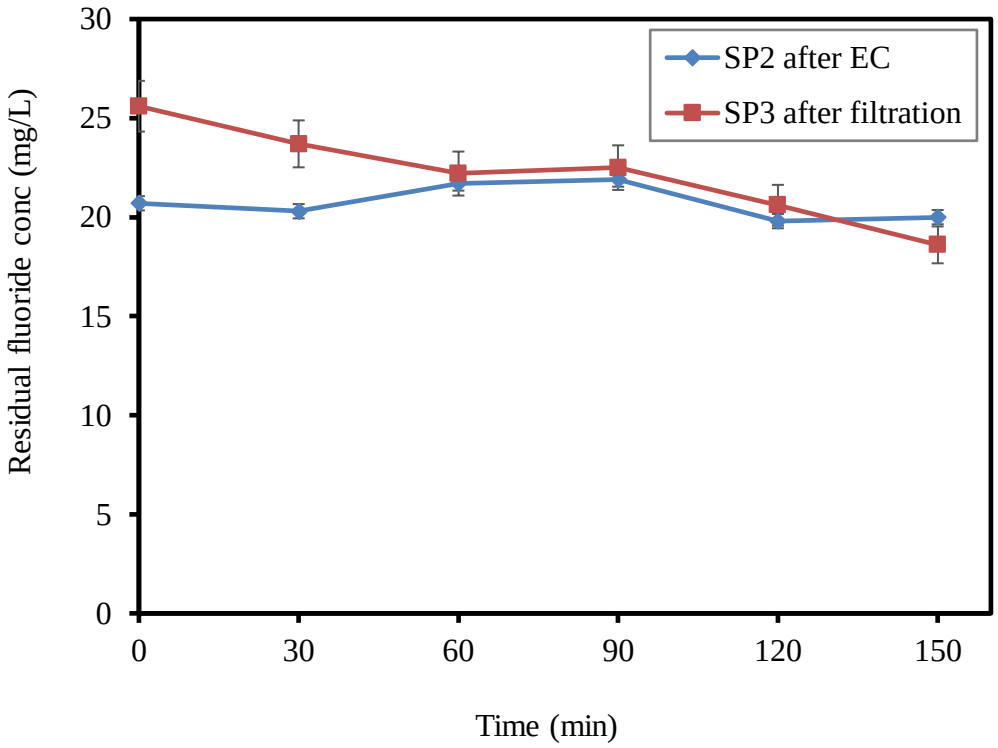


**Figure 14: Effect of residence time (0-150 min) on treated water before filtration step at constant initial fluoride concentration (36.5 mg/L), current density (9.4 mA/cm$^2$), pH (6.9), and flow rate (10 L/h)**

### 4.6 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. The $H^+$ ions react with $F^-$ ions to produce 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*., 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 to 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 9.4-18.4 mA/cm$^2$ at the EC cell. The lowest residual fluoride of 0.164 mg/L (98.7%) has been found at pH of 6 and current density of 18.4 mA/cm$^2$. This was attributed to the formation of sufficient form of $Al(OH)_3$. This study agreed with the batch findings reported previously by Mureth *et al*. (2021) that the optimum pH for defluoridation was 6. Figure 15 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 Fig. 14, a residual fluoride

concentration of 1.44 mg/L (88.29%) was achieved at a constant initial pH of 6.8 of raw water, current density (9.4 mA/cm$^2$), flow rate (10 L/h), and initial fluoride concentration (12.3 mg/L).

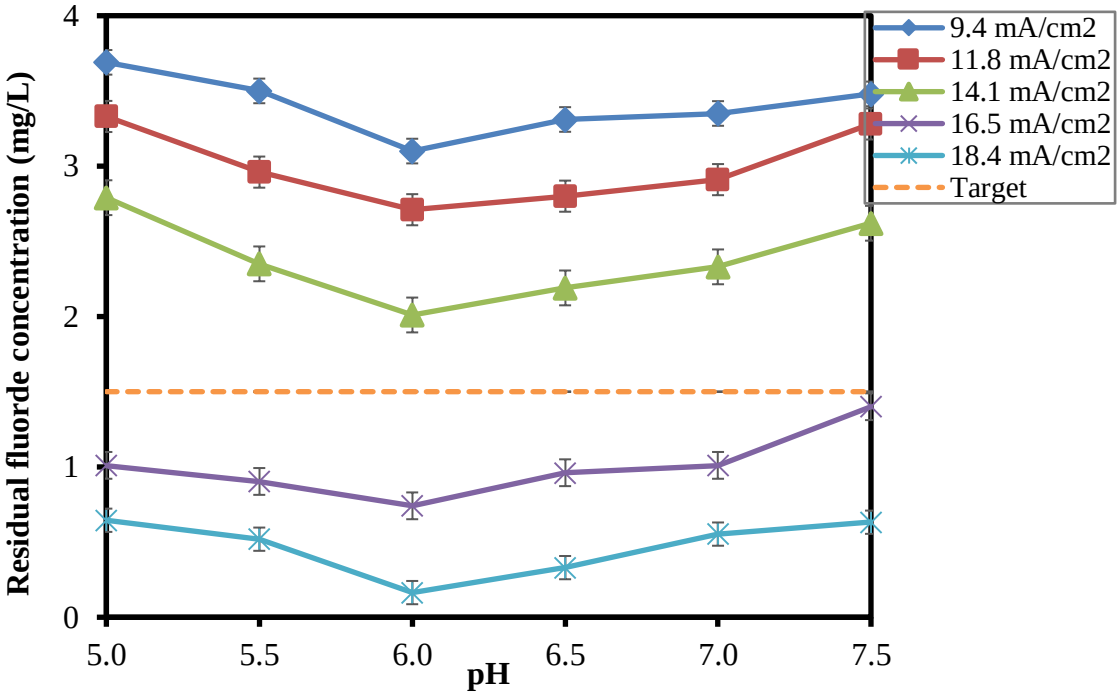


**Figure 15: Effect of initial pH (5.0-7.5) on residual fluoride concentration at constant flow rates (20 L/h), initial fluoride concentration (12.3 mg/L), and varied current density (9.4-18.4 mA/cm$^2$)**

Moreover, the initial pH of the feed water has been found to have no significant effect on the production of free chlorine. A slight variation of the produced free chlorine concentration was found at both the electrocoagulation cell ($SP_2$) and the $ECl_2$ cell ($SP_3$) of the system at a varied pH range of 5.0-7.5. Figure 16 shows that the free chlorine production has been observed to be influenced by the amount of applied current and not the water pH. According to this study, initial water pH has an effect on the speciation (specific chemical form) of the produced active chlorine species (hypochlorous acid or hypochlorite) but not on free chlorine production.

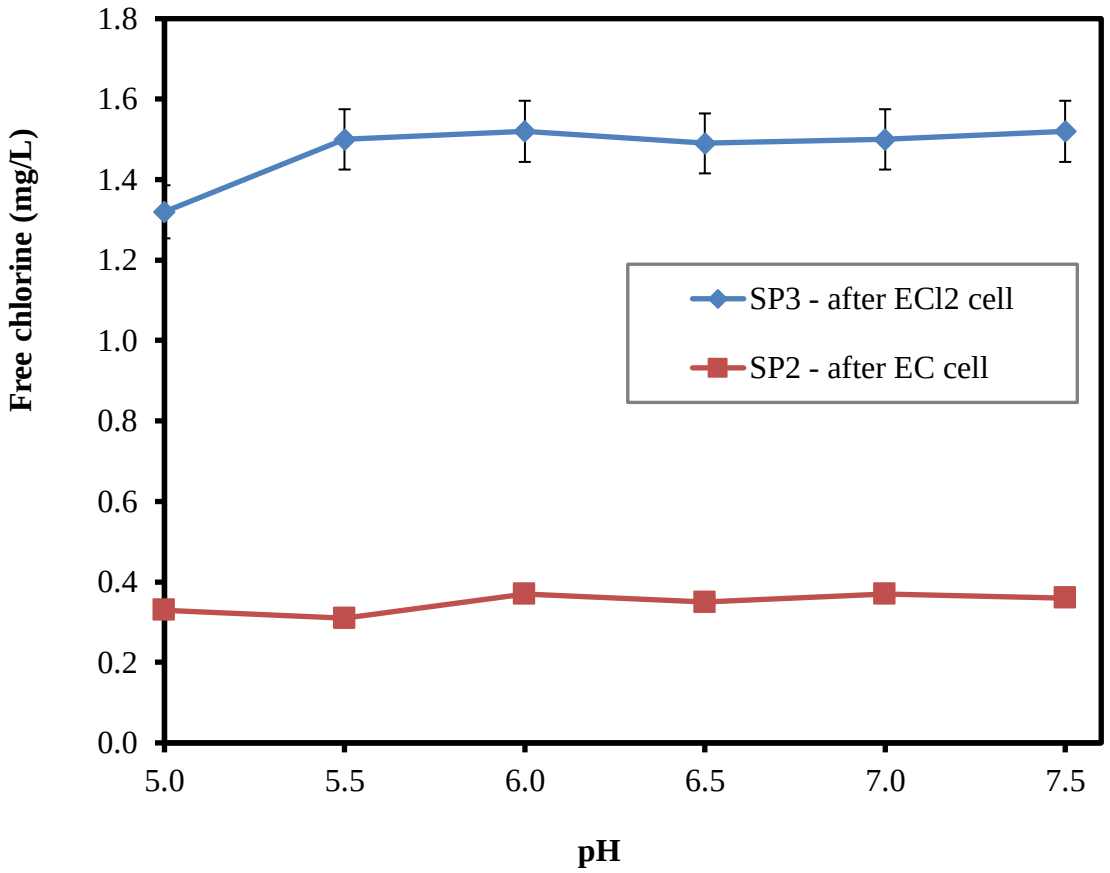


**Figure 16: Effect of initial pH (5.0-7.5) on the production of free chlorine at constant flow rate (20 L/h) and current density (9.4 mA/cm$^2$ for EC cell and 3.1 mA/cm$^2$ for $ECl_2$ cell)**

On the other hand, the final pH of the water was found to be slightly higher than the initial pH, as shown in Table 5. This means that during water treatment, cathodes generate extra basic related materials ($OH^-$) that are not incorporated into the formation of $Al(OH)_3$ or its complexes. Consider the chemical Equation (4):

$$Al_n(OH)_{3n(s)} + {}_mF^-{}_{(aq)} \rightarrow Al_nF_m(OH)_{3n-m(s)} + {}_nOH^-{}_{(aq)} \qquad (4)$$

The final pH of the optimum initial pH was found to be 6.63, which is within the WHO and TBS permissible limit of 6.5-8.5. As a result, pH adjustment is not necessarily required to meet the standards of the final treated drinking water.

