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The Treatment of Contaminated Water Using Electrocoagulation

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ABSTRACT

THE TREATMENT OF CONTAMINATED WATER USING ELECTROCOAGULATION

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Wastewater treatment is a process that is used to remove significant amounts of contaminant concentration from wastewater, which prevents harm to the environment. Chemical coagulation (CC) is one of the most influential processes used to remove contaminants, and it is the addition of chemicals called coagulants that cause particles to stick together, forming bigger flocs. However, as effective as CC is, it requires bench scale testing for determining the appropriate dosing rate for specific raw water properties, which makes the process time consuming. To eliminate these issues, the introduction of the electrocoagulation (EC) process is considered. The EC process is an emerging technology in wastewater treatment as it combines the applications of coagulation, electrochemistry, and flotation. The coagulants are generated by the electrolytic oxidation of an appropriate anode material (sacrificial anode). The electrogenerated coagulant will undergo hydrolysis reactions to form various monomeric and polymeric species, part of which generate hydroxide flocs. Larger surface area flocs are beneficial in EC for rapid adsorption of pollutants. Although the EC process has existed for many years, more time has been devoted in the past decade because of its high-efficiency in treating pollutants that are not easily removed. Currently, multiple mathematical models have been developed using EC process to determine the removal rates of contaminants. However, more research needs to be conducted to fully understand the effects and optimum operating conditions of EC to remove any contaminants. The introduction

of mass transfer of ionic species is crucial in modeling of the EC process because it considers the effect of convection, electromigration, and diffusion, which are influential factors in EC. By controlling key design parameters such as current density, pH, and pollutant concentration using mathematical modeling and considering effect of mass transfer, the optimum operating conditions for a high-efficiency EC process can be determined.

NORTHERN ILLINOIS UNIVERSITY
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THE TREATMENT OF CONTAMINATED WATER USING ELECTROCOAGULATION

BY

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DEDICATION

To my siblings, Mayowa, Remi, and Gbenga Adejinle

TABLE OF CONTENTS

LIST OF FIGURES

Figure Page

LIST OF TABLES

CHAPTER 1: INTRODUCTION

1.1 Wastewater Treatment

Wastewater treatment is a process that is used to remove significant amounts of contaminant concentration from wastewater. Depending on the application used to treat the water, the effluent water can be reused. The treatment of contaminated water is very crucial to the health of the environment. Most of the time it is industrial-based wastewater that requires more treatment options in order to prevent contamination to the municipal wastewater. There are three different treatment methods for a complete wastewater treatment: primary treatment, secondary treatment, and tertiary treatment.

Primary treatment involves the sedimentation or flotation of solid waste within water. After the pretreatment stage of filtering out larger solids, wastewater is passed through several filters that separate water from contaminants. The resulting effluent goes through a chemical process such as coagulation that separates generated smaller solid particles from the liquid by turning them into larger flocs. Secondary treatment involves the degradation of biological content in wastewater. There are three ways to carry out this treatment: biofiltration—use of sand filters, contact filters, or trickling filters to remove waste; aeration—use of oxygen to allow aerobic biodegradation; and oxidation ponds—lagoon ponds with the interaction of sunlight, bacteria, and algae. In an industrial setting the treatment process is usually completed at the secondary stage because it allows for safer release to municipalities. However, in municipalities, the tertiary treatment is utilized because it requires advanced removal of wastewater contents such as nutrient removal and pathogens, which ensures that water is safe for local use and drinking purposes.

Coagulation is a chemical process that requires the addition of chemicals to react with the desired contaminant and remove it. Other than the addition of chemicals to form larger particles and remove contaminants, there are other relatively new technologies that utilize the concept of electrochemistry, such as electrocoagulation (EC). Electrocoagulation is an emerging technology that utilizes the application of coagulation, flotation, and electrochemistry. Using electricity for water treatment dates back to the 19th century, when EC was used for treating wastewater for drinking purposes in the United States, but it was found impractical due to the high capital and electricity cost required (Guohua, 2004). The relevance of EC is now being noticed over the past few years as an environmentally friendly option that requires no chemical additives, generates minimal sludge, and needs minimal footprint. This paper will be focused on the mathematical modeling of EC by determining the optimum operating conditions that generate a high-efficiency EC process. The detailed theory of EC will be discussed in another section, but it is important to first understand the theory of coagulation.

1.2 Coagulation

Coagulation is a process for combining smaller particles into larger aggregates (flocs), which can be removed in a solid/liquid separation process. The chemicals used are called coagulants. Common coagulants include alum, polyaluminum chloride, ferric chloride and various cationic or anionic polymers. Aluminum and iron coagulants put a large amount of positively charged ions into the water, which form positively charged flocs that attract the negatively charged particles of color and turbidity. As the particles collide in the mixing area, they stick together and form increasingly larger particles. The modern use of coagulants for water treatment began over 100 years ago with the use of ferric chloride and aluminum sulphate in full-scale treatment (Jiang, 2015). As shown in Figure 1 below, the coagulant is added to the impurities, which forms precipitates. The precipitates and trapped impurities settle to the bottom and are separated from the liquid by decantation and filtration or, depending on the treatment application, are carried to the surface by air bubbles to be skimmed off by a mechanical device. The reactions of alum and ferric salts in water are governed by operating conditions such as mixing intensity, pH, temperature, alkalinity, and the nature and amount of turbidity (Pizzi & Lauer, 2013).

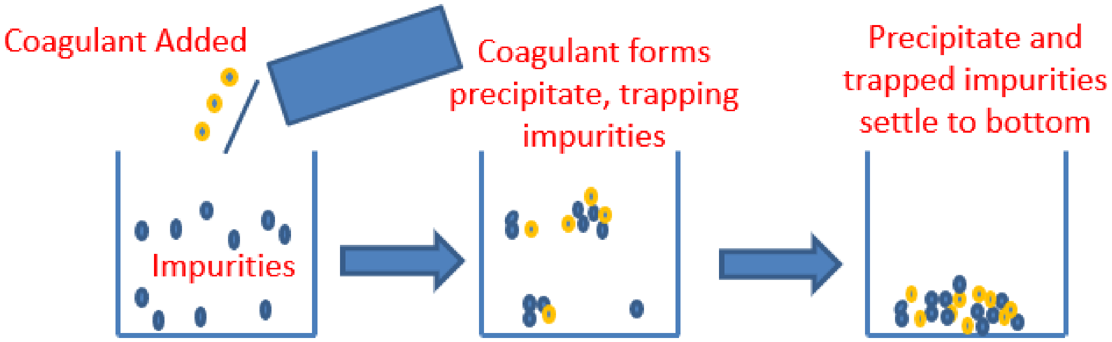

Figure 1. Description of the coagulation process.

Coagulation is an essential process for removal of suspended solids and colloidal particles from wastewater; however, there is no universally accepted mathematical theory describing all the relevant parameters that can be used to optimize and improve the process. Process optimization is based on bench scale testing (jar test) and dosing concepts, which depend on the quality of the water or wastewater.

The application of coagulation helps reduce the time required to settle out suspended solids as opposed to allowing the solids to decant for a long period of time. Coagulation is very effective in removing fine particles that cannot be removed through physical screening. It also reduces the organic matter which contributes to the biochemical oxygen demand (BOD) and chemical oxygen demand (COD). However, the determination of the optimum dosing of the coagulant is difficult, depending on water properties and operating flow conditions. It will require frequent monitoring depending on if the process utilizes a continuous or batch system. Also, coagulation requires bench scale testing to be able to predict the large-scale operating conditions. As effective as the coagulation process is, it requires a lot of attention to operate, and the replenishment of chemicals is necessary, which is why the concept of electrocoagulation is being considered as an alternative.

1.3 Electrocoagulation

Electrocoagulation (EC) is an emerging technology that is used as an alternative of chemical coagulation to treat wastewater. EC is the process of destabilizing suspended or soluble contaminants in wastewater by applying an electric current into the medium. It combines the application of coagulation, flotation, and electrochemistry. Although EC is very similar to coagulation, the process is carried out without the addition of chemicals, where the coagulating agent is generated through the electro-oxidation of a sacrificial anode such as aluminum or iron. A simple and basic EC unit consists of an electrolytic cell with anode and cathode metal electrodes connected to a DC power source and immersed in a solution to be treated as shown in

Figure 2 below. The electrodes used may be of the same or different material. Once charged, the particles bond together and form flocs, which can be removed through a physical process such as filtration or a mechanical skimming device.

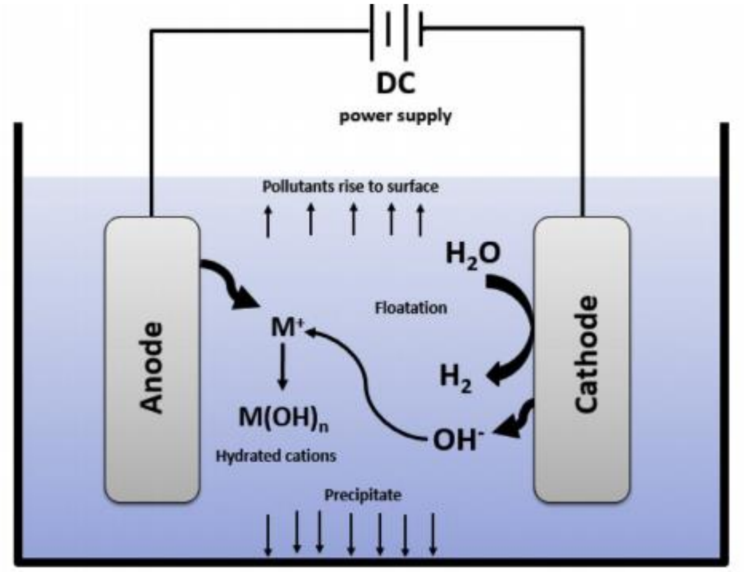

Figure 2. Schematic representation of a basic EC cell.

Iron and aluminum electrodes are the widely used metals for EC cells since these metals are available, non-toxic, and proven to be reliable (Moussa, 2017). The anode serves as the coagulant and is referred to as the sacrificial anode because it dissociates to give metal cations when electric current is applied in the cell. The anode generated from the electrochemical reaction reacts with water (H_2O) in a chemical reaction to form various metal hydroxides and hydrogen gas bubbles. The final-form of the metal hydroxides generated reacts with the pollutant, and the hydrogen gas bubbles generated carry the flocs formed to the surface in an adsorption process. The rate of generation of flocs can be controlled by the applied electric current, which produces a minimized amount of sludge generation, resulting in a lucrative technology for water or wastewater treatment (Balasubramanian, Toshinori, Ahmed, & Srinivasakannan, 2009).

In EC, since no chemicals are added, there is no presence of secondary pollution from high concentration of chemical mixing (Holt, Barton, & Mitchell, 2005). When the electric current is applied, the evolution of hydrogen gas bubbles occurs at the cathode. These gas bubbles facilitate the removal of pollutants by floating them to the surface of the solution for easy collection (Mollah, Morkovsky, Gomes, Kesmez, & Parga, 2004). EC is also easily operated due to the simplicity of the equipment used, which allows for complete automation (Bazrafshan, Mohammadi, Ansari-Moghaddam, & Mahvi, 2015). The wastewater treatment by EC produces clear, colorless, and odorless water (Holt, Barton, & Mitchell, 2005). The floc formation in the EC process is much larger compared to coagulation floc formation, making it easier for filtration (Mollah, Morkovsky, Gomes, Kesmez, & Parga, 2004). EC also produces much less sludge volume compared to coagulation process (Bazrafshan, Mohammadi, Ansari-Moghaddam, & Mahvi, 2015). Since electric current is applied in the EC process, the collision of particles is faster, thus removing even smaller particles (Holt, Barton, & Mitchell, 2005). However, for the EC process it is necessary to replace the sacrificial anode since it dissolves into the solution (Mollah, Morkovsky, Gomes, Kesmez, & Parga, 2004).

EC process is used to treat numerous wastewaters such as textile wastewater, food industry wastewater, paper industry wastewater, petroleum industry wastewater, and water containing heavy metals.

1.4 Factors Influencing Efficiency of Electrocoagulation

There are several factors that can influence the efficiency of EC, such as pH, flow rate, electrode arrangement, cell gap, current density, current efficiency, initial concentration of pollutant, type of anode, type of power supply, and other wastewater features. These factors are mainly operating conditions that can be modified. However, the most important factors will be focused on for this section

1.4.1 Current Density and Current Efficiency

Current density is the ratio of the electric current over the electrode surface area and it plays an integral role in the efficiency of EC. Current density determines the dosage of the coagulant at the anode and the evolution of the hydrogen gas at the cathode. The bubble density affects the hydrodynamics of the system, which affects the mass transfer between pollutants, coagulant, and gas microbubbles and also influences the collision rate of coagulated particles that results in floc formation (Holt, Barton, Wark, & Mitchell, 2002). A current density that will produce the optimum EC process has to be determined so as to not waste electrical energy by using too much or produce a less efficient EC process by not using enough. The determination of the optimum current density is also affected by other parameters such as pH, temperature, and water flowrate (Guohua, 2004). Current efficiency is the ratio of the actual current density being used in an electrochemical reaction to the total current density applied to the system. The current efficiency should be monitored to see if there is any effect of fouling on the electrodes.

1.4.2 pH

In wastewater treatment, the pH of the solution is crucial to how the wastewater will be treated. In coagulation process, there are different types of coagulants that perform excellently at specific solution pH, so the pH of that solution has to be adjusted to meet the requirements of the coagulant. The same applies for the EC process, where the pH of the solution is critical to achieving a high-efficiency EC process. For the EC process, the pH governs the hydrolyzed metal species generated (Malakootian, Masoorian, & Moosazadeh, 2010). The metal hydroxides formed from the chemical reaction in the EC process are influenced by the pH of the solution, which means the appropriate pH has to be selected to achieve an optimum condition for EC.

1.4.3 Flow Rate

As in any wastewater treatment plant, the flow of the water being treated is vital to the whole operation. Wastewater treatment technologies and equipment are sized by a manufacturer for a specific customer based on the operating conditions of the customer's plant. In a coagulation process, the flow of the solution is very crucial, especially when conducting the bench scale testing. A flow rate that is too high might not give the chemical coagulant enough time to mix and coagulate properly, and a low flow rate could introduce an amount of chemical coagulant in excess of what is needed. Improper accountability for the flow rate could waste chemicals or minimize the efficiency of the treatment. The same consideration applies for the EC process. If the flow is not controlled, it can greatly affect the coagulant generation, which would affect the metal hydroxides generated and the reaction with the pollutants, thereby affecting the efficiency of the EC process.

1.4.4 Initial Concentration of Pollutant

One of the most important factors that influence the efficiency of the EC process is the initial concentration of the pollutant. The removal rate of the pollutant is dependent on how much pollutant there is to remove. In an optimum EC process, and depending on the operation time, the removal rate of the pollutant will eventually reach a steady and high removal point. However, in the beginning stages of the EC process, or without a high-efficiency EC process, a difference in the removal rate will occur for the same pollutant at different initial concentrations.

1.4.5 Cell Gap

The EC device mainly consists of the electrodes, and the distance between both anode and cathode is crucial as the wastewater being treated passes through the channel. In a real-life practical wastewater treatment for EC, there would be multiple anodes and cathodes in a specific cell arrangement. The effect of cell gap must be monitored.