**Table 5: Initial pH before treatment and final pH of the treated water at constant flow rates (20 L/h), initial fluoride concentration (12.3 mg/L), and varied current density (9.4-18.4 mA/cm$^2$)**

| Initial pH | Final pH | Change |
|---|---|---|
| 5.0 | 5.56 | +0.56 |
| 5.5 | 6.03 | +0.53 |
| 6.0 | 6.63 | +0.63 |
| 6.5 | 7.01 | +0.51 |
| 7.0 | 7.45 | +0.45 |
| 7.5 | 8.32 | +0.82 |

### 4.7 Effect of current density on disinfection

During an experiment using the $ECl_2$ cell (current density range of 1.6-9.4 mA/cm$^2$, pH of 6, and a flow rate of 20 L/h) water containing numerous fecal coliforms up to 19 700 CFU/100 mL was disinfected. Figure 17 shows the concentration of free and total chlorine with increasing current density passing through the $ECl_2$ cell. Moreover, even at the low range of current density 1.6-3.1 mA/cm$^2$, a great 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 to 4-log reduction. 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) but compromise the acceptable limit 1.5 mg/L of free chlorine concentration produced at 3.1 mA/cm$^2$, was sufficient to disinfect the water with 19 700 CFU/100 mL to the permissible limit set by WHO and TBS for treated drinking water of 0 CFU/100 mL (4-log reduction), Appendix 10(c). This study is supported by Ndjomgoue-Yossa *et al*. (2015) who demonstrated that microbial inactivation is influenced by the increase in current density. Therefore, the optimum current density for water disinfection with numerous pathogens in this setup is considered to be 3.1 mA/cm$^2$.

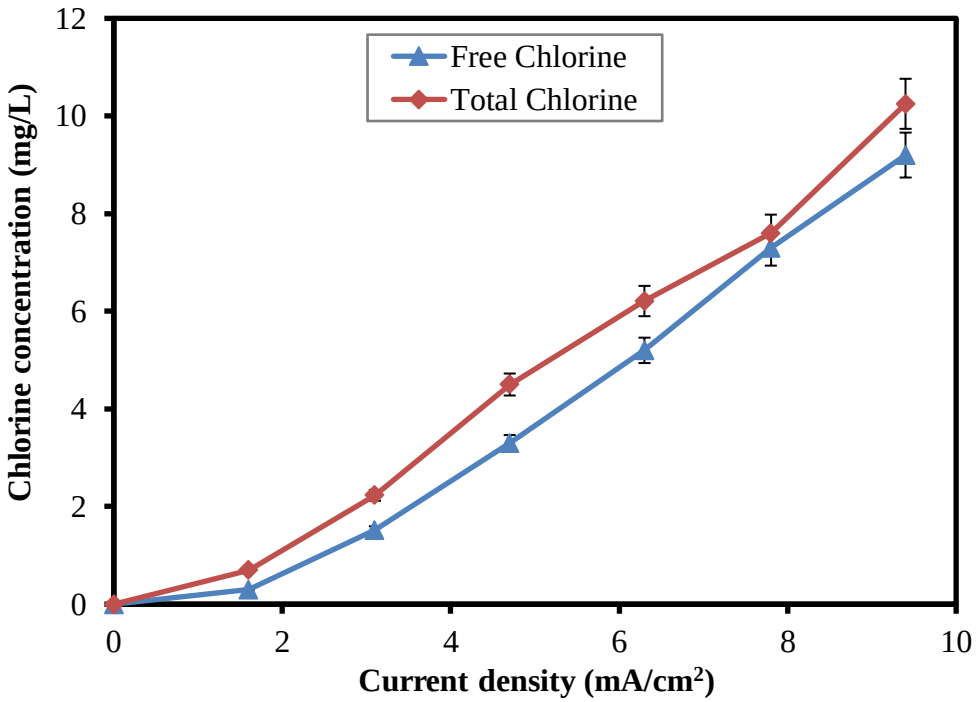


**Figure 17: Effect of current density (1.6-9.4 mA/cm$^2$) on disinfectants production at constant pH (6.0) and flow rate (20 L/h)**

### 4.8 Effect of supporting electrolyte

The 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). The NaCl can significantly reduce the effects of anionic species such as Sulphate ($SO_4^{2-}$) and bicarbonate ($HCO_3^-$). In fact, $SO_4^{2-}$ ions were recognized as a one of the agents of passivation, whereas $Cl^-$ causes the passive layer to decompose (Dura, 2013).

#### 4.8.1 Breaking of 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 electrolyte 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 aluminum electrode. In this study 99 and 165 mg/L of NaCl were added into the water to be treated to promote 60 mg/L 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 13).

$$2Al_2O_3 + 12Cl^- \rightarrow 4AlCl_3 + 3O_2 + 12e^- \quad (13)$$

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 18 shows the slight variation in the current density passing through the EC cell implies that the anodic passive layer has completely broken down. Also, at 100 mg/L of chloride concentration, enough free chlorine was electrochemically produced to efficiently disinfect water (consider, Fig. 17). This study was supported by Ndjomgoue-Yossa *et al*. (2015), who demonstrated that even at low chloride concentrations (100 mg/L), enough free chlorine is produced to disinfect water.

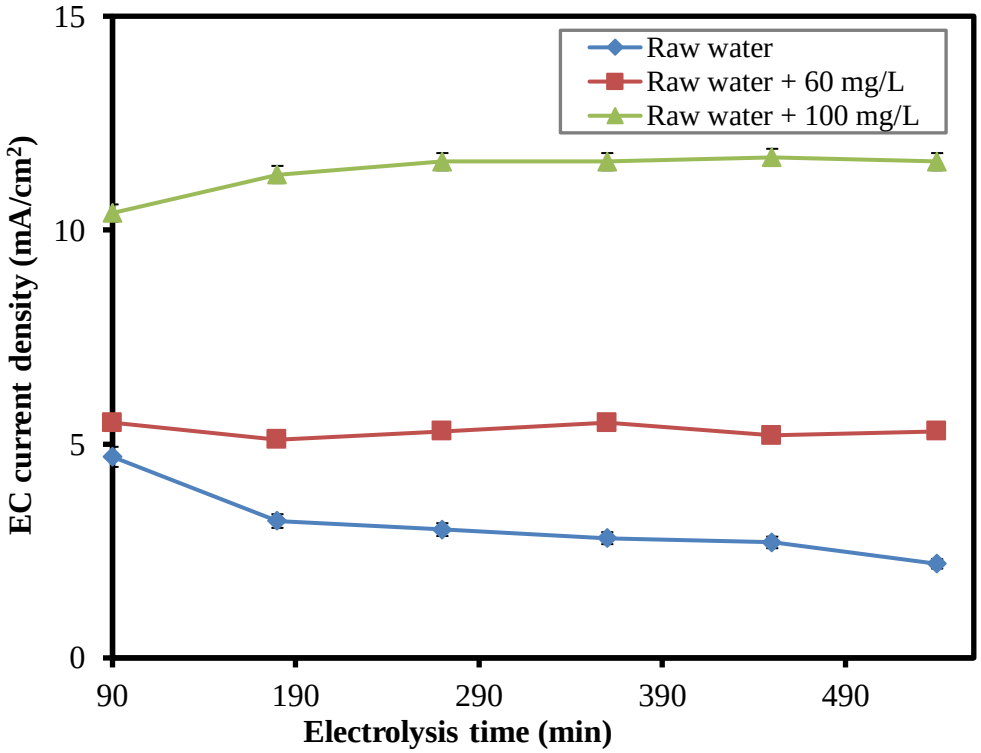


**Figure 18: Effect of supporting electrolyte, $Cl^-$ (60-100 mg/L) on current density passed through EC cell at constant pH 6.8**

### 4.8.2 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. The addition of the supporting electrolyte (99 to 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 (Fig. 18). Apshankar and Goel (2018) reported less energy consumption as the conductivity of the solution increased due to reduced resistance. However, on a laboratory scale the addition of supporting electrolyte to the water was feasible. In a field application this addition may not be feasible and the application of the here presented treatment approach limited to water with the given conductivity ranges.

## 4.9 Energy consumption

Figure 19 shows the energy consumption of the process under laboratory scale conditions.

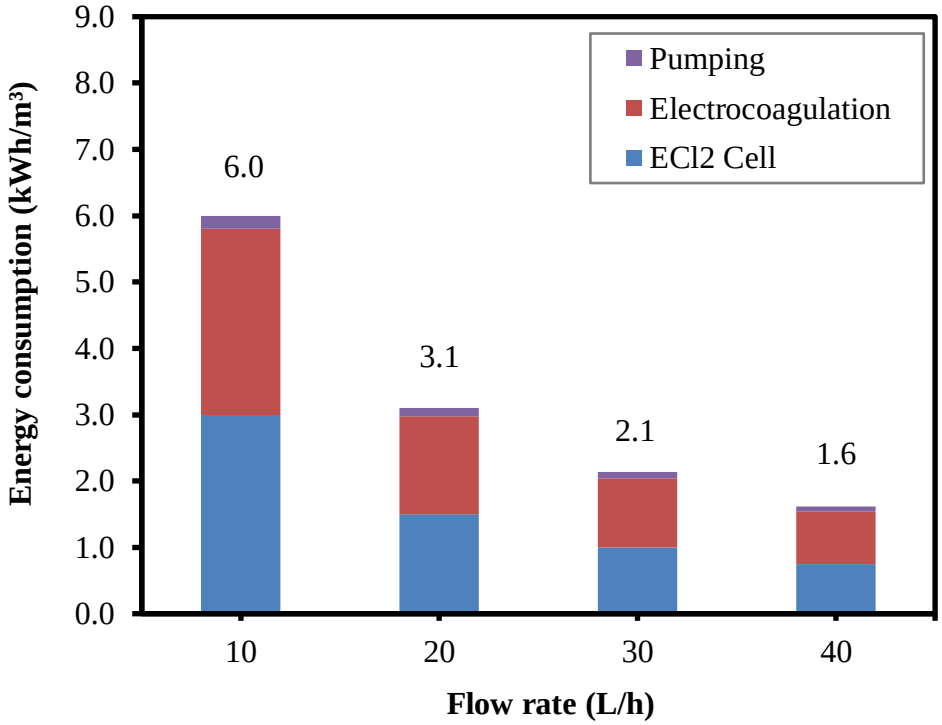


**Figure 19: Total energy consumption of treatment process under lab scale conditions**

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 compared to alternative treatment technologies such as nano-filtration (~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 regards to the alignment between the initial fluoride concentration and current applied to the EC-cell. Emamjomeh and Sivakumar (2009) suggested to limiting the current density in electrocoagulation treatment system purposely to avoid excessive energy consumption when the target fluoride concentration range was successful reached.

# CHAPTER FIVE

# CONCLUSION AND RECOMMENDATIONS

## 5.1 Conclusion

Fluoride and pathogens in contaminated water can effectively be consecutively removed by using the two separate processes (electrocoagulation and electro-disinfection) 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, respectively. Under the optimized conditions, the concentration of fluoride and pathogens in treated water were found to be 1.44 mg/L (% removal: 88.29%) and 0 CFU/100 mL (> 4 log units), respectively, below the limits set by WHO and TBS of 1.5 mg/L and 0 CFU/100 mL. The EC-$ECl_2$ system is a promising solution for 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 an 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 electrolyte to the water was feasible. In a field application this addition may not be feasible and the application of the here presented treatment approach limited to water with the given conductivity ranges.

## 5.2 Recommendations

The water supply is purposeful for drinking, the 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 (DOM) in water is recommended for future studies as well as the influence of the seasonal variations on 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 minutes as reported in the results. Testing the system for defluoridation at higher fluoride concentrations

and estimation of 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 a NaCl brine, hereby reducing the energy consumption of chlorine generation substantially.

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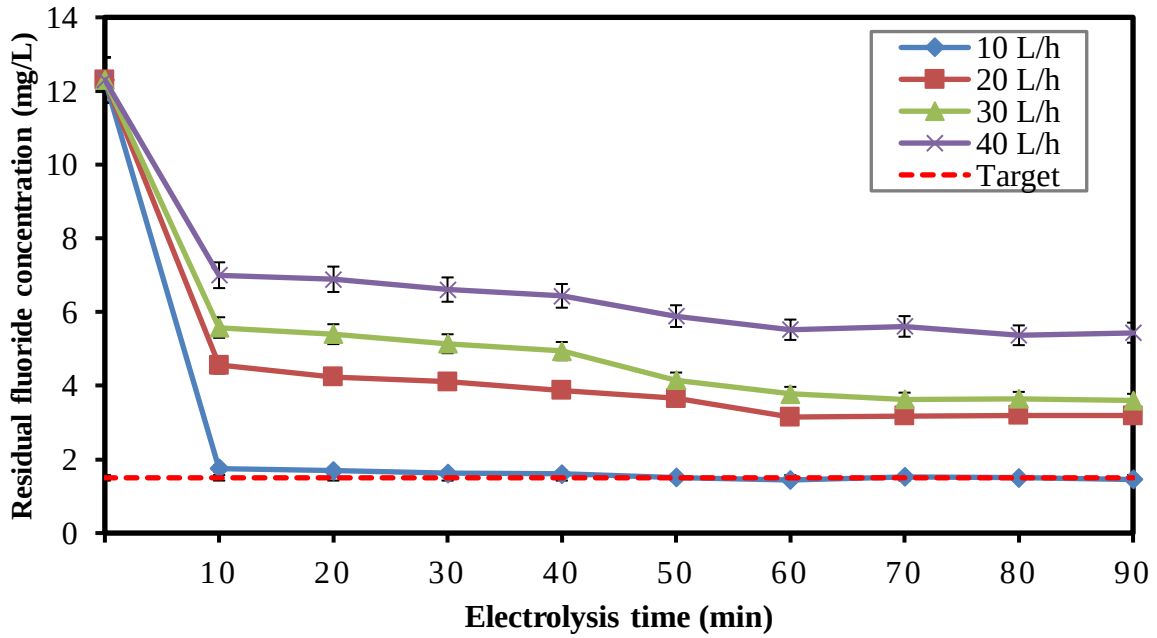
## APPENDICES

**Appendix 1: Effect of flow rates on residual $F^-$ concentrations at constant current density (9.4 mA/cm$^2$), pH (6.8), initial $F^-$ concentration (12.3 mg/L) and varied electrolysis time (10-90 min)**

**(a)**

| Electrolysis time (min) | Fluoride (mg/L): $SP_1$ | Flow rate (L/h): pH = 6.8, 9.4 mA/cm$^2$ $SP_2$ | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | 10 L/h | % Removal | 20 L/h | % Removal | 30 L/h | % Removal | 40 L/h | % Removal |
| 10 | 12.3 | 1.75 | 85.77 | 4.56 | 62.93 | 5.58 | 54.63 | 7.00 | 43.09 |
| 20 | 12.3 | 1.69 | 86.26 | 4.24 | 65.53 | 5.40 | 56.10 | 6.89 | 43.98 |
| 30 | 12.3 | 1.62 | 86.83 | 4.11 | 66.59 | 5.14 | 58.21 | 6.61 | 46.26 |
| 40 | 12.3 | 1.61 | 86.91 | 3.87 | 68.54 | 4.94 | 59.84 | 6.44 | 47.64 |
| 50 | 12.3 | 1.51 | 87.72 | 3.66 | 70.24 | 4.15 | 66.26 | 5.89 | 52.11 |
| 60 | 12.3 | 1.44 | 88.29 | 3.15 | 74.39 | 3.78 | 69.27 | 5.52 | 55.12 |
| 70 | 12.3 | 1.53 | 87.56 | 3.18 | 74.15 | 3.63 | 70.49 | 5.61 | 54.39 |
| 80 | 12.3 | 1.50 | 87.80 | 3.20 | 73.98 | 3.65 | 70.33 | 5.37 | 56.34 |
| 90 | 12.3 | 1.46 | 88.13 | 3.19 | 74.07 | 3.60 | 70.73 | 5.44 | 55.77 |

**(b)**

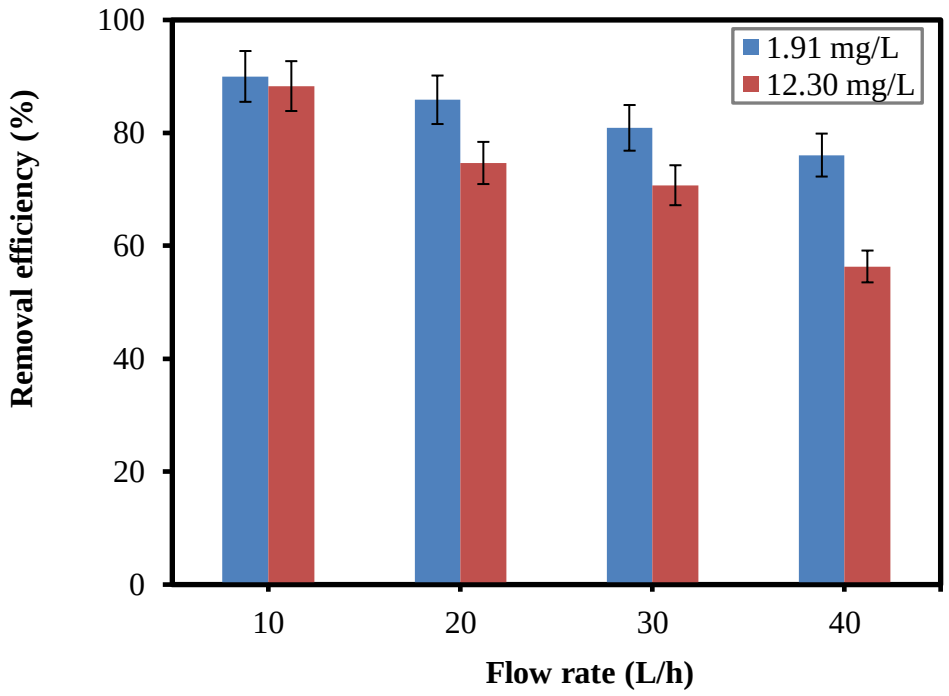


**Appendix 2: Effect of initial fluoride concentrations (mg/L) on defluoridation efficiency after EC cell at constant pH (6.8), current density (9.4 mA/cm$^2$), and varied flow rates (10-40 L/h)**