1.5 Literature Review

The scarcity of water is one of the greatest current and future difficulties that humankind is facing as the world’s population and water consumption rates continue to increase. This has led to the importance in developing cost-effective, reliable, and environmentally friendly wastewater treatment technologies that would be able to reuse a significant amount of water produced from various industries (Moussa, 2017). There are many different technologies that use membrane technology to further treat the wastewater so as to give access and opportunity to reuse. However, these technologies require high maintenance and are very costly. However, for the EC process, with the proper high-efficiency operating conditions, the treated wastewater can be reused depending on the application.

The soluble aluminum resulting from the electrochemical process can undergo different hydrolysis reactions producing several aluminum hydroxide monomeric species such as

$Al(OH)^{2+}$, $Al(OH)^{+}{}_{2}$, and $Al(OH)_3$ and polymeric species such as $Al_2(OH)_2{}^{4+}$, $Al_6(OH)_{15,}{}^{3+}$, and $Al_8(OH)^{4+}{}_{20}$ (Mechelhoff, Kelsall, & Graham, 2013). For the simplicity in mathematical modeling, the EC process mainly focuses on the monomeric species generated.

The generation and mass transfer of the electrogenerated hydroxides and flocs will have important effect on pollutant removal (Harif, Khai, & Adin, 2012). The percent of pollutant removal is the overall and key factor in any wastewater treatment, which is why it is best to determine the optimum condition for a high-efficiency EC process.

The flocs generated from the EC process have potential for adsorption of pollutants (Lakshmanan, Clifford, & Samanta, 2010), which means the metal ionic species distribution is a determining factor for EC efficiency (Canizares, Jimenez, Martinez, Saez, & Rodrigo, 2007). As described, flotation is a key part of the EC process; the bubbles formed through the oxygen and hydrogen evolution in the electrodes increase the efficiency of removal by flotation (Chen, Chen, & Yue, 2002).

$Al(OH)_3$ is the final-form and main species for pH at 7 (Holt, Barton, Wark, & Mitchell, 2002), and adsorption and coagulation particularly depend on pH to achieve a high-efficiency EC process (Jimenez, Saez, Martinez, & Canizares, 2012) .The optimum pH for the EC process is at a neutral stage, and based on the aluminum equilibrium solubility diagram depicted in Figure 3, it can be confirmed that $Al(OH)_3$ is the final-form species at pH of 7.

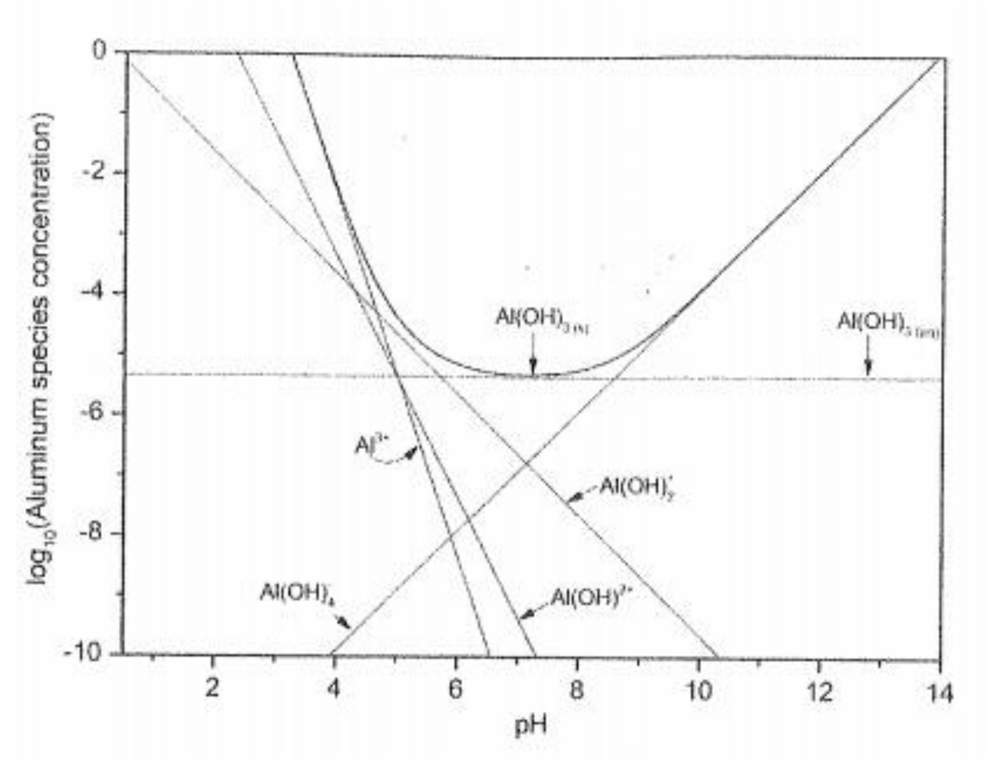

Figure 3. Aluminum equilibrium solubility diagram.

In EC process, the flow rate affects the floc formation and flotation effect caused by the hydrogen and oxygen bubbles (Peipei et al., 2018). The flow rate is introduced as a variable in the mass transport of the ionic species, which will be discussed more in the next chapter. As explained, there are two separate processes occurring in the EC process: the electrochemical reaction of the generation of ionic species and the chemical reaction taking place, which lead to the adsorption of the pollutant. The pollutant is adsorbed at the surface of the generated flocs, which makes the removal of pollutant similar to conventional adsorption except for the generation of coagulants, and since the amount of coagulant can be estimated over time, the pollutant removal can be modeled by the adsorption phenomenon (Balasubramanian, Toshinori, Ahmed, & Srinivasakannan, 2009).

Despite the success of electrocoagulation for the treatment of various types of wastewater, its application as a possible treatment option for petroleum wastewater is scarce (Elnaas, 2009). The EC process can be veered towards treating diesel and oily bilge wastewater. Electrocoagulation has the potential to be used for the treatment of agro-industry wastewater due to the ability of removing colloidal particles and suspended solids relating to organic and inorganic contents (Drogui, 2008). It is very important to treat agriculture-based waste as it contains manure and other wastes from farms, and etc. The EC process is particularly more effective in treating wastewaters with light suspended particles such as oily restaurant wastewater, with the application of electroflotation (Chen & Chen, 2000). The EC can also be used to treat textile wastewater with dye-containing solutions for the removal of COD, color, and turbidity (Kobya, 2003). The key aspects that affect the efficiency of EC must be monitored closely and discussed further in this paper.

1.6 Objective of Study

The objective of this study is to improve the efficiency of the EC process by determining the optimum operating conditions. A high-efficiency EC process will result is a high percent of removal of specific pollutants. One of the main areas in which the EC process is utilized is the removal of heavy metals, and for this paper, the focus will be on the removal of arsenic from wastewater. Arsenic has become a major concern in wastewater globally, and numerous treatment techniques are used to tackle the pollutant. Recently, USEPA has lowered the maximum contaminant level for arsenic from 50 to 10 mg/L, and the removal by adsorption using iron and aluminum salts is more commonly used because of the ability to reduce arsenic less than 2mg/L (Balasubramanian, Toshinori, Ahmed, & Srinivasakannan, 2009). The implementation of the EC process and its adsorption-like capabilities will serve as a crucial technology to reduce arsenic to an environmentally safe level. The determination of high percent removal of arsenic will depend on a high-efficiency EC process. To achieve this, the study will focus on determining and utilizing the optimum operating conditions for the key factors that influence the performance of the EC process: current density, pH, flow rate, and the initial concentration of the pollutant. This will be performed using a mathematical model that will be simulated using COMSOL Multiphysics 5.3 software.

CHAPTER 2: MATHEMATICAL MODELING OF ELECTROCOAGULATION

The EC process has two main parts: the generation of the final-form metal hydroxides and the adsorption process, where the pollutant reacts with the metal hydroxide and rises to the surface because of the evolution of hydrogen gas. However, as shown in Figure 4 below, there are three different reactions required to complete the EC process. As the electric current is applied to the system, there is an anodic or oxidation reaction occurring at the anode and a cathodic or reduction reaction taking place at the cathode. The anodic reaction produces Al^{3+}, which acts as the coagulant in the anode, and the cathodic reaction produces OH^- as the cathode.

The first reactions describe the generation of ionic species from electrochemical reaction. The second reaction is the formation of ionic complex by chemical reaction. The chemical reaction between the Al^{3+} and H_2O forms various monomeric and polymeric species (metal hydroxides), and at a pH of 7 the final-form of $Al(OH)_3$ is achieved. The last reaction taking place is the adsorption of pollutants. Figure 5 below describes the adsorption process in which the precipitates formed from the first two reactions mix with the pollutants and are carried to the surface by the hydrogen bubbles created by the cathodic reaction as shown in Figure 2.

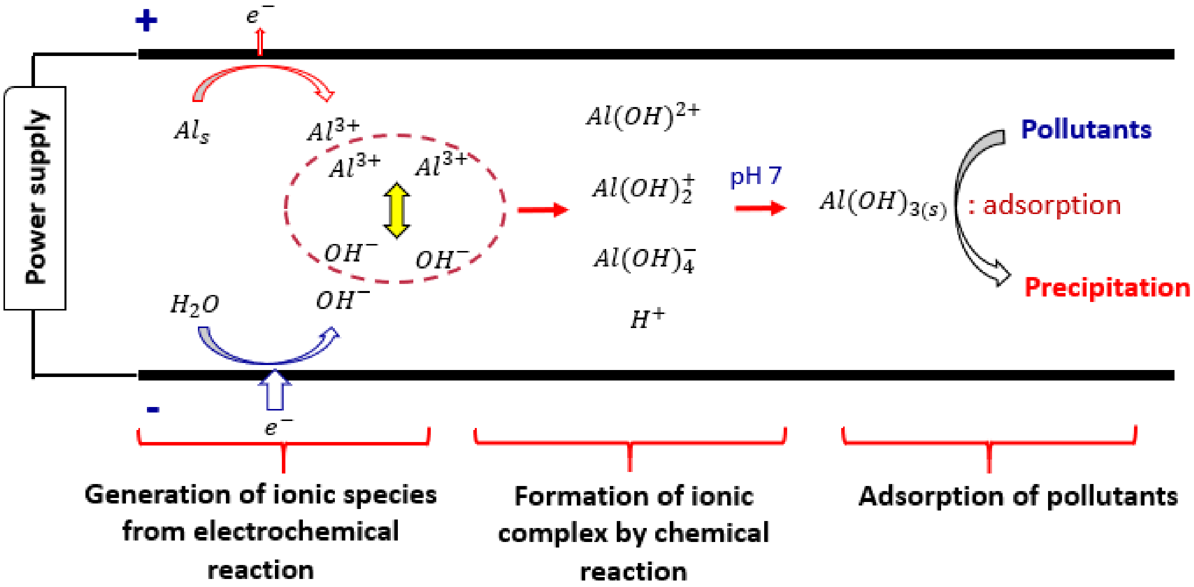

Figure 4. The three reactions in a complete EC process.

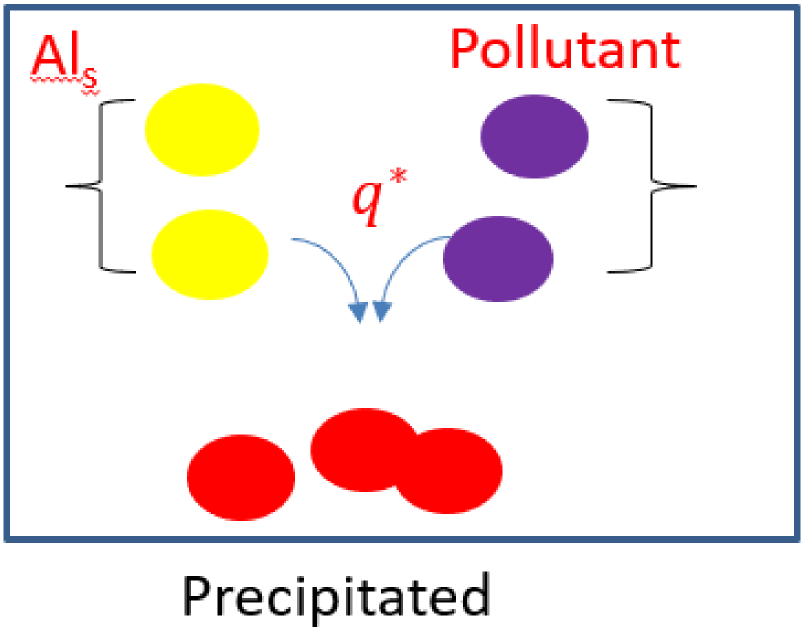

Figure 5. The adsorption process in EC.

As mentioned earlier, the mathematical modeling in consideration will focus on the removal rate of arsenic. Arsenic is one the most concerning heavy metals present in wastewater and is hazardous to the environment. Graca (2019) conducted an EC process experiment and model for the removal of arsenic. The mathematical model considers unsteady-state and zero dimension, which means the average removal rate is being considered and assumed to be the same at all locations. The dissolution of aluminum effect was considered, and the physical adsorption phenomena was utilized for the model. However, the effect of mass transfer was not considered. A second model that was assessed was by Lu, Wang, Ma, Tang, and Li (2017), who considered a steady-state two-dimensional model. This model considers the effect of mass transfer, but the dissolution of aluminum and the physical adsorption phenomena was not considered.

The goal is to incorporate the properties of the two models and take into consideration the affecting factors in determining the optimum operating conditions for the EC process. The main factors that influence the efficiency of the EC process will be considered to determine a high removal rate of arsenic. There are three Phases to this model as shown in Table 1 below. The first Phase will consider a zero-dimensional unsteady-state model, the second Phase will consider a one-dimensional unsteady-state model, and the third Phase will consider a two-dimensional unsteady-state model. Each Phase will add a new operating condition to see the effect on the simulations and determine the optimum EC process and high removal rate of arsenic.

Table 1 : Operating Conditions for the Three Modeling Phases (Unsteady-state)

Phase 1	Phase 2	Phase 3
0D	1D	2D
Effect of current	Diffusion	Effect of current and current efficiency
Initial concentration of pollutant		Convection
		Cell gap

2.1 Phase 1

Phase 1 considers a time-dependent unsteady-state zero-dimensional analysis with the effect of current intensity to determine the removal rate of arsenic. The assumptions for this model are shown in Table 2. There is anode dissolution in the EC process, which means the metal anode will have to be replaced after dissolving and is deemed ineffective. In the models that will be analyzed, the shrinkage of the anode is neglected. Three different current densities (40 mA, 100 mA, and 190 mA) will be considered for this model, and the goal is to determine which current density will produce a higher solid particle generation or floc formation. Once the current is determined, it will be used to determine the removal rate of arsenic and also used in the other Phases considered in this paper. Also, three different initial concentrations (2 mol/m^3, 3 mol/m^3, and 4 mol/m^3) of arsenic are considered to show how initial concentration affects the EC process.

Table 2 : Assumptions for All Models

pH of 7 is assumed because the EC process performs better at neutral pH
$Al(OH)_3$ is the final-form and main species for pH at 7
The shrinkage of the anode due to its dissolution is neglected
The process is considered isothermal
The effect of the presence sodium and chloride ions is not accounted in the model
Ionic complex with contaminant is governed by Langmuir isotherm

2.1.1 Formulations and Operating Conditions

The two main parts of the EC process will consider the generation of the total and soluble aluminum concentration and the removal rate of arsenic. The simulation is run for 90 minutes and the electric current is shut off after 30 minutes to see the effect on the solid and soluble aluminum concentration. Before considering the formulations in the model, it is best to show the key reactions in the EC process.