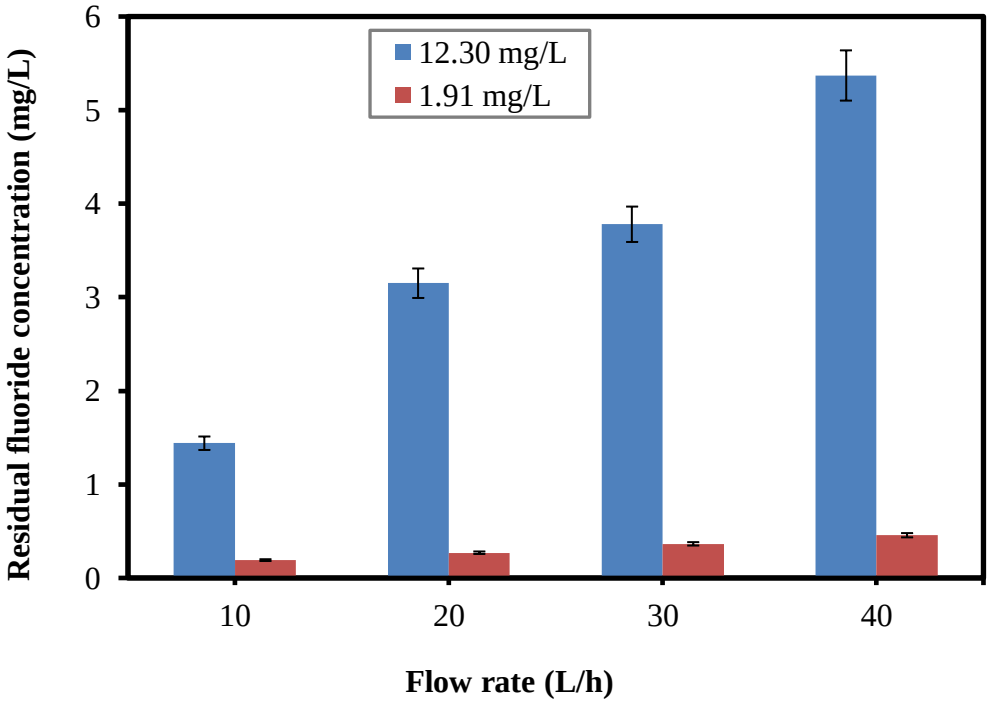
**(a)**

| Electrolysis time (min) | $SP_2$: 12.3 mg$F^-$/L, 9.4 mA/cm$^2$, pH = 6.8 | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | 10 L/h | % Removal | 20 L/h | % Removal | 30 L/h | % Removal | 40 L/h | % Removal |
| 10 | 1.75 | 85.77 | 4.56 | 62.93 | 5.58 | 54.63 | 7.00 | 43.09 |
| 20 | 1.69 | 86.26 | 4.24 | 65.53 | 5.40 | 56.10 | 6.89 | 43.98 |
| 30 | 1.62 | 86.83 | 4.11 | 66.59 | 5.14 | 58.21 | 6.61 | 46.26 |
| 40 | 1.61 | 86.91 | 3.87 | 68.54 | 4.94 | 59.84 | 6.44 | 47.64 |
| 50 | 1.51 | 87.72 | 3.66 | 70.24 | 4.15 | 66.26 | 5.89 | 52.11 |
| 60 | 1.44 | 88.29 | 3.15 | 74.39 | 3.78 | 69.27 | 5.52 | 55.12 |
| 70 | 1.53 | 87.56 | 3.18 | 74.15 | 3.63 | 70.49 | 5.61 | 54.39 |
| 80 | 1.50 | 87.80 | 3.20 | 73.98 | 3.65 | 70.33 | 5.37 | 56.34 |
| 90 | 1.46 | 88.13 | 3.19 | 74.07 | 3.60 | 70.73 | 5.44 | 55.77 |
| **$SP_2$: 1.91 mg$F^-$/L, 9.4 mA/cm$^2$, pH = 6.8** | | | | | | | | |
| 10 | 0.259 | 86.44 | 0.279 | 85.39 | 0.370 | 80.63 | 0.457 | 76.07 |
| 20 | 0.267 | 86.02 | 0.281 | 85.29 | 0.365 | 80.89 | 0.463 | 75.76 |
| 30 | 0.230 | 87.96 | 0.270 | 85.86 | 0.392 | 79.48 | 0.479 | 74.92 |
| 40 | 0.225 | 88.22 | 0.296 | 84.50 | 0.388 | 79.69 | 0.481 | 74.82 |
| 50 | 0.221 | 88.43 | 0.315 | 83.51 | 0.389 | 79.63 | 0.491 | 74.29 |
| 60 | 0.191 | 90.00 | 0.296 | 84.50 | 0.400 | 79.06 | 0.495 | 74.08 |
| 70 | 0.234 | 87.75 | 0.317 | 83.40 | 0.403 | 78.90 | 0.499 | 73.87 |
| 80 | 0.205 | 89.27 | 0.320 | 83.25 | 0.417 | 78.17 | 0.501 | 73.77 |
| 90 | 0.201 | 89.48 | 0.326 | 82.93 | 0.426 | 77.70 | 0.512 | 73.19 |

**(b)**


100
80
60
40
20
0
Removal efficiency (%)
1.91 mg/L
12.30 mg/L
10
20
30
40
Flow rate (L/h)

**(c)**

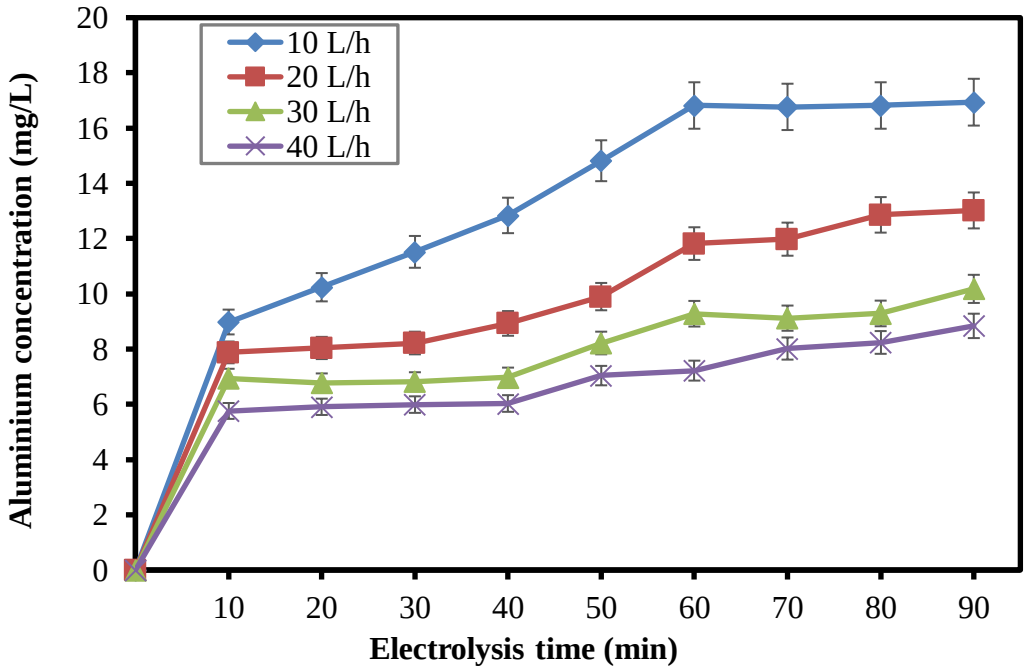

6
5
4
3
2
1
0
Residual fluoride concentration (mg/L)
12.30 mg/L
1.91 mg/L
10
20
30
40
Flow rate (L/h)

**Appendix 3: Effect of flow rates (L/h) on production and residual aluminum concentrations (mg/L) at constant current density (9.4 mA/cm$^2$), pH (6.8) and initial fluoride concentration (12.3 mg/L)**

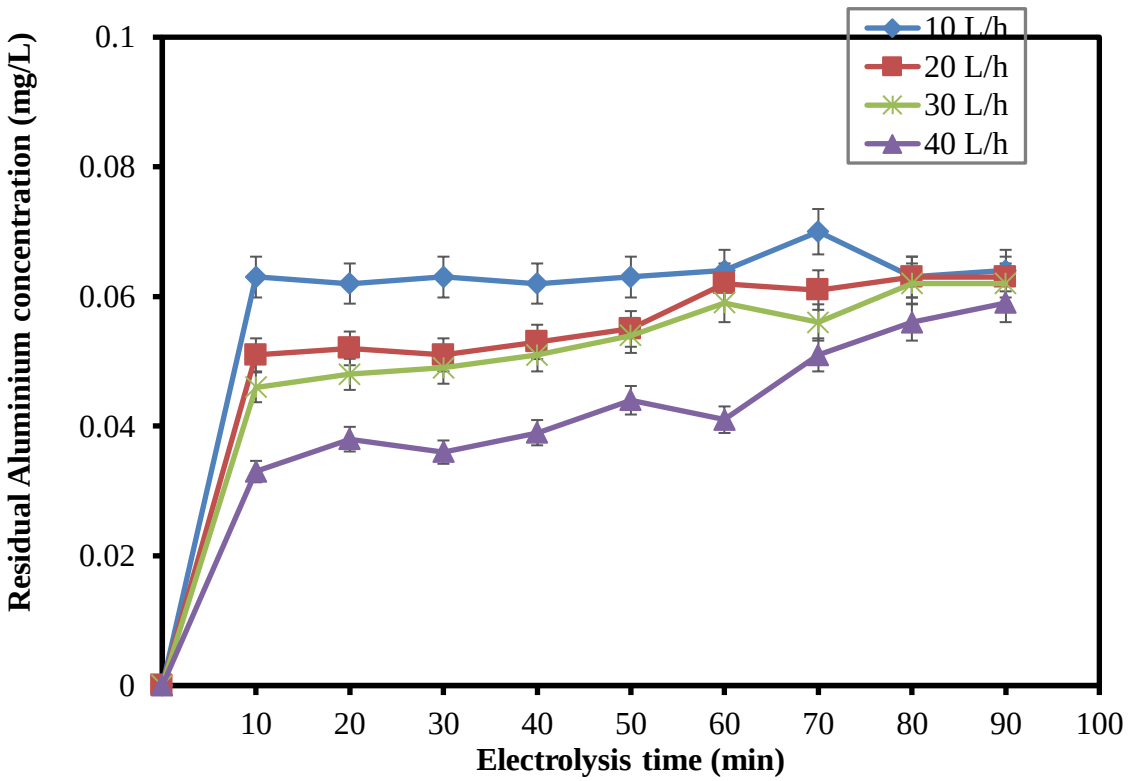
**(a)**

| Electrolysis time (min) | Aluminum concentration: 12.3 mgF$^-$/L, pH = 6.8 and 9.4 mA/cm$^2$ | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | $SP_2$ – $Al^{3+}$ after EC cell | | | | $SP_3$ – Residual $Al^{3+}$ after filtration step | | | |
| | 10 L/h | 20 L/h | 30 L/h | 40 L/h | 10 L/h | 20 L/h | 30 L/h | 40 L/h |
| 10 | 8.98 | 7.88 | 6.94 | 5.76 | 0.063 | 0.051 | 0.046 | 0.033 |
| 20 | 10.24 | 8.04 | 6.78 | 5.91 | 0.062 | 0.052 | 0.048 | 0.038 |
| 30 | 11.52 | 8.22 | 6.82 | 5.99 | 0.063 | 0.051 | 0.049 | 0.036 |
| 40 | 12.84 | 8.93 | 6.98 | 6.03 | 0.062 | 0.053 | 0.051 | 0.039 |
| 50 | 14.82 | 9.9 | 8.22 | 7.04 | 0.063 | 0.055 | 0.054 | 0.044 |
| 60 | 16.82 | 11.82 | 9.28 | 7.22 | 0.064 | 0.062 | 0.059 | 0.041 |
| 70 | 16.77 | 11.98 | 9.12 | 8.02 | 0.07 | 0.061 | 0.056 | 0.051 |
| 80 | 16.82 | 12.86 | 9.29 | 8.24 | 0.063 | 0.063 | 0.062 | 0.056 |
| 90 | 16.94 | 13.02 | 10.18 | 8.84 | 0.064 | 0.063 | 0.062 | 0.059 |

**(b)**



**(c)**

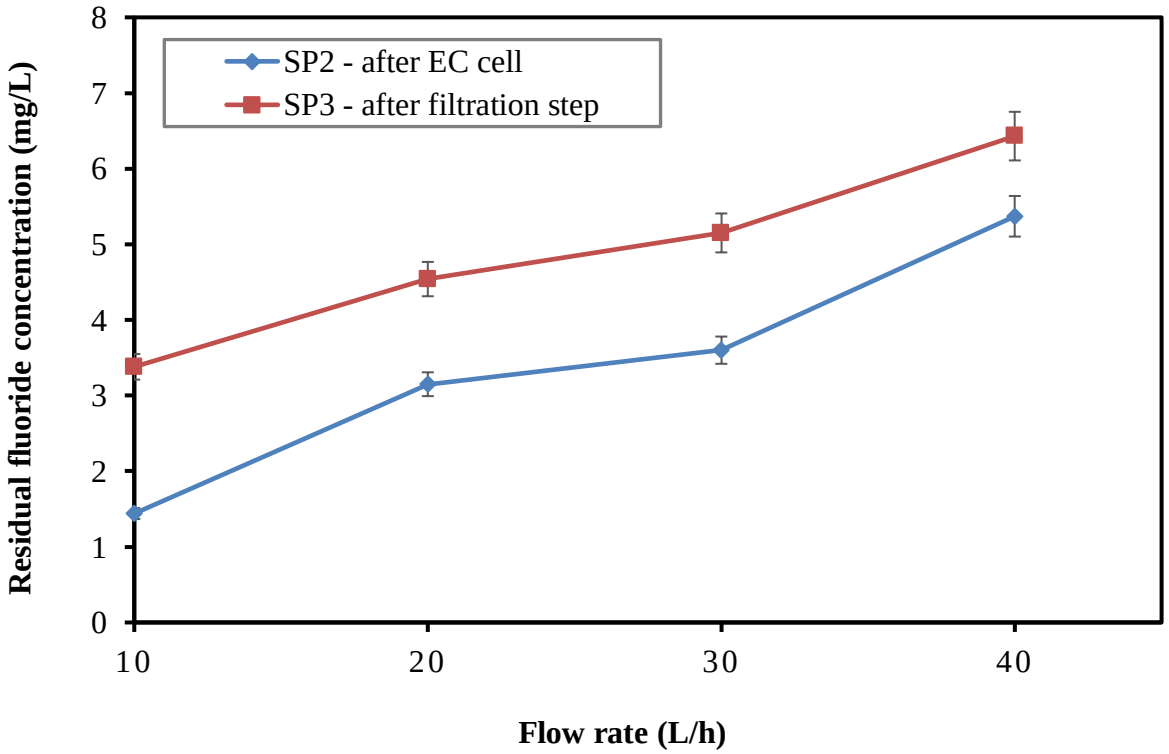

10 L/h
20 L/h
30 L/h
40 L/h
Residual Aluminium concentration (mg/L)
0.1
0.08
0.06
0.04
0.02
0
10
20
30
40
50
60
70
80
90
100
Electrolysis time (min)