The electrogeneration of the coagulant, Al^{3+}, and OH^- is given below:

Anode: $Al \rightarrow Al^{3+} + 3e^-$ (2.1)

Cathode: $2H_2O + 2e^- \rightarrow H_2 + 2OH^-$ (2.2)

The formation of soluble and insoluble hydroxides is given below:

$$\mathrm{Al^{3+} + H_2O \rightleftharpoons Al(OH)^{2+} + H^+} \quad (2.3)$$

$$Al(OH)^{2+} + H_2O \rightleftharpoons Al(OH)_2^+ + H^+ \quad (2.4)$$

$$Al(OH)_2^+ + H_2O \rightleftharpoons Al(OH)_3 + H^+ \quad (2.5)$$

$$Al(OH)_3 + H_2O \rightleftharpoons Al(OH)_4^- + H^+ \quad (2.6)$$

$$H_2O \rightleftharpoons H^+ + OH^- \quad (2.7)$$

The reaction rates (Graca, 2019) of each chemical reaction are given below:

$$\mathrm{R_1} = \mathrm{k_{1-f}}\left(\mathrm{Al^{3+}} - \frac{\mathrm{Al(OH)^{2+}\cdot H^+}}{\mathrm{K_1}}\right) \tag{2.8}$$

$$\mathrm{R_2} = \mathrm{k_{2-f}}\left(\mathrm{Al(OH)^{2+}} - \frac{\mathrm{Al(OH)_2^{+}\cdot H^+}}{\mathrm{K_2}}\right) \tag{2.9}$$

$$\mathrm{R_3} = \mathrm{k_{3-f}}\left(\mathrm{Al(OH)_2^{+}} - \frac{\mathrm{Al(OH)_3\cdot H^+}}{\mathrm{K_3}}\right) \tag{2.10}$$

$$\mathrm{R_4} = \mathrm{k_{4-f}}\left(\mathrm{Al(OH)_3} - \frac{\mathrm{Al(OH)_4^{-}\cdot H^+}}{\mathrm{K_4}}\right) \tag{2.11}$$

$$\mathrm{R_w} = \mathrm{k_{wf}}\left(1 - \frac{\mathrm{OH^-\cdot H^+}}{\mathrm{K_w}}\right) \tag{2.12}$$

where R_i, k_i, and K_i, are the reaction rates, kinetic constant of hydrolysis reaction, and thermodynamic equilibrium constants respectively. The kinetic constants and equilibrium constants are shown in Table 3 below.

Table 3 : Thermodynamic Equilibrium Data and Kinetic Constants for the Hydrolysis Reactions

i	k_f (Abodi et al., 2012)	Ki (Xiao, Zhang, & Lee, 2008)
1	4.2×10^{4} L/(mol·s)	9.6×10^{-6}
2	4.2×10^{4} L/(mol·s)	5.3×10^{-5}
3	5.6×10^{4} L/(mol·s)	2.0×10^{-6}
4	2.5×10^{-7} L/(mol·s)	2.7×10^{-9}
w	1.52×10^{-6} L/(mol·s)	1.0×10^{-14}

The governing equations shown below are general form of flux of ionic species in solution, mass conservation, charge conservation, and current relation.

$$\vec{N}_i = \underbrace{-z_i u_i F c_i \vec{\nabla}\Phi}_{\text{Migration}} - \underbrace{D_i \vec{\nabla} c_i}_{\text{Diffusion}} + \underbrace{\vec{v} c_i}_{\text{Convection}} \tag{2.13}$$

$$\frac{\partial c_i}{\partial t} = -\vec{\nabla} \cdot \vec{N}_i + R_i \tag{2.14}$$

$$\sum_i z_i c_i = 0 \tag{2.15}$$

$$\vec{\imath} = F \sum_i z_i \vec{N}_i \tag{2.16}$$

where:

D_i = diffusion coefficient

z_i = charge number of species

Φ = electric potential

v = flow velocity vector

μ_i = ionic migration rate

F = Faraday constant

C_i = concentration of ionic species

The chemical reaction rate equation (Graca, 2019) of each species is given below:

$$R_i|_{i=Al^{3+}} = -R_1 \tag{2.17}$$

$$R_i|_{i=Al(OH)^{2+}} = R_1 - R_2 \tag{2.18}$$

$$R_i|_{i=Al(OH)_2^+} = R_2 - R_3 \tag{2.19}$$

$$R_i|_{i=Al(OH)_{3(s)}} = R_3 - R_4 \tag{2.20}$$

$$R_i|_{i=Al(OH)_4^-} = R_4 \tag{2.21}$$

$$R_i|_{i=H^+} = R_1 + R_2 + R_3 + R_4 + R_w \tag{2.22}$$

$$R_i|_{i=OH^-} = R_w \tag{2.23}$$

$$\frac{\mathrm{d[Al]_D}}{\mathrm{dt}} = \frac{\mathrm{I}\in_\mathrm{c}}{\mathrm{FZV}} - \mathrm{k_{cg}}(\mathrm{[Al]_D} - \mathrm{[Al]_{sat}}) \tag{2.24}$$

where:

$[Al]_D$ = soluble aluminum

k_{cg} = coagulation constant

$[Al]_{sat}$ = saturation concentration of aluminum

V = solution volume

ϵ_c = current efficiency

Now considering the adsorption of the pollutant, the Langmuir adsorption isotherm theory is used to determine the amount of adsorbate adsorbed as a function of concentration, which will be used to determine the removal rate of arsenic. The mathematical modeling approach utilized is similar to the one used by Graca (2019), which considers the equilibrium of the solid and liquid Phase of the concentration. The equations are given below:

$$C_o - C^* = q^*[Al]_s \tag{2.25}$$

$$[Al]_s = [Al]_T - [Al]_D \tag{2.26}$$

$$q^* = \frac{QKC^*}{C_0 + (K-1)C^*} \tag{2.27}$$

$$C^* = -\frac{C_0(2-K) + QKAl_S}{(K-1)} + \frac{C_0^2}{C^*(K-1)} \tag{2.28}$$

$$RR = \frac{C_0 - C^*}{C_0} * 100 \tag{2.29}$$

where:

Q = Solid capacity

K = Separation factor

C^* = Liquid equilibrium pollutant concentration

C_0 = Initial pollutant concentration

q^* = Solid equilibrium pollutant concentration

These are the conditions that will be simulated for this model. All equations described above will be utilized except for the addition of mass transfer effect, which considers migration, convection, and diffusion.

2.2 Phase 2

Phase 2 considers an unsteady-state one-dimensional analysis with the effect of diffusion. Diffusion is a mass transfer phenomenon that causes distribution of a chemical species to become more uniform in space as time passes. If the concentration of a species is not uniformly distributed, the effect of diffusion causes the evolution of the concentration to be more uniform. The results achieved in Phase 1 for the current intensity, which yields high removal rate for arsenic, will serve as the optimum operating condition for Phase 2.

The model in Phase one is time-dependent while Phase 2 considers the effect of diffusion in the spatial dimension (x-axis). The mass transport equation in equation (2.13) will now be used in the Phase 2 model for diffusion, while neglecting convection and migration. Figure 6 below shows the distance between the anode and cathode, which is the region where the EC reaction takes place on the x-axis. In Phase 1, it is assumed that the removal rate of arsenic is an average since the model is zero dimensional. However, in Phase 2 the removal rate at any time and location (x-axis) between the anode and cathode can be determined.

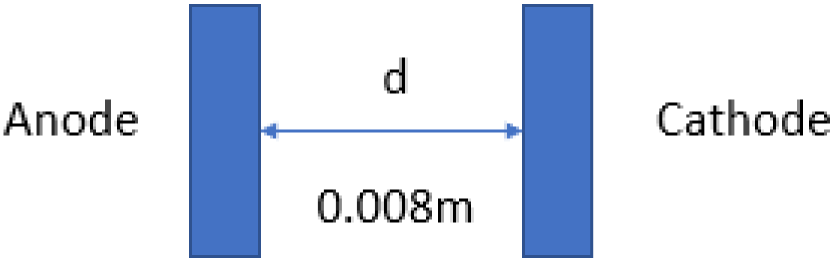

Figure 6. Phase 2: Distance between electrodes.

The simulation assumptions made in Table 2 applies for Phase 2. The boundary conditions (Lu, Wang, Ma, Tang, & Li, 2017) considered for this model are given below:

Anode: $N_i|_{i=Al^{3+}} = \frac{Io}{3F}$; $N_i|_{i=other\ species} = 0$ (2.30)

Cathode: $N_i|_{i=OH^-} = -\frac{Io}{F}$; $N_i|_{i=other\ species} = 0$ (2.31)

where:

$N_i|_{i=Al^{3+}}$ = mass transfer flux species at anode surface

$N_i|_{i=OH^-}$ = mass transfer flux species at cathode surface

With these operating conditions, the model in Phase 2 will produce the removal rate of arsenic in 1D. There were different sets of diffusivities for Al^{3+} and OH^- simulated to see the effects on the removal rate of arsenic. However, diffusion coefficient is an intrinsic component that cannot be adjusted by physical means, which is why the concept of convection is crucial for the model and mass transfer. Table 4 below shows the diffusivities that were simulated.

Table 4 : Diffusion Coefficient of Ionic Species

Species	D1 (m^2/s)	D2 (m^2/s)	D3 (m^2/s)
Al^{3+}	1.011×10^{-9}	1.011×10^{-7}	1.011×10^{-5}
H^+	9.308×10^{-9}	9.308×10^{-9}	9.308×10^{-9}
OH^-	5.280×10^{-9}	5.280×10^{-7}	5.280×10^{-5}
Others	1.0×10^{-9}	1.0×10^{-9}	1.0×10^{-9}

2.3 Phase 3

Phase 3 considers an unsteady-state two-dimensional analysis with the effect of diffusion, convection, cell gap, current, and current efficiency. Convection is mass transfer due to the average velocity of all molecules. For dilute species transport, one component (either convection or diffusion) dominates the momentum of the system; it is normal to assume velocity as the dominant component (COMSOL, 2015). Figure 7 shown below is the 2D geometry of the domain considered for the model in Phase 3, which is the height of the anode and cathode and the distance between the electrodes. The inlet and outlet of the fluid are defined, and the same operating conditions for flux used in Phase 2 are considered. The distance between the electrodes are important factors as well, as discussed, and different conditions will be modeled to see the effect on EC. Also, current and current efficiency are factors that will be controlled and considered for the 2D model. The objective for this model is to determine how these operating conditions affect the performance of the EC process and the best conditions for an optimum EC process, which ultimately determines the best removal rate for arsenic.

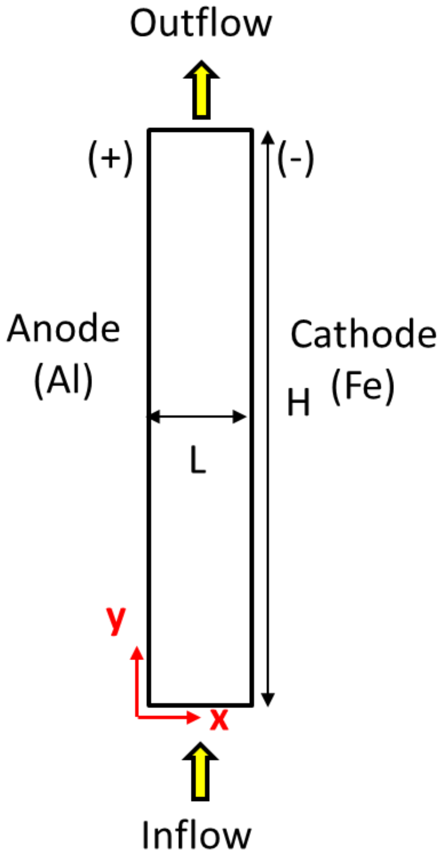

Figure 7. 2D EC model for Phase 3.

In any electrochemical cell, laminar flow is mainly considered. In this case, if the retention time is long, the fluid will take time to pass, which will give more time for the diffusion and hydrolysis reaction to have an effect on the pollutant. The simulation ran for three different velocities, currents, current efficiencies, and cell gaps to compare the effects on the EC process for the removal of arsenic. Navier-Stokes equation for fluid flow and velocity profile equation is given below:

$$\rho\left(\frac{\partial \vec{v}}{\partial t} + \vec{v} \cdot \nabla \vec{v}\right) = -\nabla p + \mu \nabla^2 \vec{v} + \rho \vec{g} \tag{2.32}$$

$$U_p = \left(1/(2\mu)\right) * \frac{dP}{dx} * [x^2 - hx] \tag{2.33}$$

where:

ρ = density of fluid

g = gravity

U_p = velocity profile

P = Pressure

μ = dynamic viscosity of fluid

The initial and boundary conditions are given below:

Initial concentration of arsenic = 4 mol/m^3

Initial concentration of Al^{3+} = 6 mol/m^3

Initial concentration of other species = 0 mol/m^3

At the surface of the electrode (x = 0):

$$N_i|_{i=Al^{3+}} = \frac{Io}{3F} \quad ; \quad N_i|_{i=other\ species} = 0$$

At the surface of the electrode (x = L):

$$N_i|_{i=OH^-} = -\frac{Io}{F} \quad ; \quad N_i|_{i=other\ species} = 0$$

At the inlet (y = 0):

$N_i|_{i=other\ species} = 0$; Velocity = 0.33 – 1.32 cm/s

At the outlet (y = H):

$N_i|_{i=other\ species} = 0$; Pressure = 0 Pa

No slip condition was set at the surface of the electrodes, and gravity effect in both directions is neglected. A 3-minute simulation time was used for the models.

2.4 Objective

Each model that was simulated to determine the effect on the aluminum solid particle generation and the removal rate of arsenic. Phase 1 considers a 0D model for the removal rate over time by showing the effect of current density and initial concentration of pollutant. Phase 2 considers the 1D distribution of the species between the electrodes and the effect of diffusion over time, while Phase 3 considers the 2D distribution of species with the effect of current, current efficiency, cell gap, and convection. The overall objective was to attain a high-efficiency EC process by obtaining the optimum operating conditions.

CHAPTER 3: RESULTS AND DISCUSSION

Three different models were simulated to determine the total and soluble aluminum generation and the removal rate for the heavy metal, arsenic. Many different conditions were considered for all the models to better understand the factors that influence the efficiency of EC operation.

3.1 Outcomes of Phase 1

In an EC experiment conducted by Graca (2019), the total and soluble aluminum concentrations were determined for three different current intensities (40 mA, 100 mA, 190 mA) to analyze the best of the three. The optimum current intensity is used to determine the removal rate of arsenic. The experiment was conducted for 90 minutes and the current was shut off after 30 minutes. The 0D mathematical model in Phase 1 is used to compare total aluminum concentration of the experimental results for the first 30 minutes before shutting off the current. As shown in Figure 8 below, the mathematical model depicting the effect of the current intensities reasonably predicts the experimental results. From the graph it is noticed that total aluminum concentration increases with increasing current intensity.

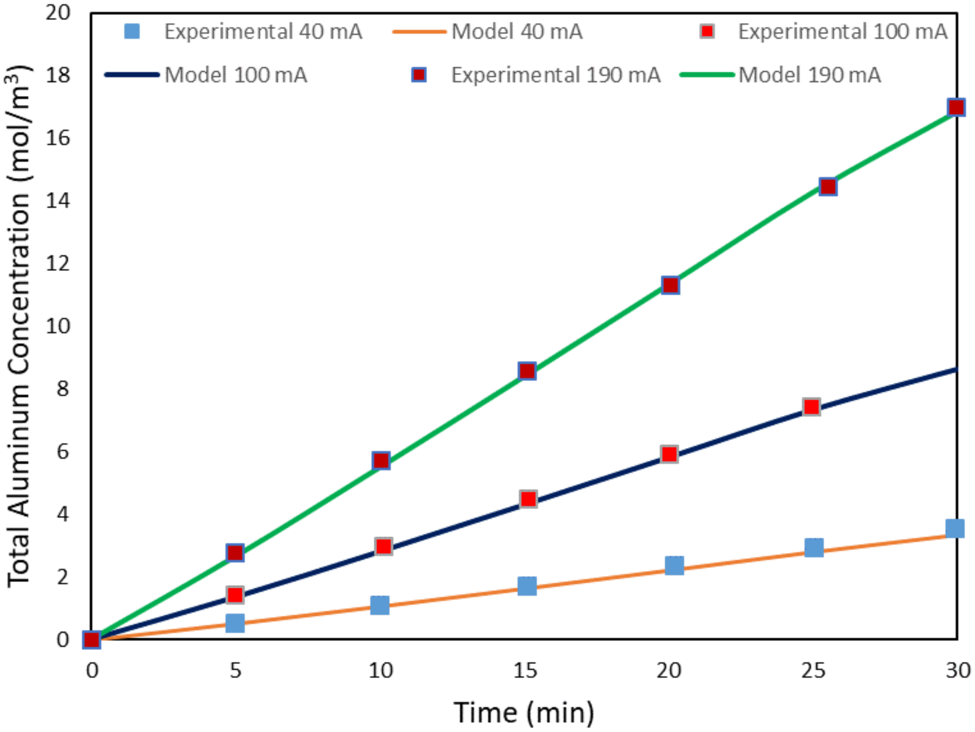

Figure 8. Total aluminum concentrations at different current intensities.

Now looking at the complete simulation results for the model, the total and soluble aluminum concentrations are compared. The goal is to determine the highest concentration difference between the total and soluble aluminum concentration. The difference in the two concentrations is the solid aluminum concentration as shown in equation (2.26), which should show the highest and fastest solid particle formation. The results are shown in Figures 9-11.

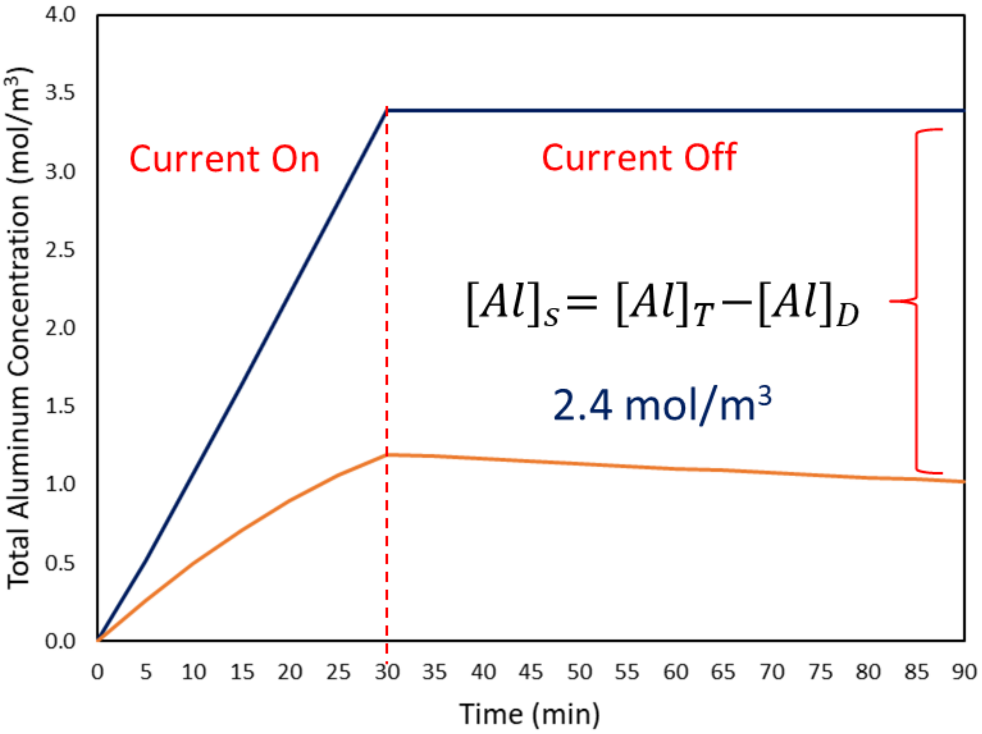

Figure 9. Total and soluble aluminum concentration at 40mA.

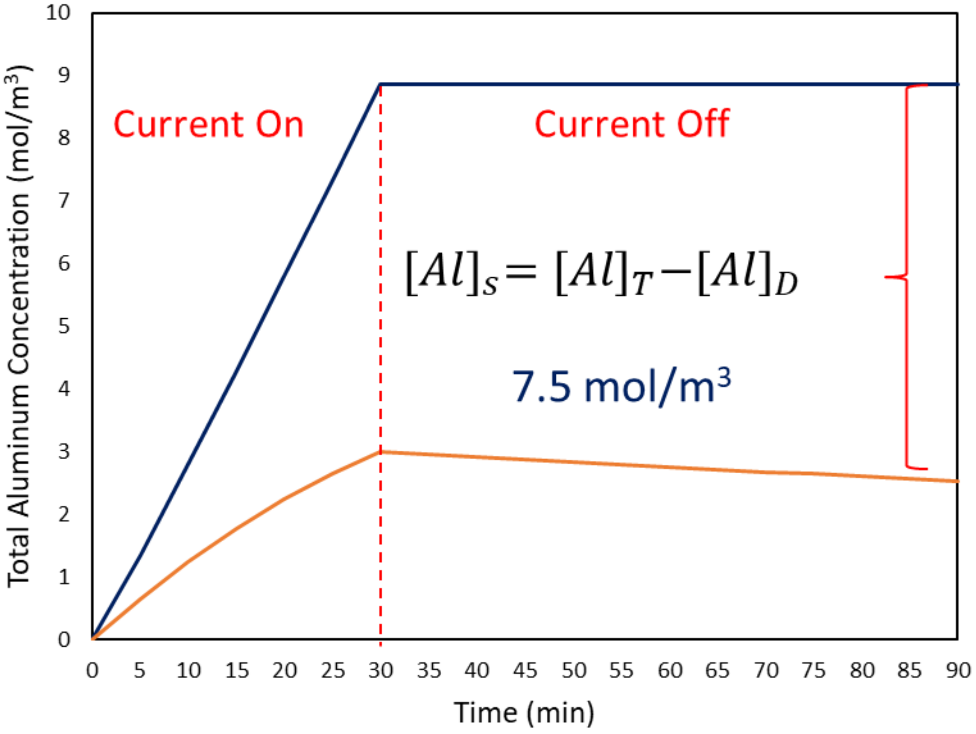

Figure 10. Total and soluble aluminum concentration at 100 mA.

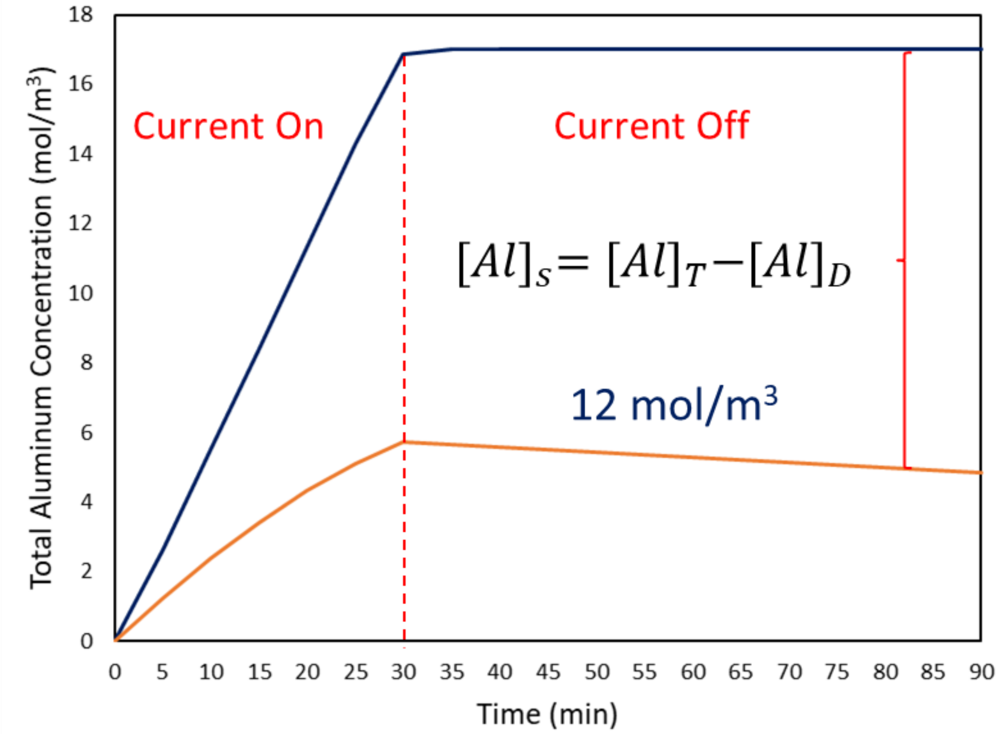

Figure 11. Total and soluble aluminum concentration at 190 mA.

Figure 9 shows the total and soluble aluminum concentration for a current intensity of 40 mA. As shown, the soluble aluminum concentration decreases after 30 minutes when the current is off and a final solid aluminum concentration of 2.4 mol/m^3 was reported. The final reported solid aluminum concentration in Figure 10 for the 100 mA current intensity was 7.5 mol/m^3 and 12 mol/m^3 for the 190 mA simulation in Figure 11. This suggests that the highest current intensity produced a higher and faster solid particle formation, and in the EC process this is favorable for the removal of the pollutant.

The optimum operating condition in terms of current intensity has been determined to be 190 mA. This result is used to determine the removal rate of arsenic while comparing the effect of three different initial concentrations of arsenic on the EC operation. In the initial stages as shown in Figure 12, the lower the initial concentration, the faster the removal rate. As treatment time continues, it can be noticed that the removal rate of arsenic for all initial concentrations achieve closer percent pollutant removal (2 mol/m^3 – 98%, 3 mol/m^3 – 97%, 4 mol/m^3 – 96%).

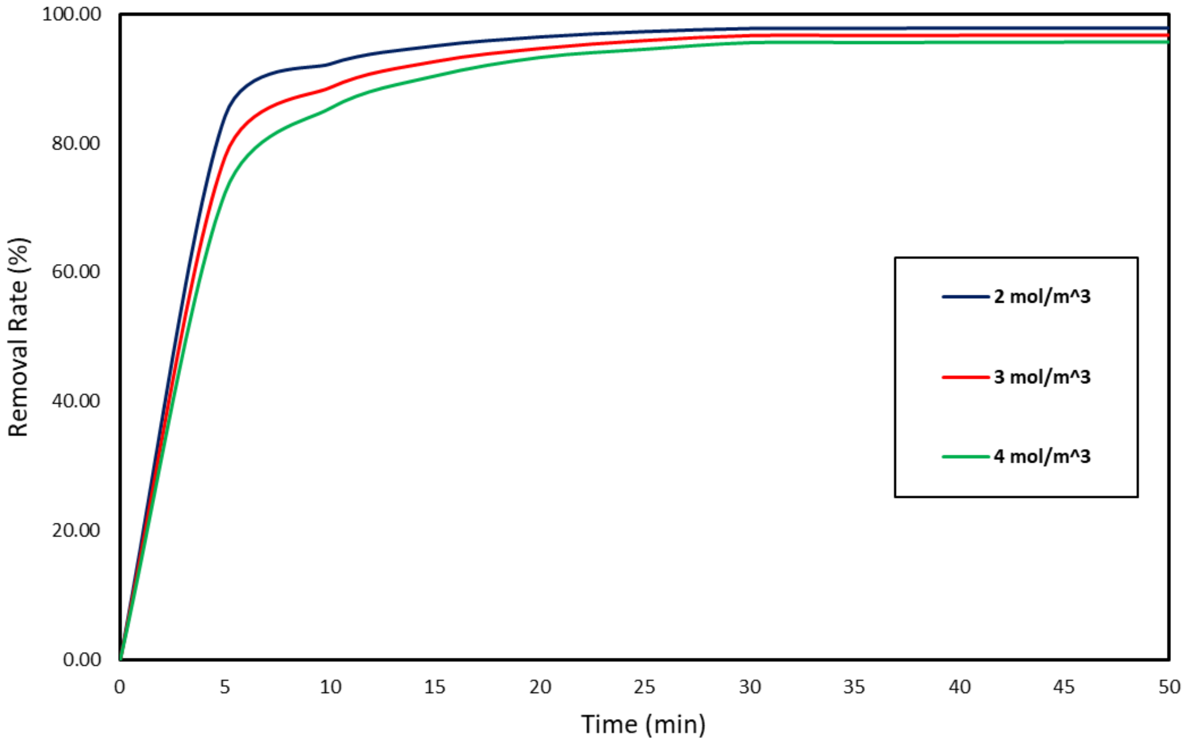

Figure 12. Removal rate of arsenic, Phase 1.

3.2 Outcomes of Phase 2

The model in Phase 2 considered the effect of diffusion in the EC process and also the distribution of species in the x-axis. Different sets of diffusion coefficients were used for Al^{3+} and OH^- (anode and cathode) while the other species were kept at constant. The results for total and soluble aluminum concentration and removal rate of arsenic for a simulation time of 50 minutes are shown in Figures 13-18.

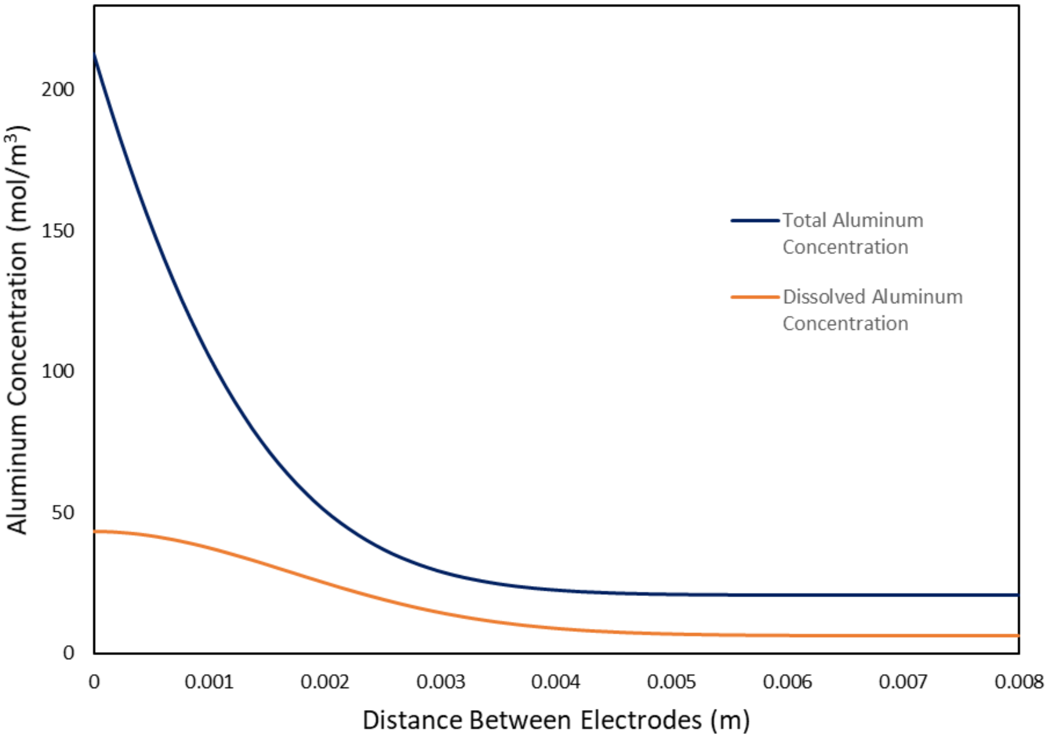

Figure 13. Total and soluble aluminum concentration, diffusivity D1.