**Appendix 4: Residual fluoride concentration after EC cell ($SP_2$) and filtration step ($SP_3$) at constant initial fluoride concentration (12.3 mg/L), current density (9.4 mA/cm$^2$), pH (6.8) and varied flow rates (10-40 L/h)**

**(a)**

| Fluoride at $SP_1$ = 12.3 mg/L, 9.4 mA/cm$^2$, pH = 6.8 | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| **Electrolysis Time (min)** | **$SP_2$ - after Al cell** | | | | **$SP_3$ - after filtration** | | | |
| | 10 L/h | 20 L/h | 30 L/h | 40 L/h | 10 L/h | 20L/h | 30L/h | 40L/h |
| 10 | 1.75 | 3.96 | 5.58 | 7.00 | 4.46 | 6.33 | 6.94 | 7.57 |
| 20 | 1.69 | 3.74 | 5.40 | 6.89 | 4.11 | 5.82 | 6.98 | 7.51 |
| 30 | 1.62 | 3.51 | 5.14 | 6.61 | 3.95 | 5.71 | 6.87 | 7.32 |
| 40 | 1.61 | 3.37 | 4.94 | 6.44 | 3.88 | 5.21 | 6.76 | 7.22 |
| 50 | 1.51 | 3.19 | 4.15 | 5.89 | 3.68 | 5.02 | 6.37 | 7.09 |
| 60 | 1.44 | 3.15 | 3.78 | 5.52 | 3.38 | 4.94 | 5.77 | 6.86 |
| 70 | 1.53 | 3.18 | 3.63 | 5.61 | 3.41 | 4.78 | 5.73 | 6.57 |
| 80 | 1.50 | 3.20 | 3.65 | 5.37 | 3.44 | 4.54 | 5.41 | 6.53 |
| 90 | 1.46 | 3.19 | 3.60 | 5.44 | 3.42 | 4.67 | 5.15 | 6.43 |

**(b)**

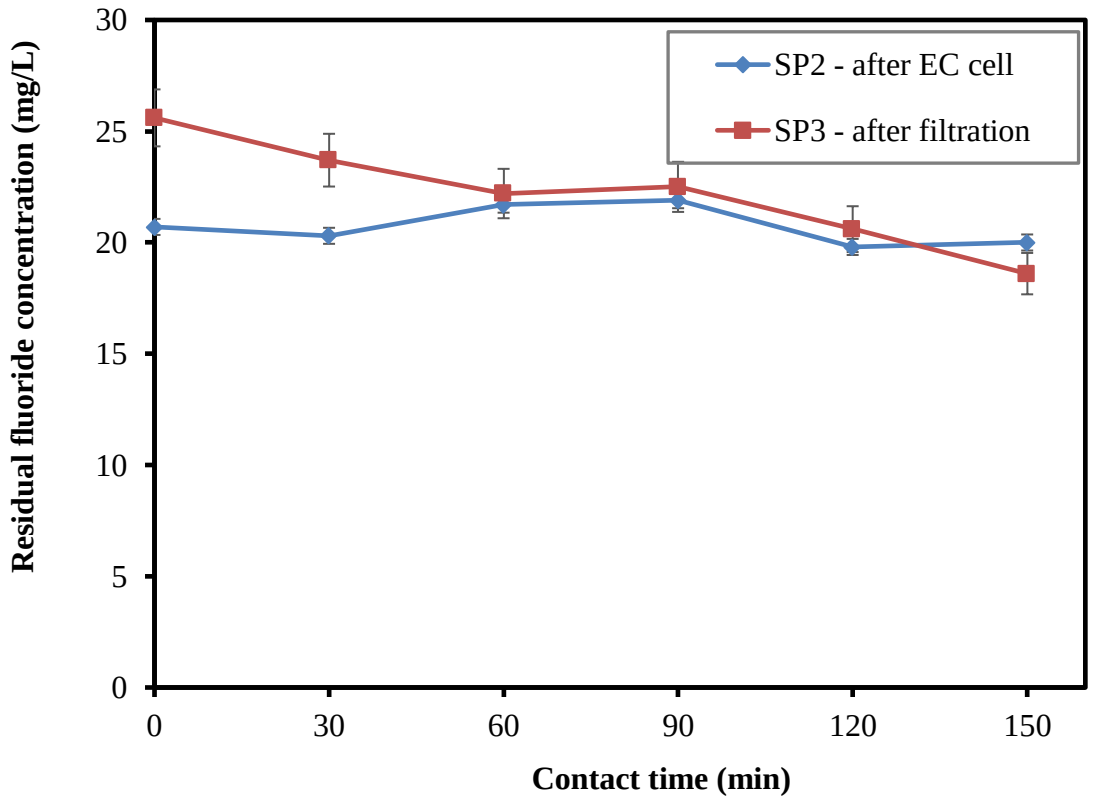


**Appendix 5: Effect of residence time on treated water before filtration step at constant initial fluoride concentration (36.5 mg/L), current density (9.4 mA/cm$^2$), pH (6.9), and flow rate (10 L/h)**

**(a)**

| **$C_o$ = 36.5 mg$F^-$/L, 9.4 mA/cm$^2$, pH = 6.9 and 10 L/h** | | |
|---|---|---|
| **Time (min)** | **$SP_2$ – unfiltered samples** | **$SP_3$ – filtered samples** |
| 0 | 20.70 | 25.60 |
| 30 | 20.30 | 23.70 |
| 60 | 21.70 | 22.20 |
| 90 | 21.90 | 22.50 |
| 120 | 19.80 | 20.60 |
| 150 | 20.00 | 18.60 |

**(b)**

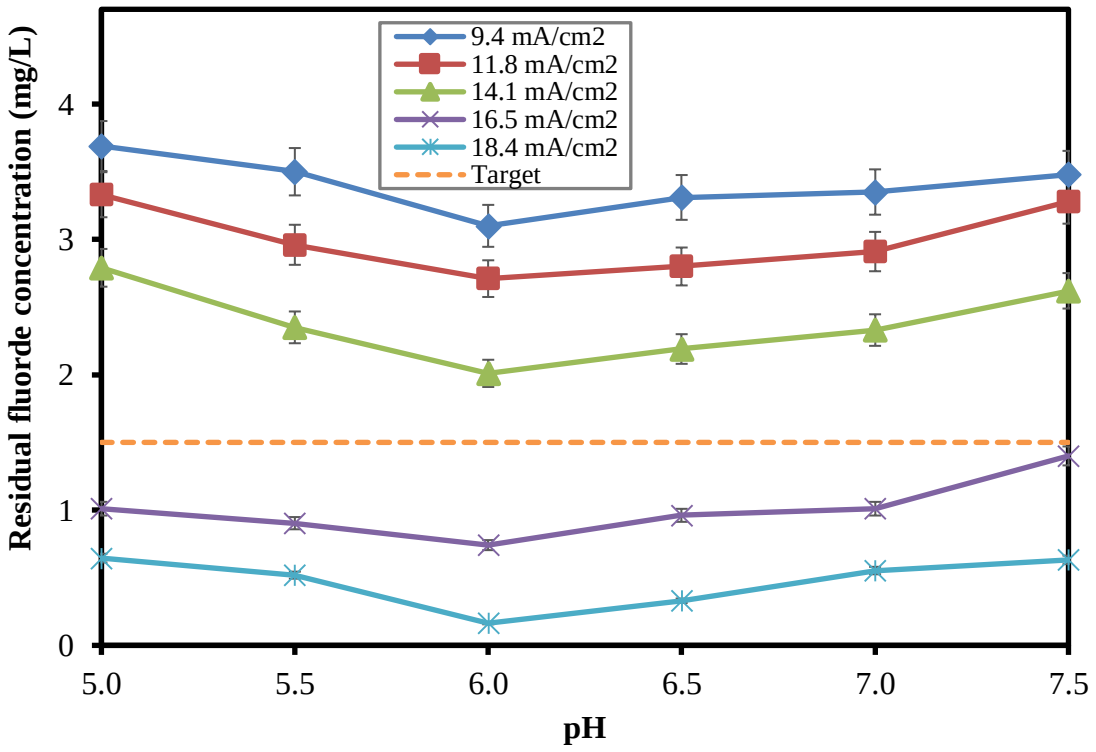


**Appendix 6: Effect of initial pH on residual fluoride concentration at constant flow rate (20 L/h), initial fluoride concentration (12.3 mg/L), and varied current density (9.4-18.4 mA/cm$^2$)**