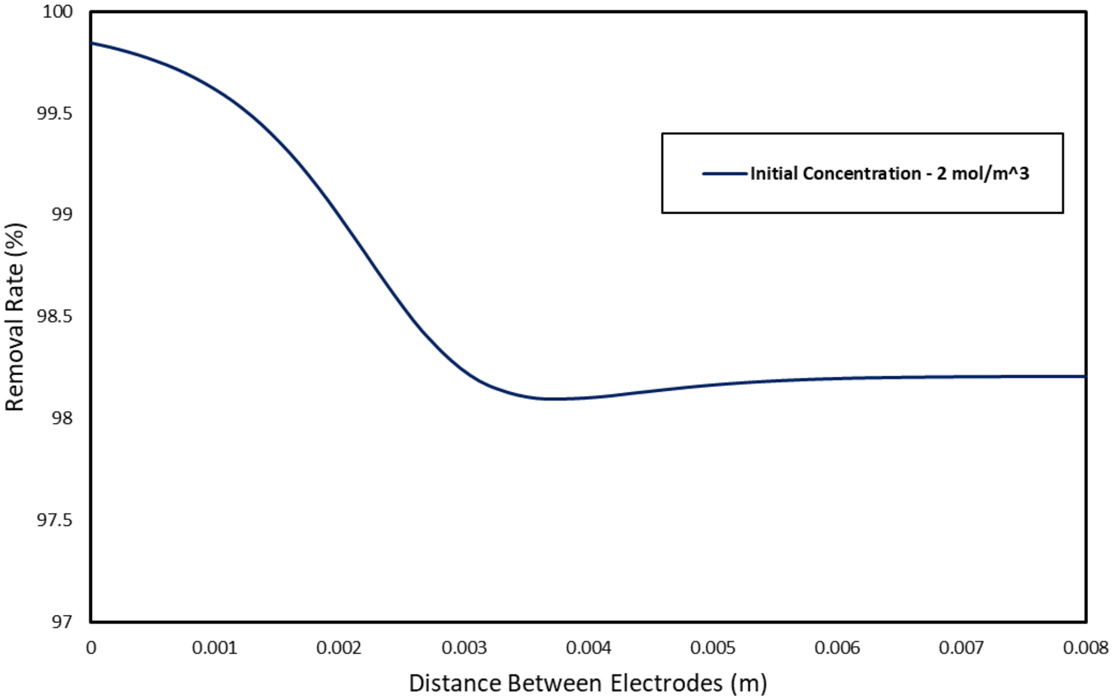

Figure 14. Removal rate of arsenic, diffusivity D1.

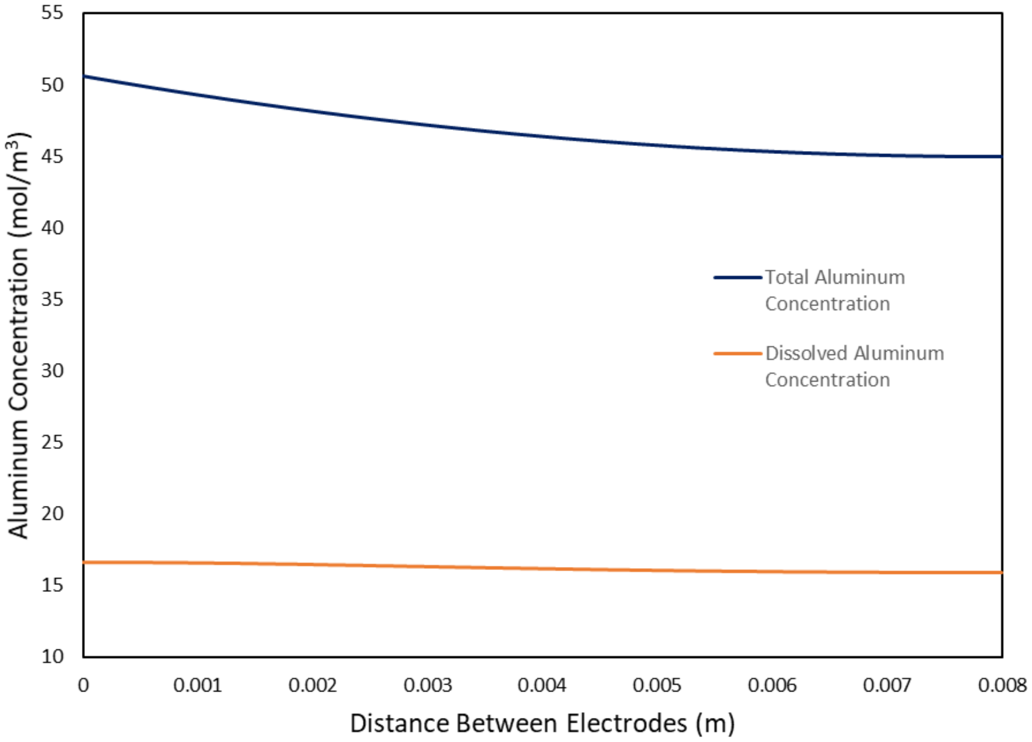

Figure 15. Total and soluble aluminum concentration, diffusivity D2.

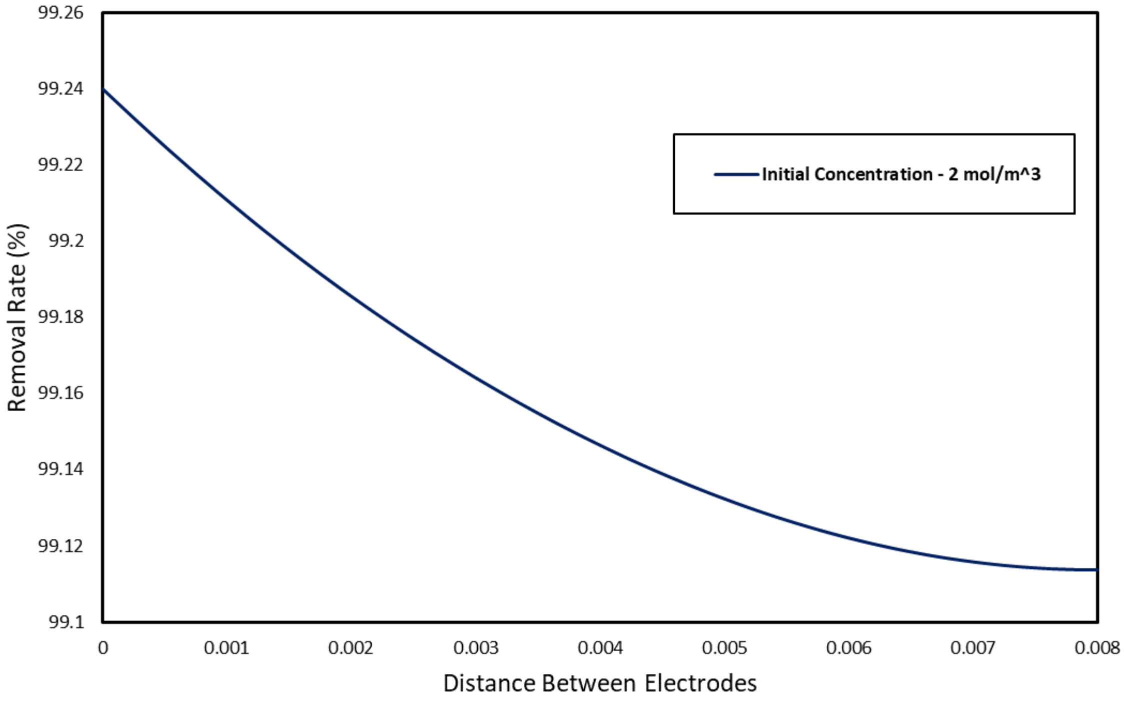

Figure 16. Removal rate of arsenic, diffusivity D2.

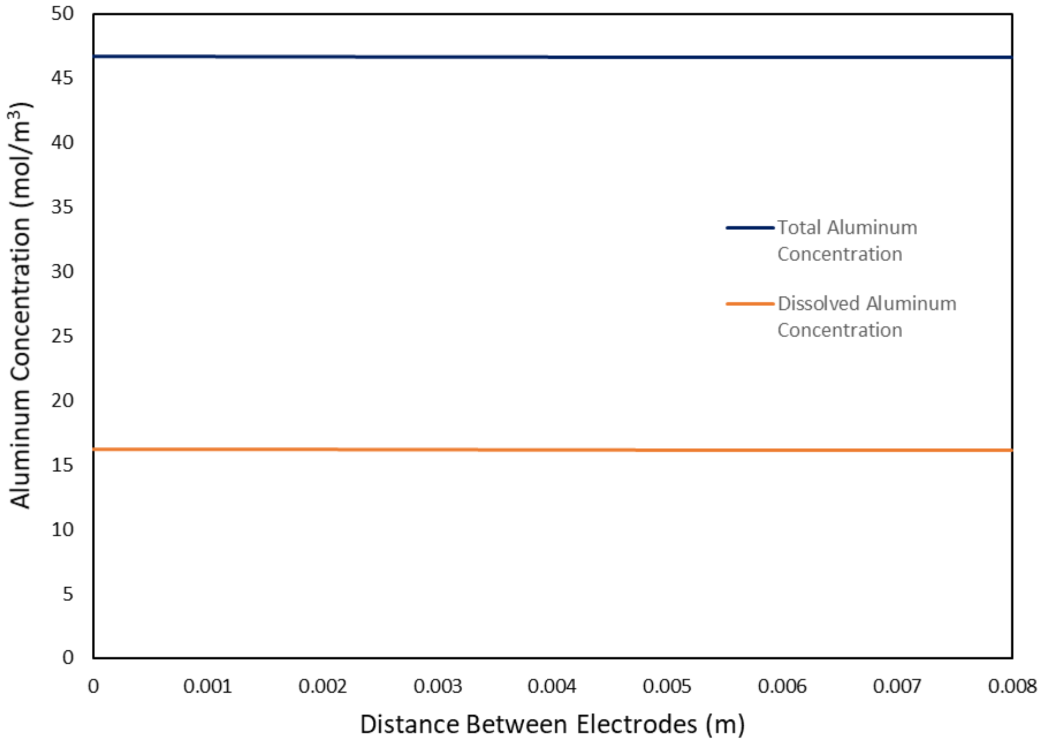

Figure 17. Total and soluble aluminum concentration, diffusivity D3.

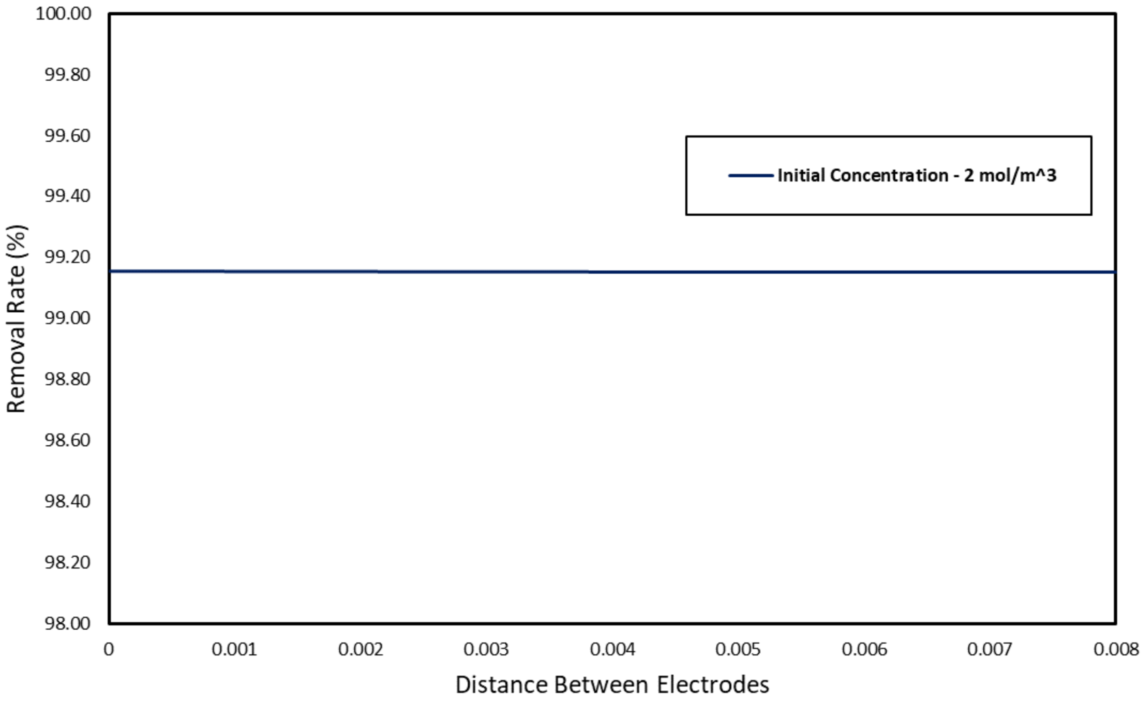

Figure 18. Removal rate of arsenic, diffusivity D3.

In Figure 13, with diffusion coefficient group D1, there is a major difference between the anode and cathode for the total aluminum concentration and a slight difference in the soluble aluminum. This is explaining that there is more aluminum concentration and generation on the anode than on the cathode. The removal rates of arsenic between the anode and cathode are not majorly different, but there is more removal on the anode as shown in Figure 14.

In Figure 15, with diffusion coefficient group D2, there is no major difference between anode and cathode in terms of total and soluble aluminum concentration generated. The removal rate of arsenic as shown in Figure 16 has a very small difference between the anode and cathode. And looking at Figure 17 with diffusion coefficient group D3, there is no difference between the anode and cathode in terms of total and soluble aluminum concentration generated. The removal rate of arsenic is also the same for both anode and cathode as shown in Figure 18.

As shown in Table 5, the average solid aluminum concentrations for the three groups are very similar. Also, comparing the average removal rates for the three groups, the results show very similar removal rate for arsenic. For this model there is no major difference in terms of removal rate of arsenic for the effect of diffusion in the EC process.