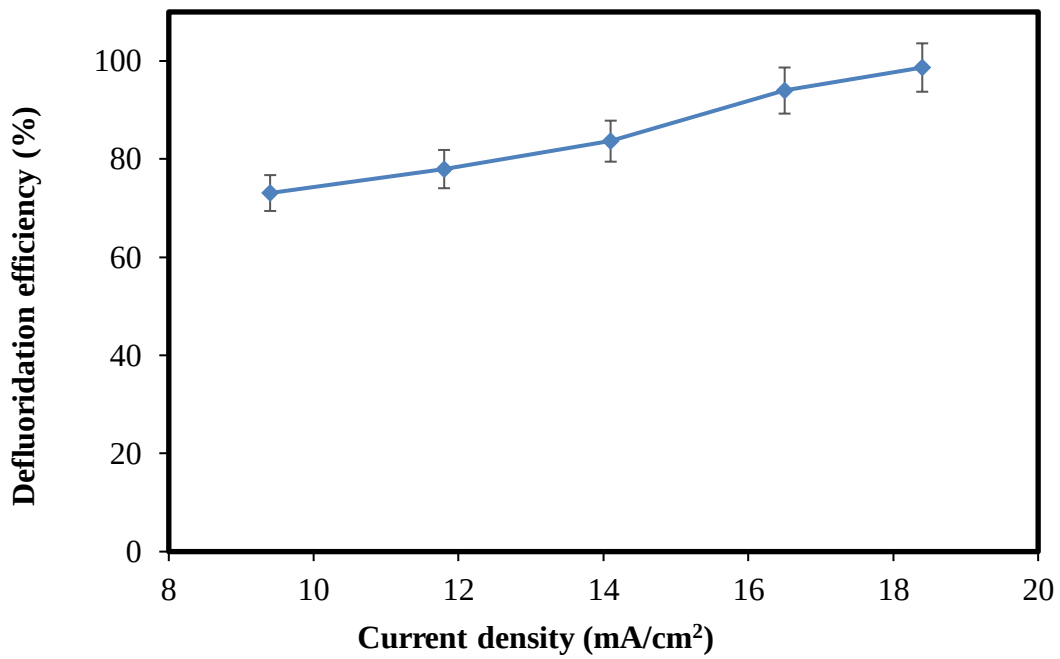
**(a)**

| pH | Electrolysis time (min) | Fluoride (mg/L): 12.3 mgF$^-$/L, 20 L/h | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | 9.4 mA/cm$^2$ | $Al^{3+}$ (mg/L) | 11.8 mA/cm$^2$ | $Al^{3+}$ (mg/L) | 14.1 mA/cm$^2$ | $Al^{3+}$ (mg/L) | 16.5 mA/cm$^2$ | $Al^{3+}$ (mg/L) | 18.4 mA/cm$^2$ | $Al^{3+}$ (mg/L) |
| 5.0 | 60 | 3.69 | 6.63 | 3.33 | 8.59 | 2.79 | 11.67 | 1.01 | 16.27 | 0.645 | 23.11 |
| 5.5 | 120 | 3.50 | 6.93 | 2.96 | 9.61 | 2.35 | 12.23 | 0.903 | 16.82 | 0.519 | 23.89 |
| 6.0 | 180 | 3.10 | 7.42 | 2.71 | 10.22 | 2.01 | 13.43 | 0.741 | 17.92 | 0.164 | 24.54 |
| 6.5 | 240 | 3.31 | 7.19 | 2.80 | 9.71 | 2.19 | 13.19 | 0.961 | 17.41 | 0.330 | 23.75 |
| 7.0 | 300 | 3.35 | 7.01 | 2.91 | 9.14 | 2.33 | 12.86 | 1.010 | 17.09 | 0.553 | 22.53 |
| 7.5 | 360 | 3.48 | 6.74 | 3.28 | 8.97 | 2.62 | 12.67 | 1.400 | 16.24 | 0.632 | 22.12 |

**(b)**



**(c)**

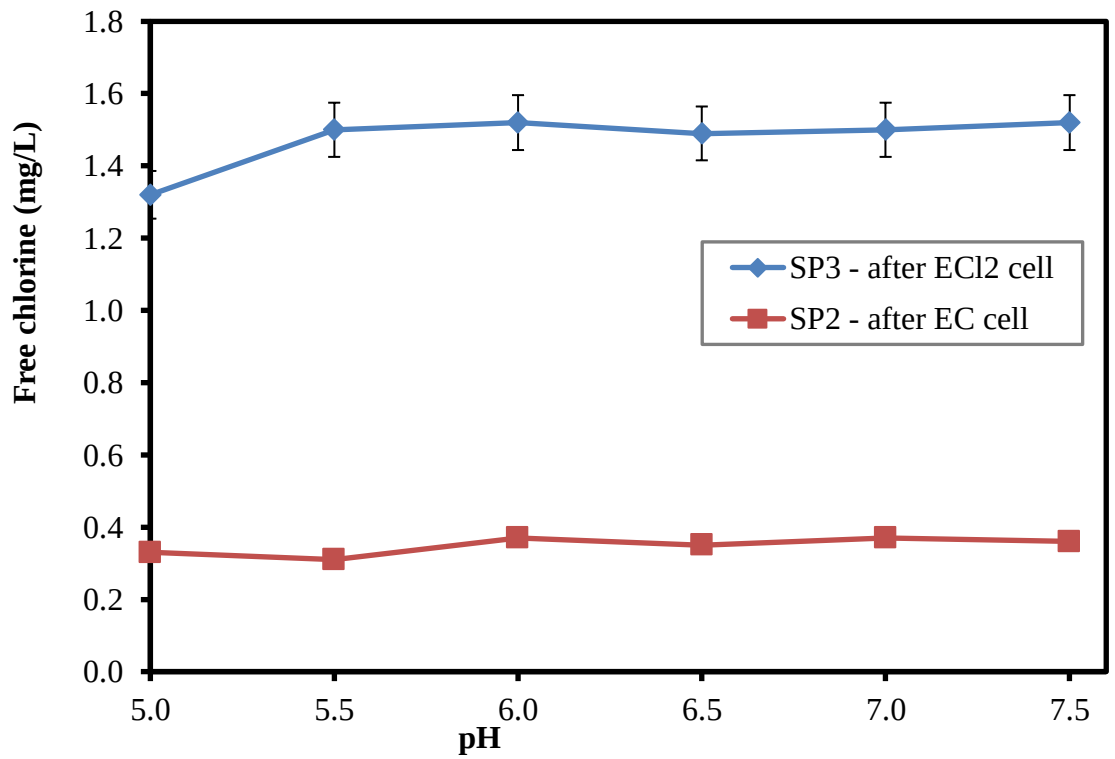

Defluoridation efficiency (%)
100
80
60
40
20
0
8
10
12
14
16
18
20
Current density (mA/cm2)

**Appendix 7: Effect of initial pH on production of free chlorine at constant flow rate (20 L/h), current (9.4 mA/cm$^2$ for EC cell and 3.1 mA/cm$^2$ for $ECl_2$ cell)**

**(a)**

| pH | Free Chlorine (mg/L): 20 L/h | |
|---|---|---|
| | $SP_2$: 9.4 mA/cm$^2$ | $SP_3$: 3.1 mA/cm$^2$ |
| 5.0 | 0.33 | 1.32 |
| 5.5 | 0.31 | 1.50 |
| 6.0 | 0.37 | 1.52 |
| 6.5 | 0.35 | 1.49 |
| 7.0 | 0.37 | 1.50 |
| 7.5 | 0.36 | 1.52 |

**(b)**

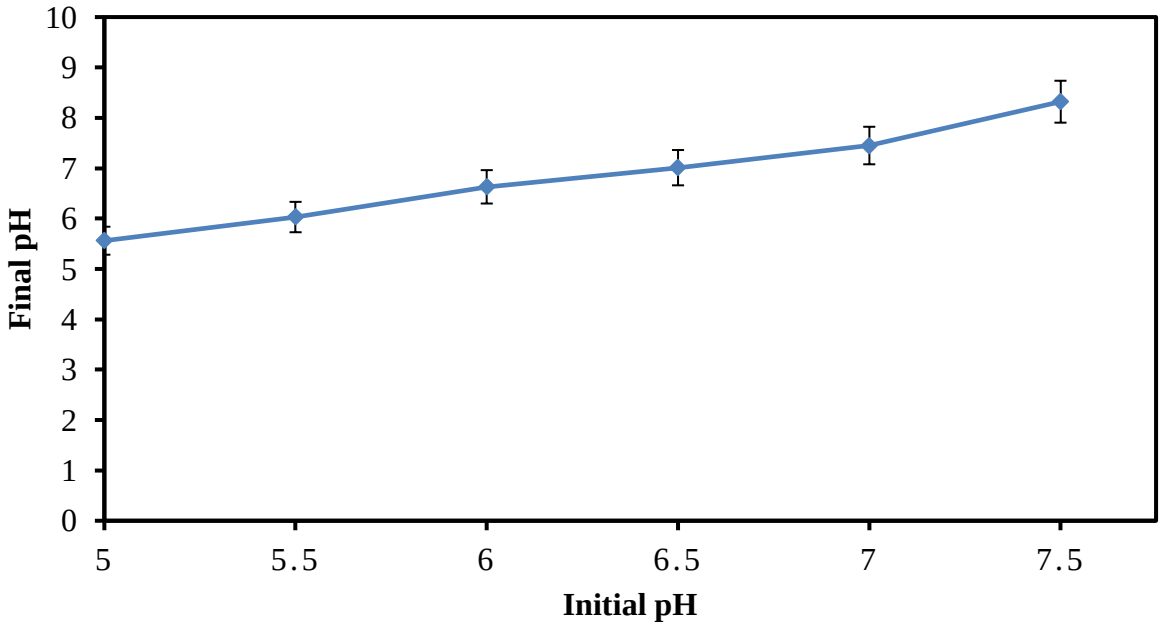


**Appendix 8: Initial pH before treatment and final pH of the treated water**

**(a)**

| 20 L/h, 9.4 mA/cm$^2$, 12.3 mgF$^-$/L | | |
|---|---|---|
| **Initial pH** | **Final pH** | **Change** |
| 5.0 | 5.56 | +0.56 |
| 5.5 | 6.03 | +0.53 |
| 6.0 | 6.63 | +0.63 |
| 6.5 | 7.01 | +0.51 |
| 7.0 | 7.45 | +0.45 |
| 7.5 | 8.32 | +0.82 |

**(b)**

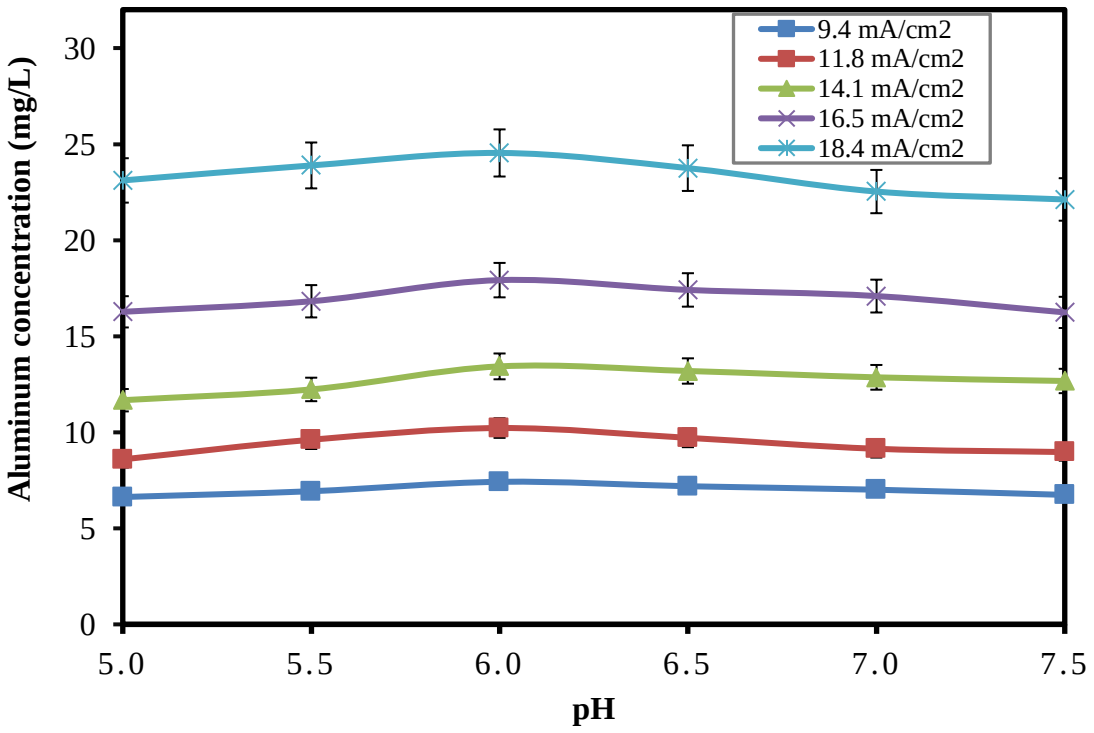


**Appendix 9:** **Effect of current density (9.4-18.4 mA/cm$^2$) on production of aluminum concentration (mg/L) at constant flow rate (20 L/h), initial fluoride concentration (12.3 mg/L) and varied pH (5.0-7.5)**