Table 5: Resulting Effect of Diffusion in Phase 2

Diffusivity Group	Total Al Concentration Avg (mol/m^3)	Soluble Al Concentration Avg (mol/m^3)	Solid Al Concentration Avg (mol/m^3)	Removal Rate of Arsenic Avg (%)
1	47.53	16.41	31.12	98.57
2	46.85	16.22	30.63	99.16
3	46.73	16.22	30.51	99.16

3.3 Outcomes of Phase 3

In order to determine the optimum operating condition for the 2D model in Phase 3, extensive study has to be conducted for a variety of parameters. Convection, current, current efficiency, and cell gap are the main factors that are monitored to see the effect on the EC process. The results show the total and soluble aluminum concentration and the removal rate of arsenic. A quadrilateral-element mesh was used for the two-dimensional EC reactor as shown below in Figure 19. The velocity profile generated is shown in Figure 20. Table 6 shows the parameters that were analyzed in the simulations. The results are shown in Figures 21-38:

Table 6: Simulated Parameters for Phase 3

Velocity (cm/s)	Current (mA)	Current Efficiency	Cell Gap (m)
0.33	20	1	0.008
0.66	10	0.5	0.004
0.132	5	0.25	0.002

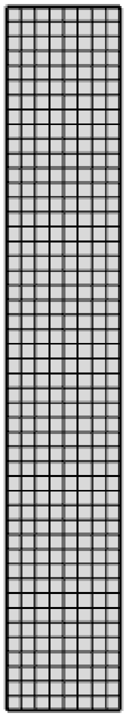

Figure 19. Quadrilateral mesh for the model.

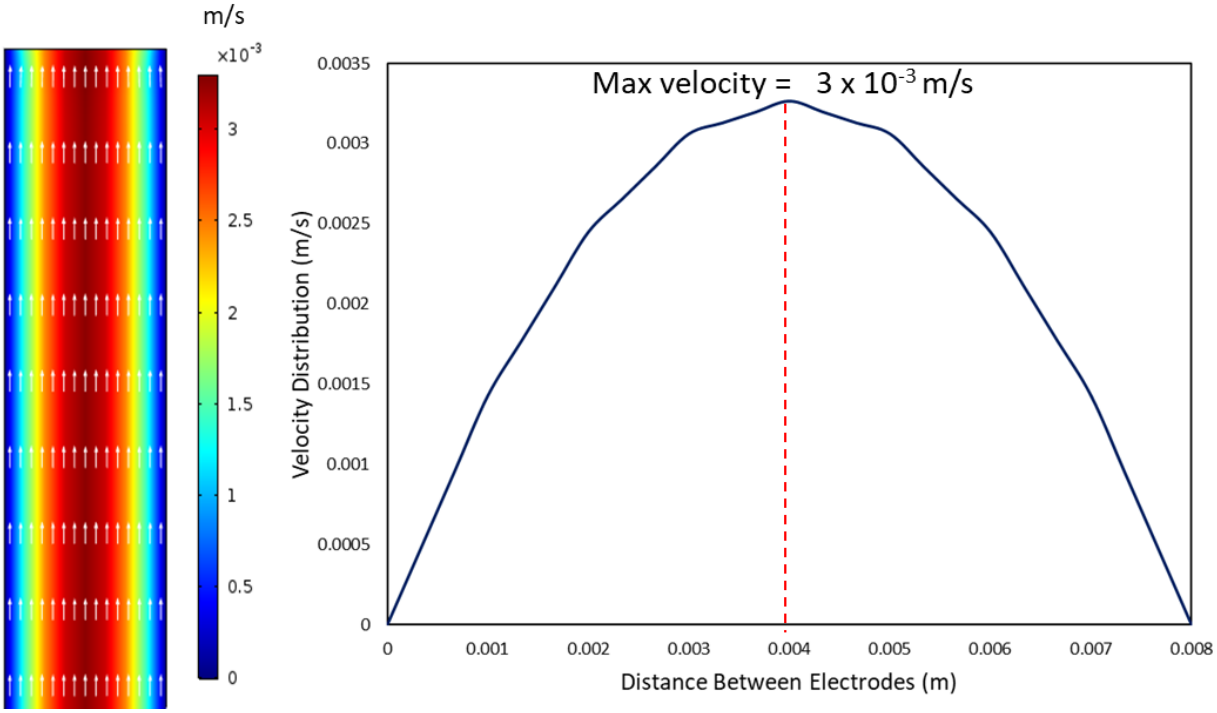

Figure 20. Velocity flow profile inside the channel and distribution at y = 2.5 cm.

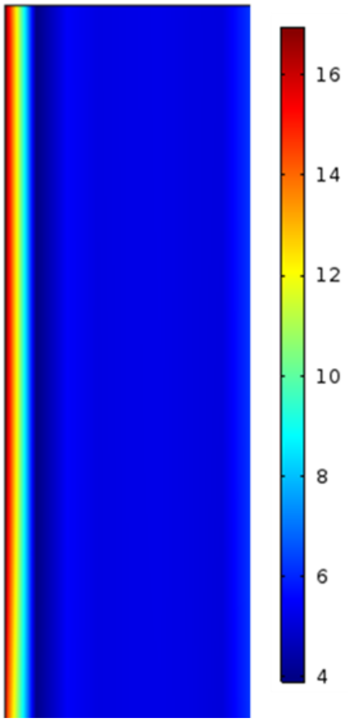

Figure 21. 2D total aluminum concentration (velocity = 0.0033 m/s, cell gap = 0.008 m, current = 20 mA, current efficiency = 1, time = 3 mins).

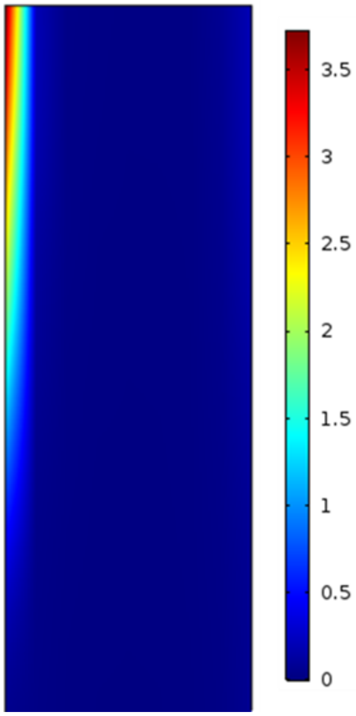

Figure 22. 2D soluble aluminum concentration (velocity = 0.0033 m/s, cell gap = 0.008 m, current = 20 mA, current efficiency = 1, time = 3 mins).

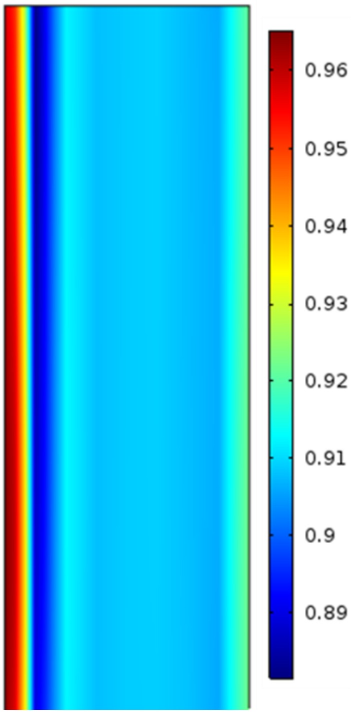

Figure 23. 2D removal rate of arsenic (velocity = 0.0033 m/s, cell gap = 0.008 m, current = 20 mA, current efficiency = 1, time = 3 mins).

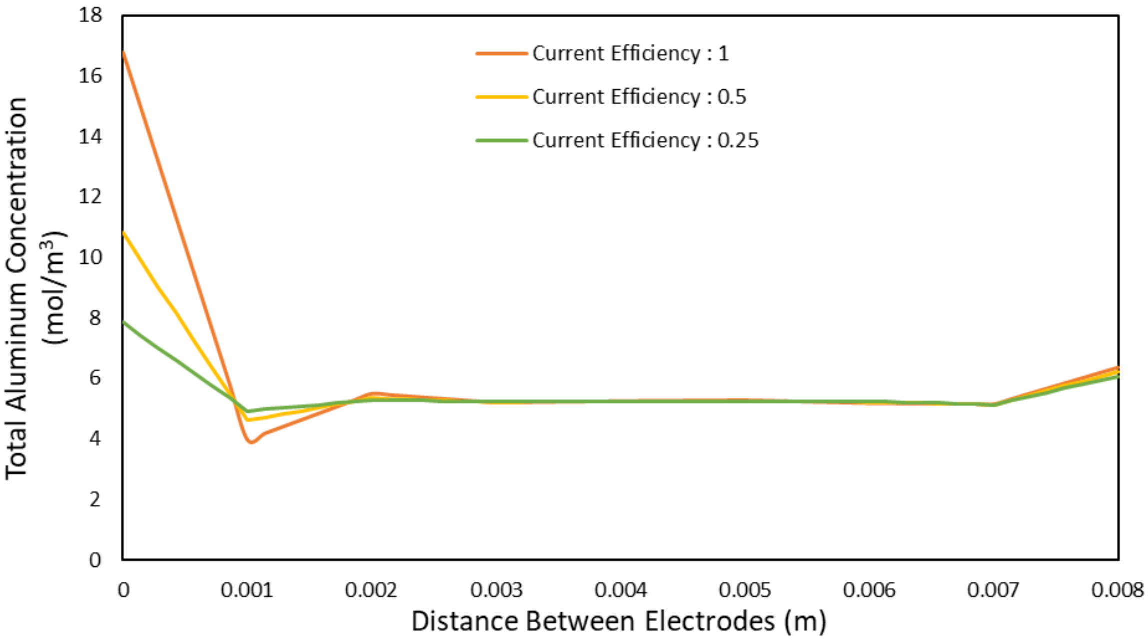

Figure 24. Total aluminum concentration at location y = 0.05m (velocity = 0.0033 m/s, cell gap = 0.008 m, current = 20 mA, current efficiency = 1, 0.5, 0.25, time = 3 mins).

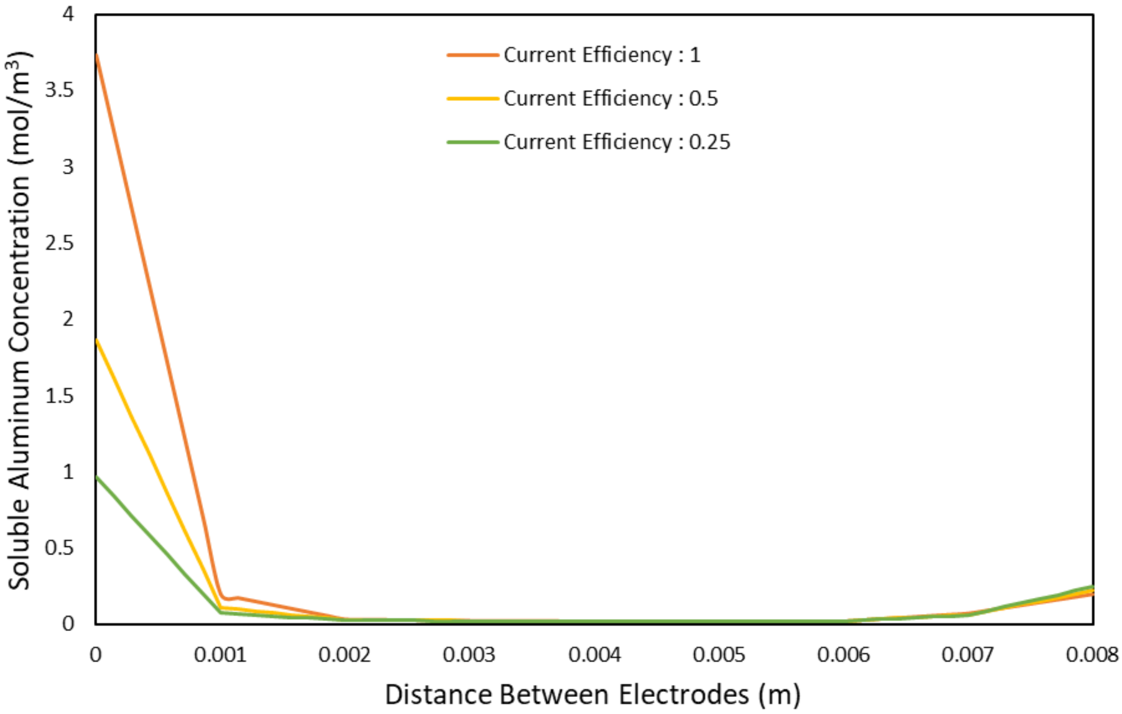

Figure 25. Soluble aluminum concentration at location y = 0.05m (velocity = 0.0033 m/s, cell gap = 0.008 m, current = 20 mA, current efficiency = 1, 0.5, 0.25, time = 3 mins).

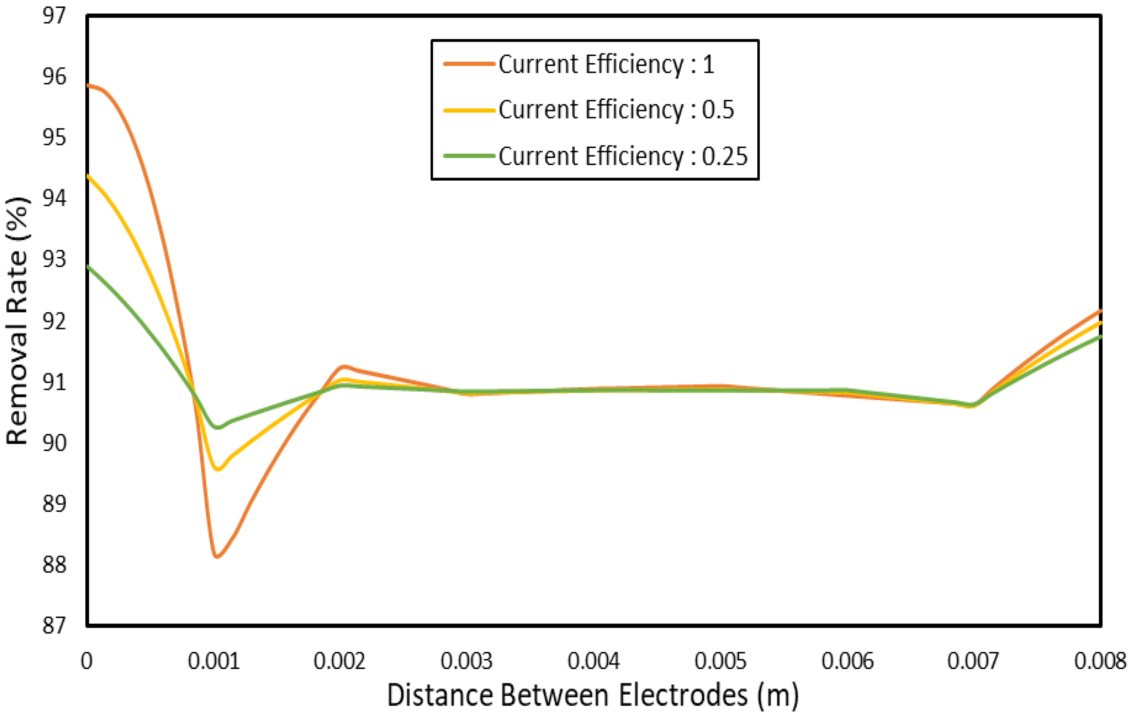

Figure 26. Removal rate of arsenic at location y = 0.05m (velocity = 0.0033 m/s, cell gap = 0.008 m, current = 20 mA, current efficiency = 1, 0.5, 0.25, time = 3 mins).

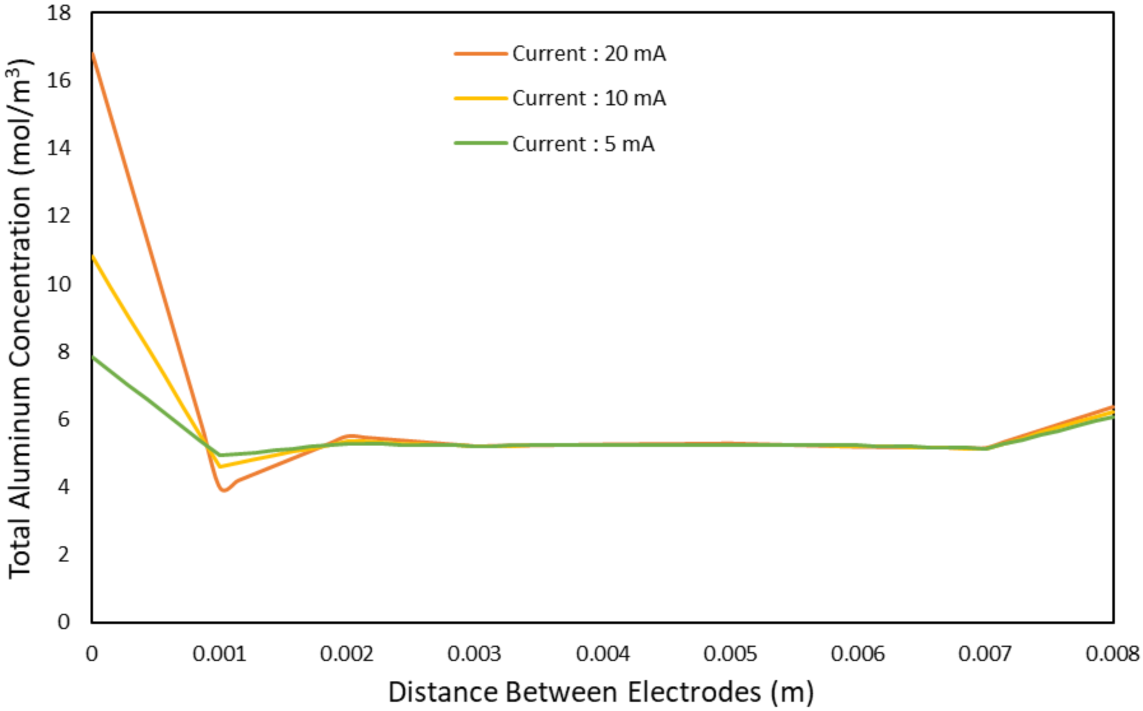

Figure 27. Total aluminum concentration at location y = 0.05m (velocity = 0.0033 m/s, cell gap = 0.008 m, current = 20 mA, 10mA, 5mA, current efficiency = 1, time = 3 mins).

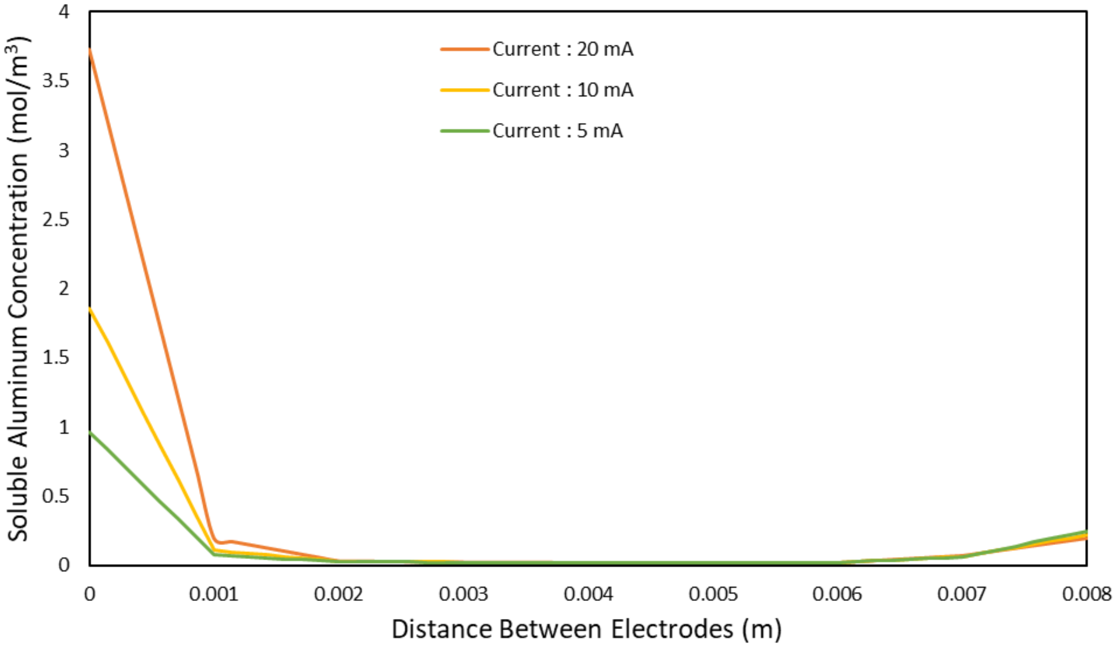

Figure 28. Soluble aluminum concentration at location y = 0.05m (velocity = 0.0033 m/s, cell gap = 0.008 m, current = 20 mA, 10mA, 5mA, current efficiency = 1, time = 3 mins).

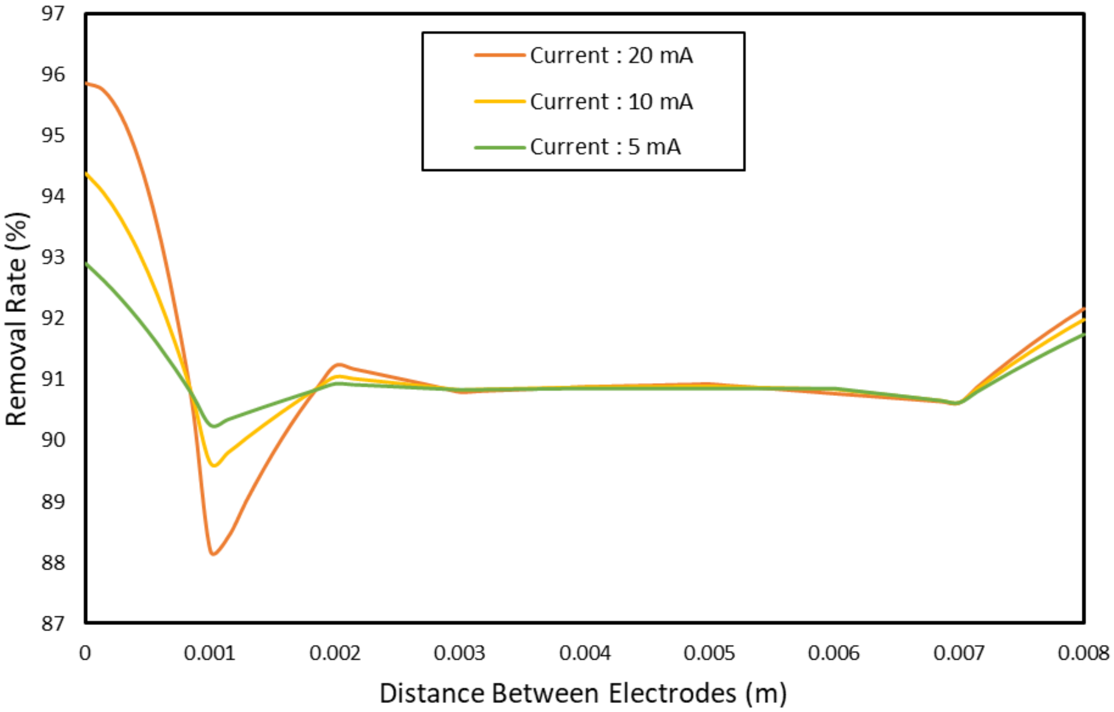

Figure 29. Removal rate of arsenic at location y = 0.05m (velocity = 0.0033 m/s, cell gap = 0.008 m, current = 20 mA, 10mA, 5mA, current efficiency = 1, time = 3 mins).

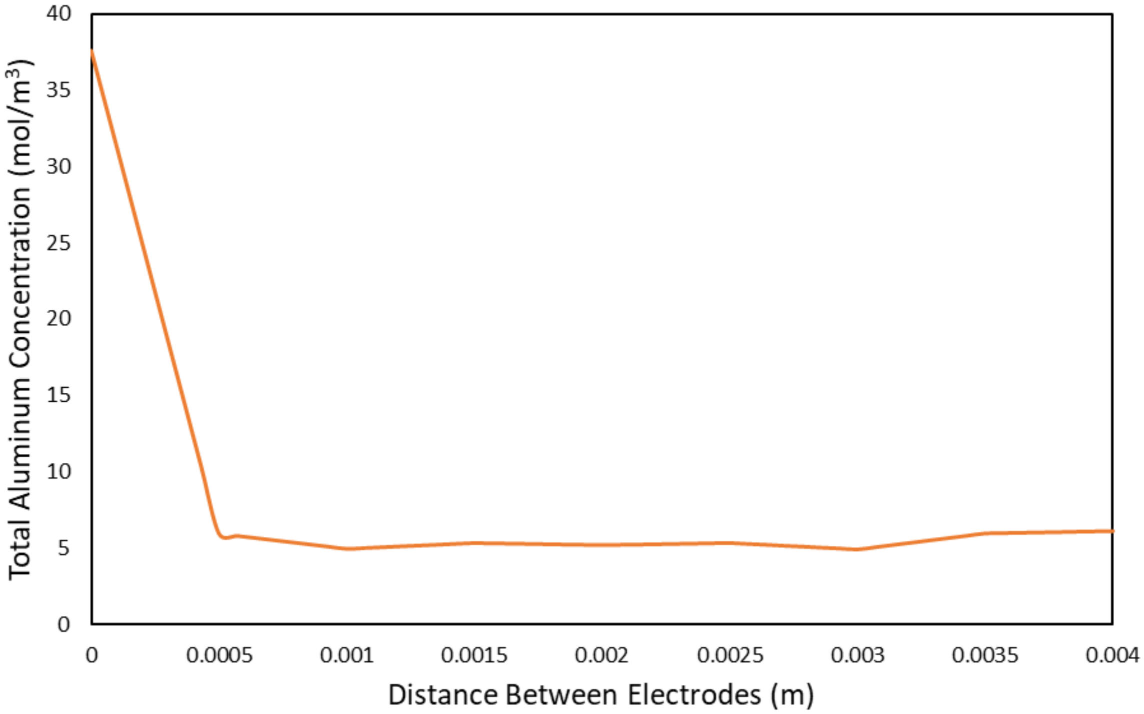

Figure 30. Total aluminum concentration at location y = 0.05m (velocity = 0.0033 m/s, cell gap = 0.004 m, current = 20 mA, current efficiency = 1, time = 3 mins).

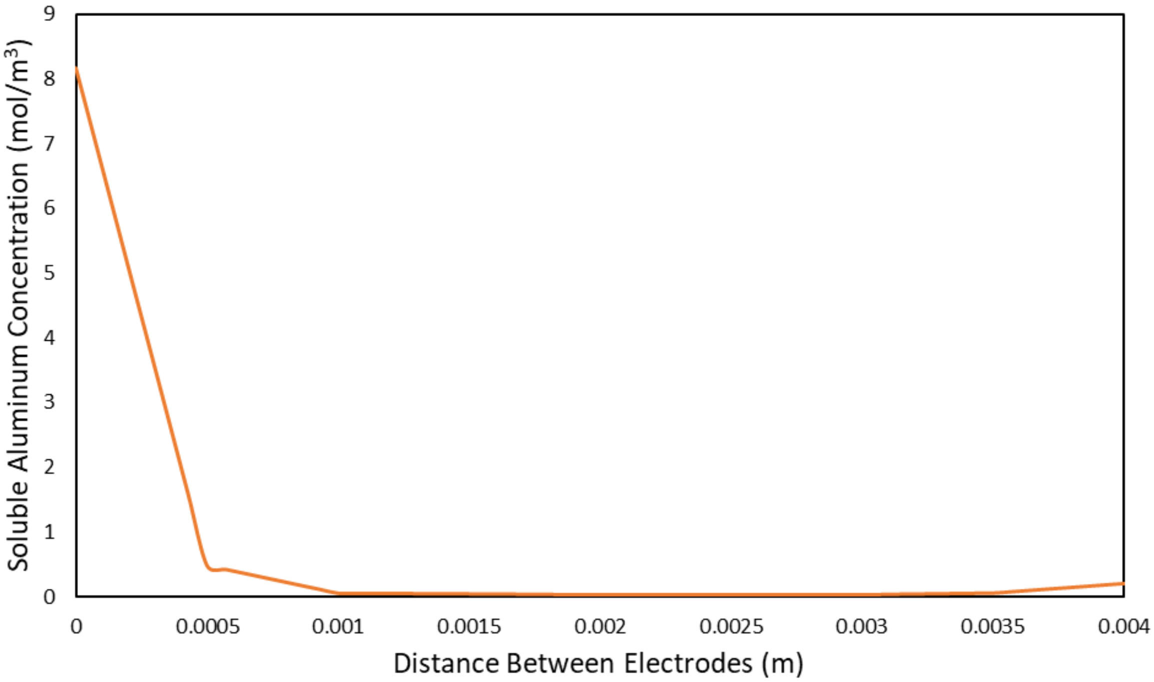

Figure 31. Soluble aluminum concentration at location y = 0.05m (velocity = 0.0033 m/s, cell gap = 0.004 m, current = 20 mA, current efficiency = 1, time = 3 mins).

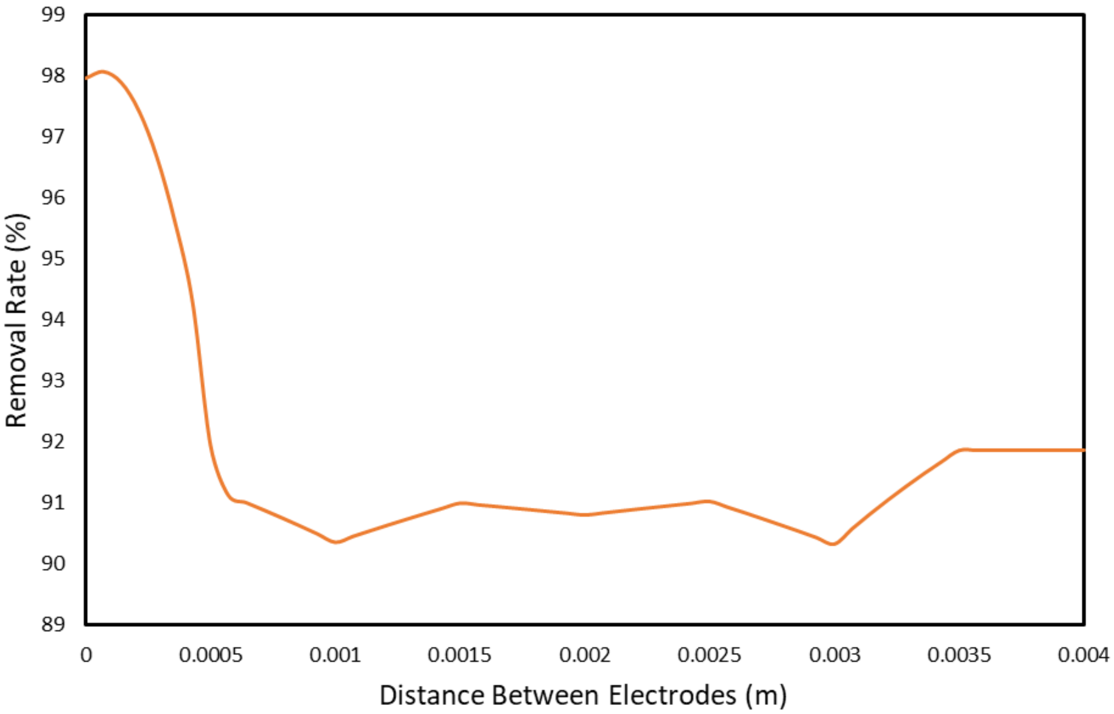

Figure 32. Removal rate of arsenic at location y = 0.05m (velocity = 0.0033 m/s, cell gap = 0.004 m, current = 20 mA, current efficiency = 1, time = 3 mins).