**(a)**

| pH | Aluminum concentration (mg/L) at $SP_2$; 20 L/h; 12.3 mg$F^-$/L | | | | |
|---|---|---|---|---|---|
| | 9.4 mA/cm$^2$ | 11.8 mA/cm$^2$ | 14.1 mA/cm$^2$ | 16.5 mA/cm$^2$ | 18.4 mA/cm$^2$ |
| 5.0 | 6.63 | 8.59 | 11.67 | 16.27 | 23.11 |
| 5.5 | 6.93 | 9.61 | 12.23 | 16.82 | 23.89 |
| 6.0 | 7.42 | 10.22 | 13.43 | 17.92 | 24.54 |
| 6.5 | 7.19 | 9.71 | 13.19 | 17.41 | 23.75 |
| 7.0 | 7.01 | 9.14 | 12.86 | 17.09 | 22.53 |
| 7.5 | 6.74 | 8.97 | 12.67 | 16.24 | 22.12 |

**(b)**

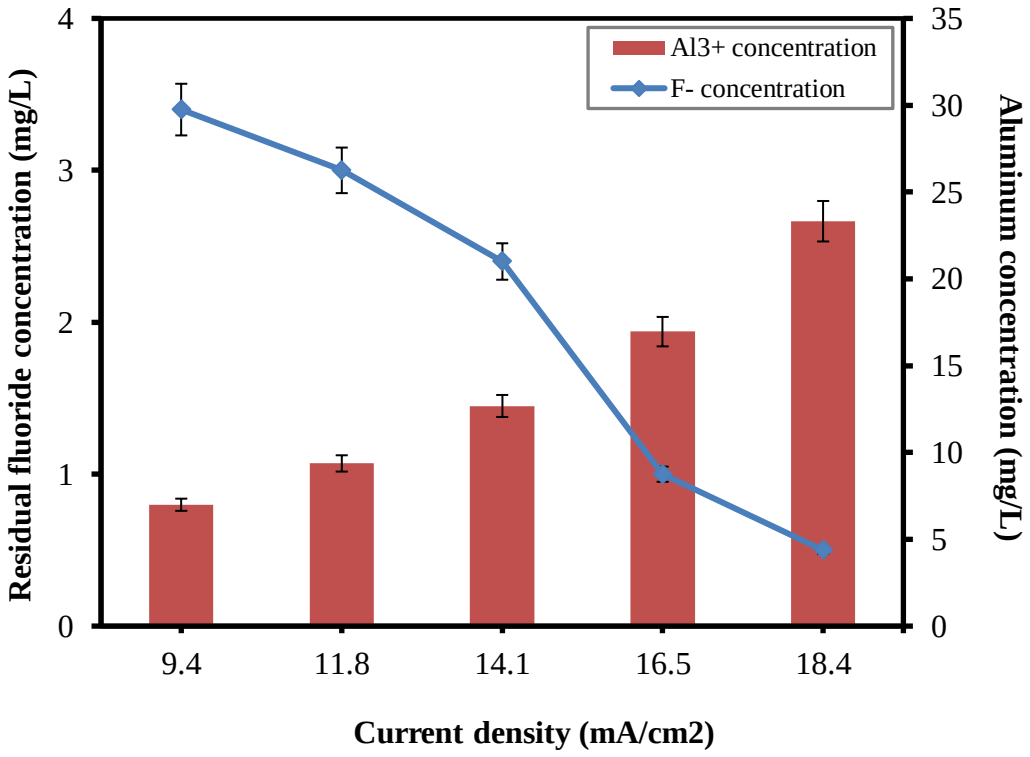



.

**(c)**

| Initial fluoride (mg/L) = 12.30 mg/L, flow rate = 20 L/h, pH = 6.25 ± 0.56 | | |
|---|---|---|
| Current density (mA/cm$^2$) | Aluminum concentration (mg/L) | Residual fluoride concentration (mg/L) |
| 9.4 | 6.99 ± 0.26 | 3.4 ± 0.20 |
| 11.8 | 9.37 ± 0.53 | 3.00 ± 0.23 |
| 14.1 | 12.68 ± 0.59 | 2.38 ± 0.26 |
| 16.5 | 16.96 ± 0.60 | 1.00 ± 0.20 |
| 18.4 | 23.32 ± 0.83 | 0.47 ± 0.17 |

**(d)**

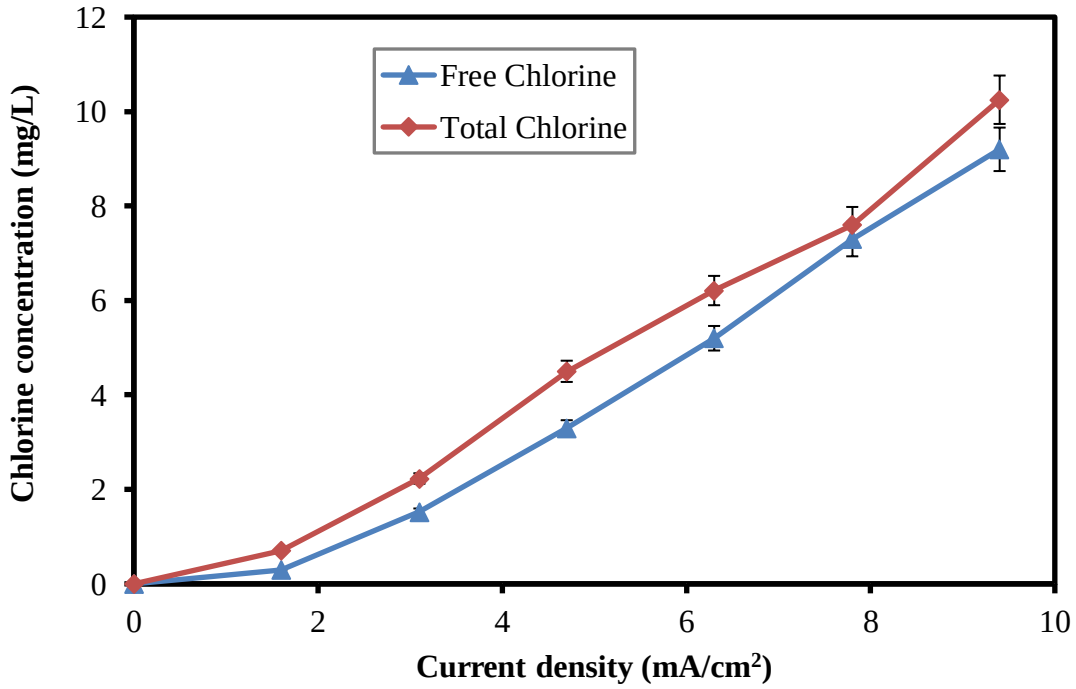


**Appendix 10: Effect of current density on disinfectants production (b) and disinfection (c) at constant pH (6.0) and flow rate (20 L/h)**

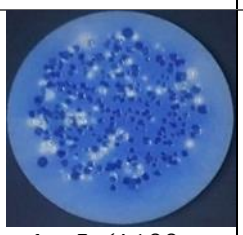
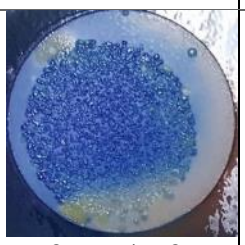
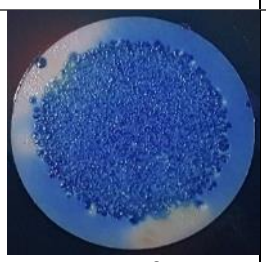
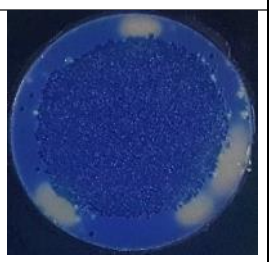
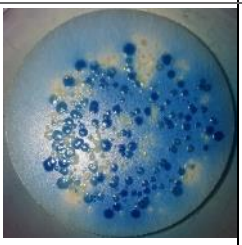
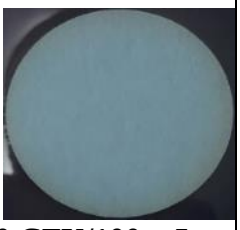
**(a)**

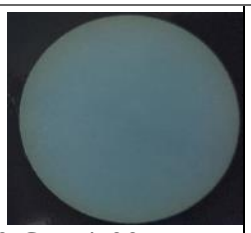
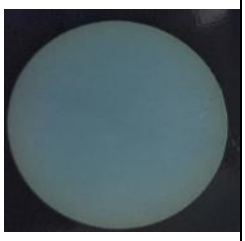
| Free and Total Chlorine after $ECl_2$ Cell | | | | Fecal Coliforms (CFU/100 mL) | | |
|---|---|---|---|---|---|---|
| **Current (mA)** | **Current density ($mA/cm^2$)** | **Free Chlorine (mg/L)** | **Total Chlorine (mg/L)** | **Before dosing - raw water** | **After dosing – 0.5 L domestic wastewater** | **After dosing – 100 L domestic wastewater** |
| 0 | 0 | 0.0 | 0.0 | 8 | 36 | 19700 |
| 1000 | 1.6 | 0.3 | 0.7 | 0 | 0 | 134 |
| 2000 | 3.1 | 1.5 | 2.2 | 0 | 0 | 0 |
| 3000 | 4.7 | 3.3 | 4.5 | 0 | 0 | 0 |
| 4000 | 6.3 | 5.5 | 6.2 | 0 | 0 | 0 |
| 5000 | 7.8 | 7.3 | 7.6 | 0 | 0 | 0 |
| 6000 | 9.4 | 9.2 | 10.3 | 0 | 0 | 0 |

**(b)**



**(c)**

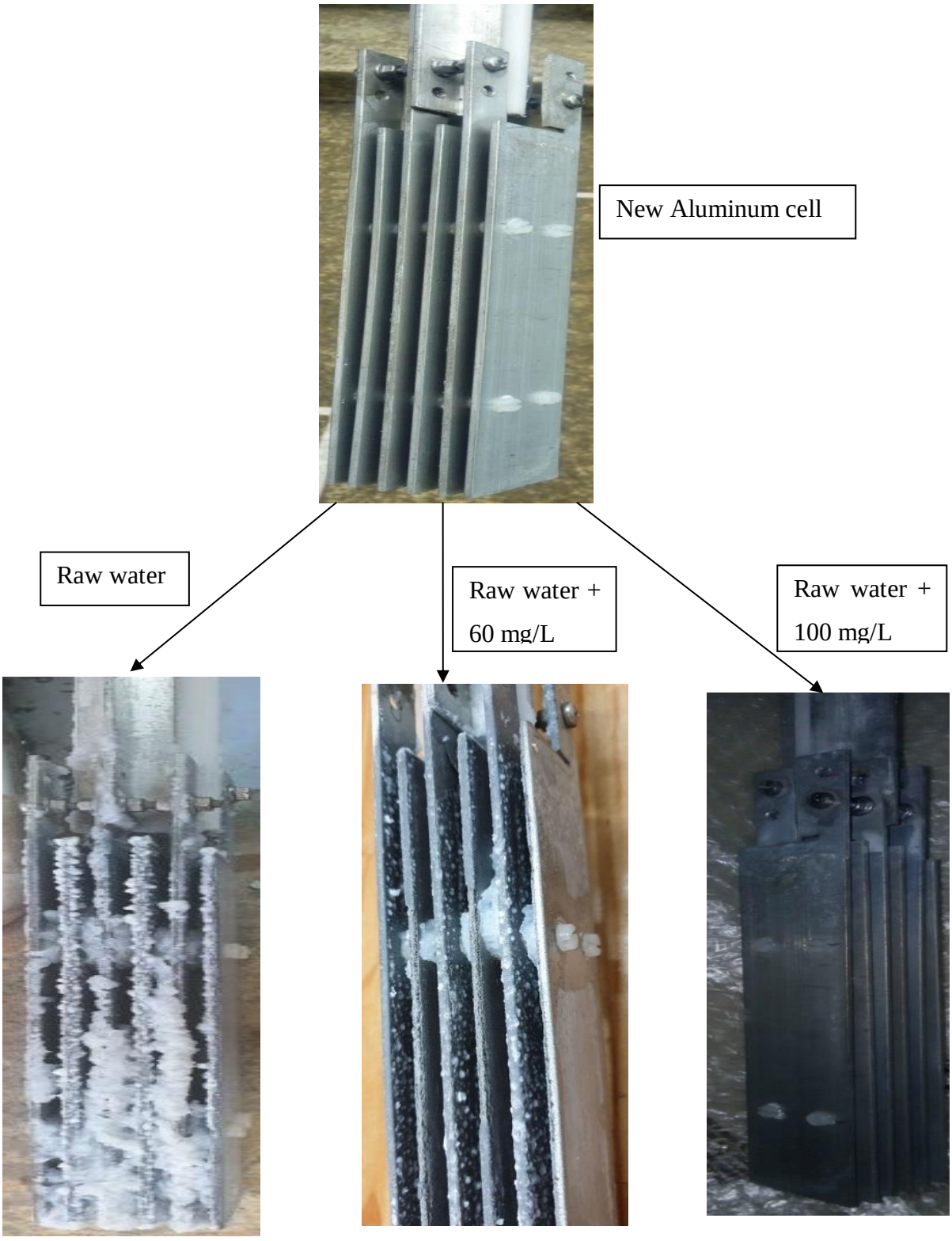
| **Current density ($mA/cm^2$)** | **Fecal coliforms (CFU/100 mL): 20 L/h, pH = 6** | | | |
|---|---|---|---|---|
| 0 | Blank | | | |
| |  **1** mL (*100 factor) 197CFU*100 = **19 700 CFU** |  **10** mL (*10 factor) |  **20** mL (*5 factor) |  **100** mL |
| 1.6 |  **134 CFU/100 mL** | | | |
| 3.1 | **0 CFU/100 mL** | | | |
| 4.7 |  **0 CFU/100 mL** | | | |

| | | | | |
|---|---|---|---|---|
| 6.3 |  **0 CFU/100 mL** | | | |
| 7.8 | **0 CFU/100 mL** | | | |
| 9.4 |  **0 CFU/100 mL** | | | |

**(d)**

| **EC current: electrode surface area = 425 $cm^2$** | | **$ECl_2$ current: electrode surface area = 640 $cm^2$** | |
|---|---|---|---|
| **Current (mA)** | **Current density ($mA/cm^2$)** | **Current (mA)** | **Current density ($mA/cm^2$)** |
| 4 000 | 9.4 | 1 000 | 1.6 |
| 5 000 | 11.8 | 2 000 | 3.1 |
| 6 000 | 14.1 | 3 000 | 4.7 |
| 7 000 | 16.5 | 4 000 | 6.3 |
| 7 800 | 18.4 | 5 000 | 7.8 |
| | | 6 000 | 9.4 |

**Appendix 11: Effect of supporting electrolyte on anodic passivation of aluminium electrode**

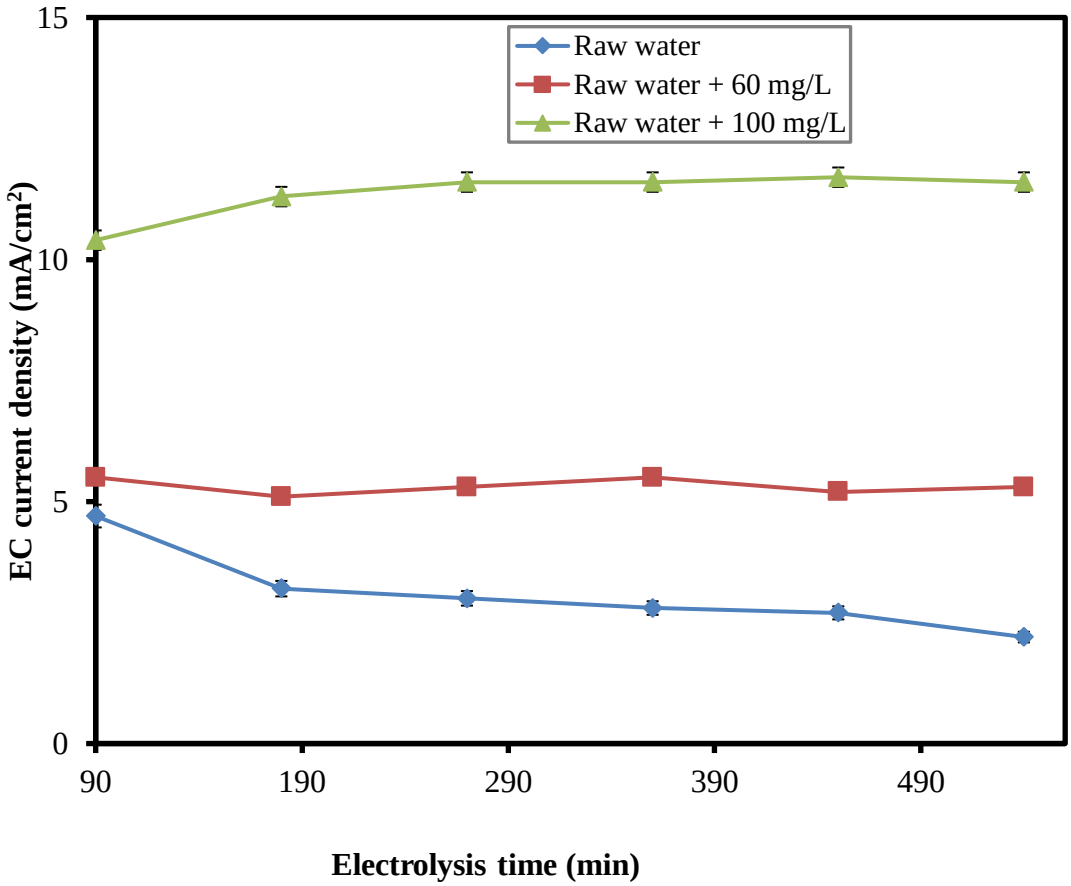


**Appendix 12:** **Effect of supporting electrolyte on current density passed through EC cell at pH 6.8**

**(a)**

| Electrolysis time (min) | EC current density (mA/cm$^2$) | | |
|---|---|---|---|
| | Raw water | Raw water + 60 mg/L | Raw water + 100 mg/L |
| 90 | 4.7 | 5.5 | 10.4 |
| 180 | 3.2 | 5.1 | 11.3 |
| 270 | 3.0 | 5.3 | 11.6 |
| 360 | 2.8 | 5.5 | 11.6 |
| 450 | 2.7 | 5.2 | 11.7 |
| 540 | 2.2 | 5.3 | 11.6 |

**(b)**



**Appendix 13: Summary of evaluated parameters for defluoridation and disinfection efficiency using EC-$ECl_2$ process in water with up to 12.30 mg$F^-$/L and 19700 CFU/100 mL**

| **Optimal operating conditions** | | | |
|---|---|---|---|
| Evaluated Factors and Conditions | Cathode-Anode/ Connection type | Optimum | Efficiency (%) |
| **Defluoridation** | | | |
| Flow rate (10-40 L/h) | Al – Al Monopolar | 10-20 L/h | 88.29-98.67% |
| Electrolysis time (10-90 min) | | 50-60 min | |
| Initial F concentration (1.91–12.3 mg/L) | | 12.3 mg/L | |
| NaCl concentration (60–100 mg/L) | | 100 mg/L | |
| Initial pH (5.0–7.5) | | 6.0-7.0 | |
| Applied current (9.4–18.4 mA/$cm^2$) | | 9.4-18.4 mA/$cm^2$ | |
| Aluminium concentration (5.76-24.94 mg/L) | | 16-24 mg/L | |
| **Disinfection** | | | |
| Free chlorine (0.3-9.2 mg/L) | Ti – Ti Monopolar | 0.3-1.5 mg/L | 99.99% |
| Applied current (1.6-9.4 mA/$cm^2$) | | 3.1 mA/$cm^2$ | |

## RESEARCH OUTPUT

### (i) Publications

Njau, O. E., Otter, P., Machunda, R., Rugaika, A., Wydra, K., & Njau, K. N. (2023). Removal of fluoride and pathogens from water using the combined electrocoagulation-inline-electrolytic disinfection process. *Water Supply*, *23*(7), 2745-2757.

### (ii) Poster presentation