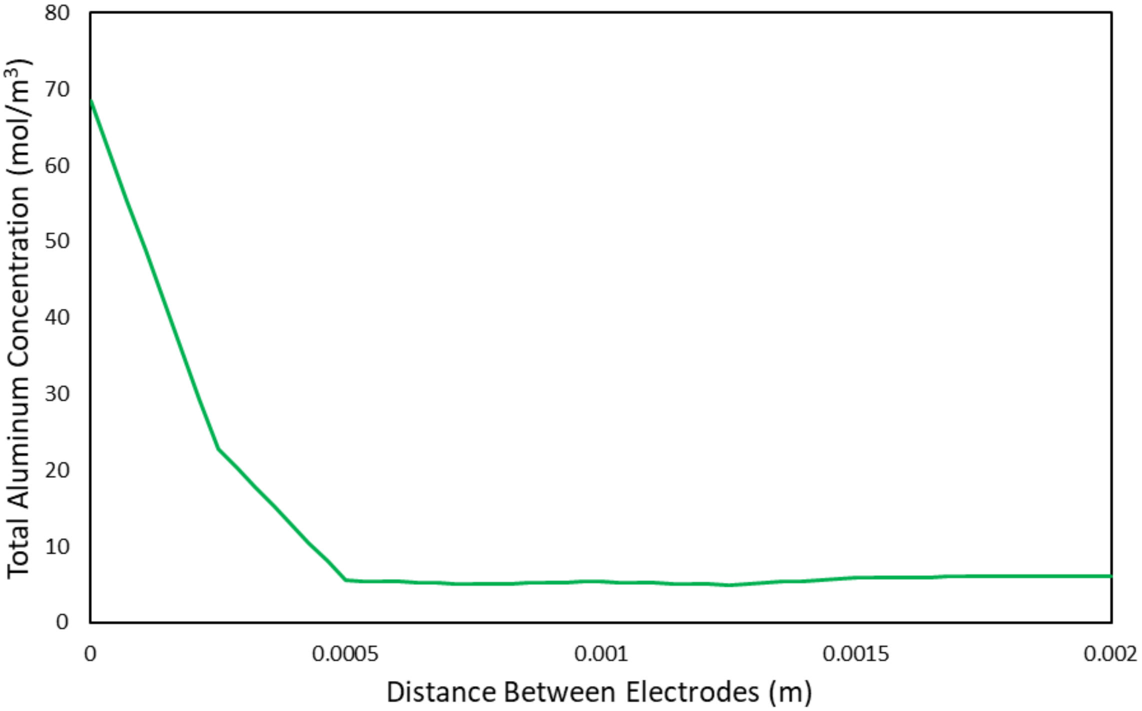

Figure 33. Total aluminum concentration at location y = 0.05m (velocity = 0.0033 m/s, cell gap = 0.002 m, current = 20 mA, current efficiency = 1, time = 3 mins).

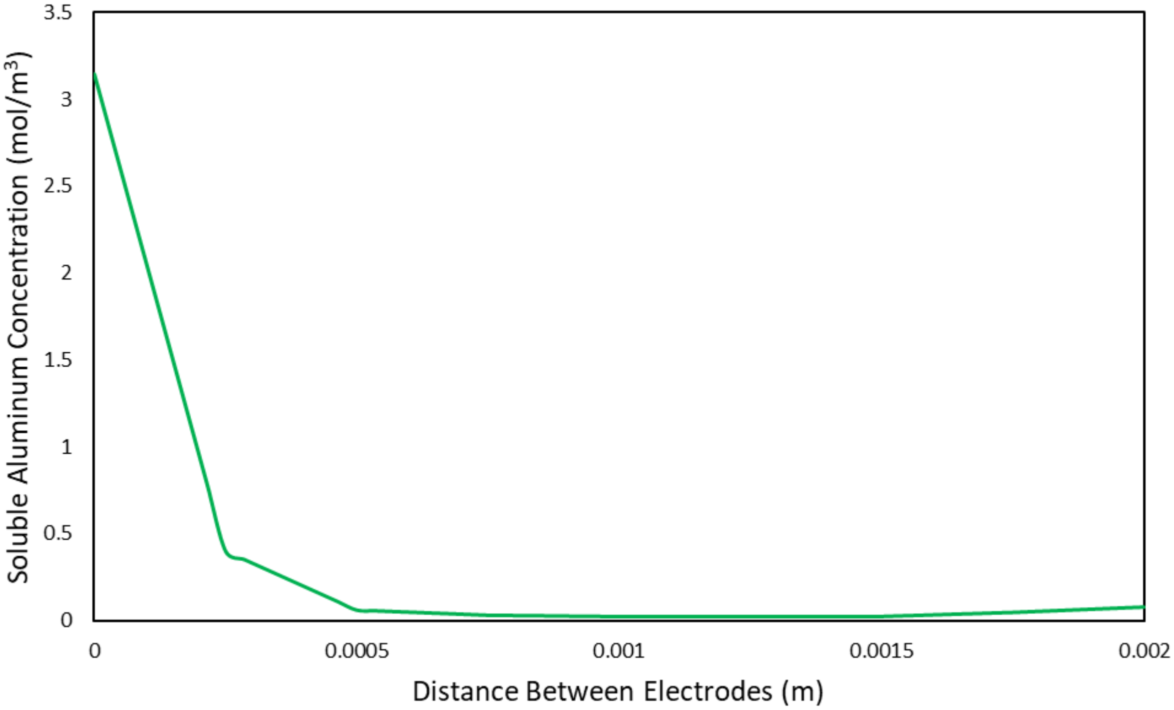

Figure 34. Soluble concentration at location y = 0.05m (velocity = 0.0033 m/s, cell gap = 0.002 m, current = 20 mA, current efficiency = 1, time = 3 mins).

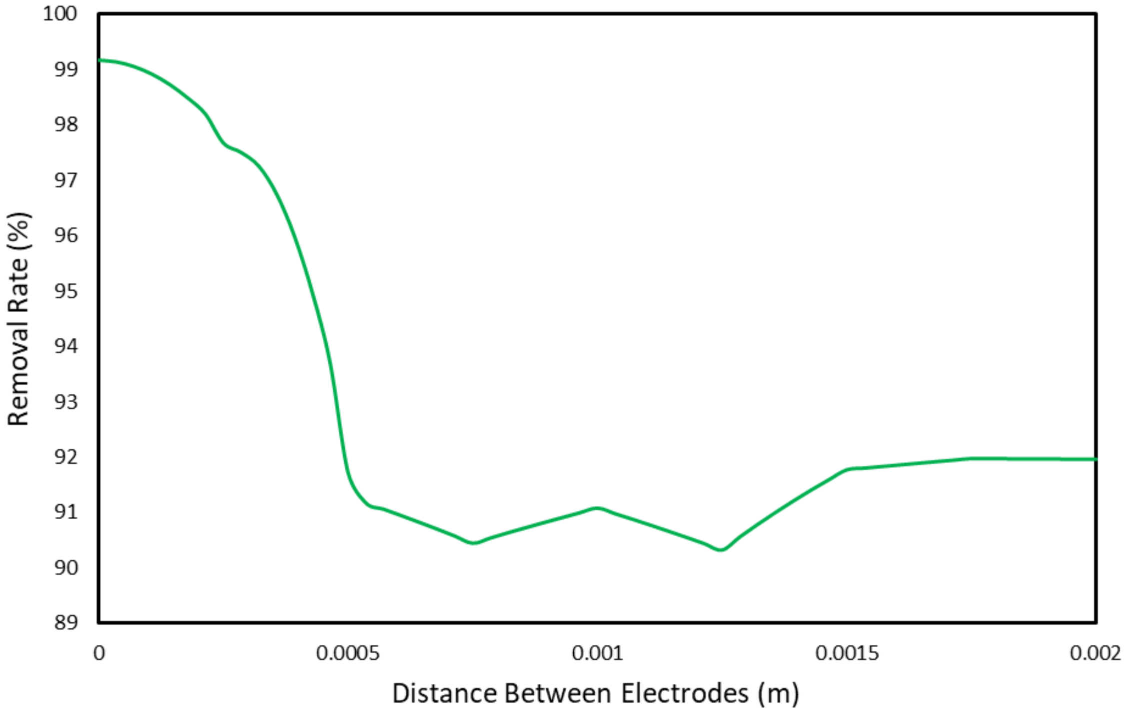

Figure 35. Removal rate of arsenic at location y = 0.05m (velocity = 0.0033 m/s, cell gap = 0.002 m, current = 20 mA, current efficiency = 1, time = 3 mins).

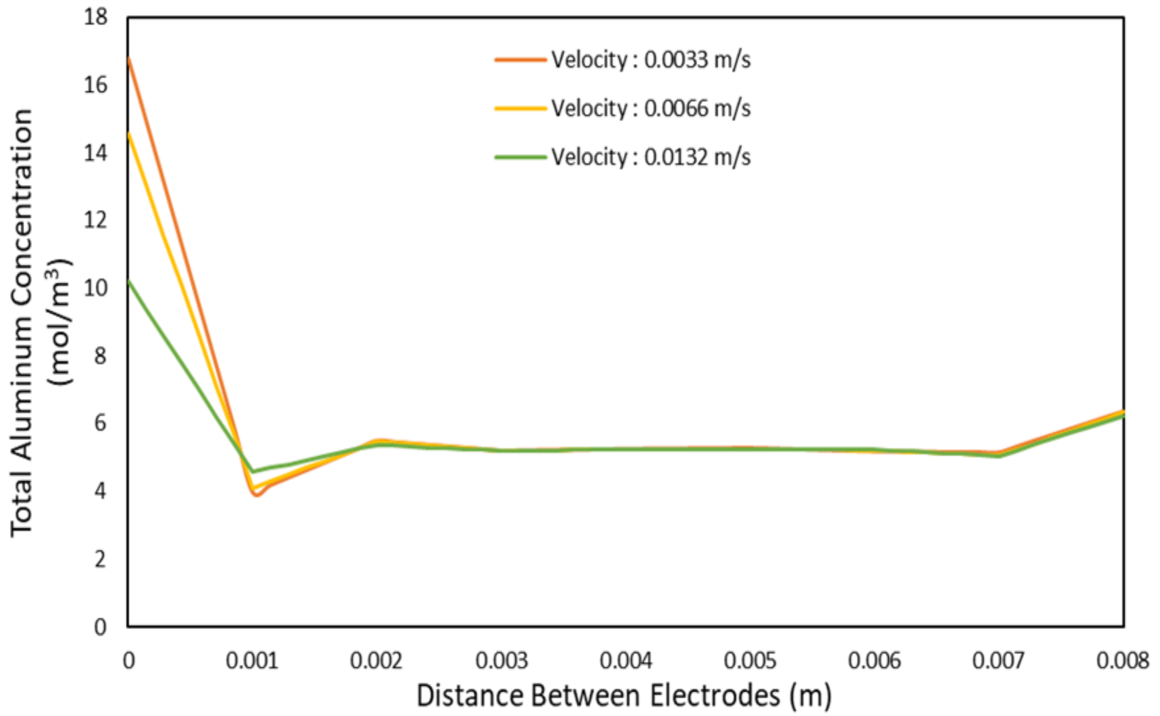

Figure 36. Total aluminum concentration at location y = 0.05m (velocity = 0.0033, 0.0066, 0.0132 m/s, cell gap = 0.008 m, current = 20 mA, current efficiency = 1, time = 3 mins).

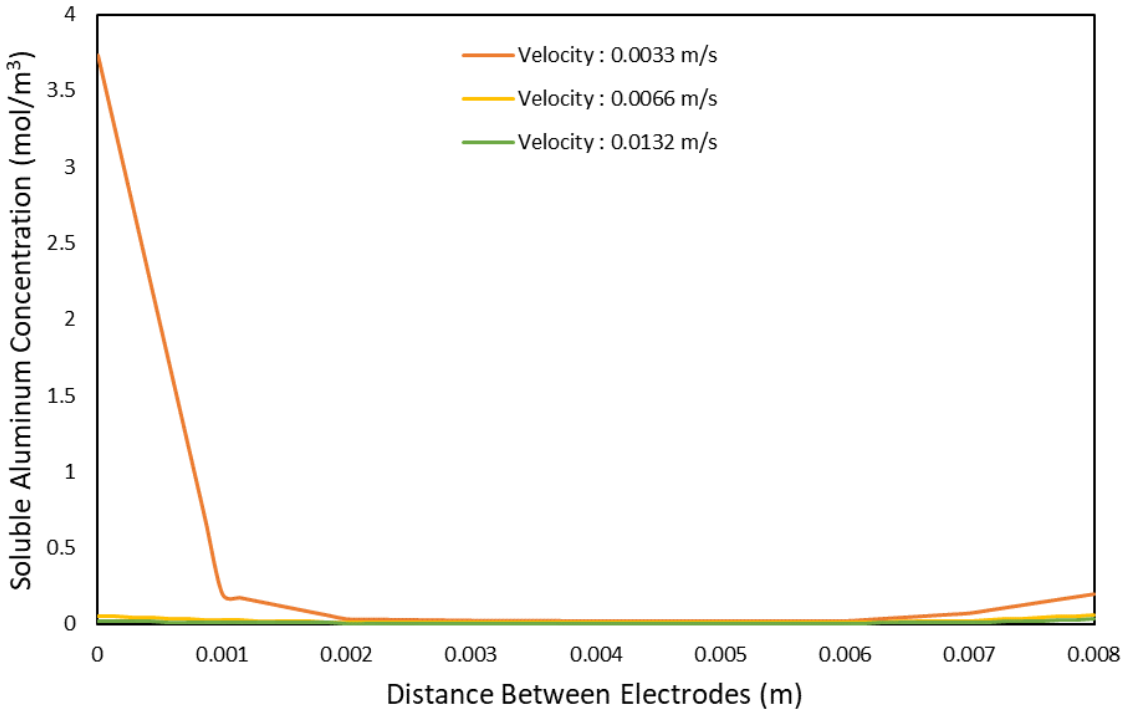

Figure 37. Soluble aluminum concentration at location y = 0.05m (velocity = 0.0033, 0.0066, 0.0132 m/s, cell gap = 0.008 m, current = 20 mA, current efficiency = 1, time = 3 mins).

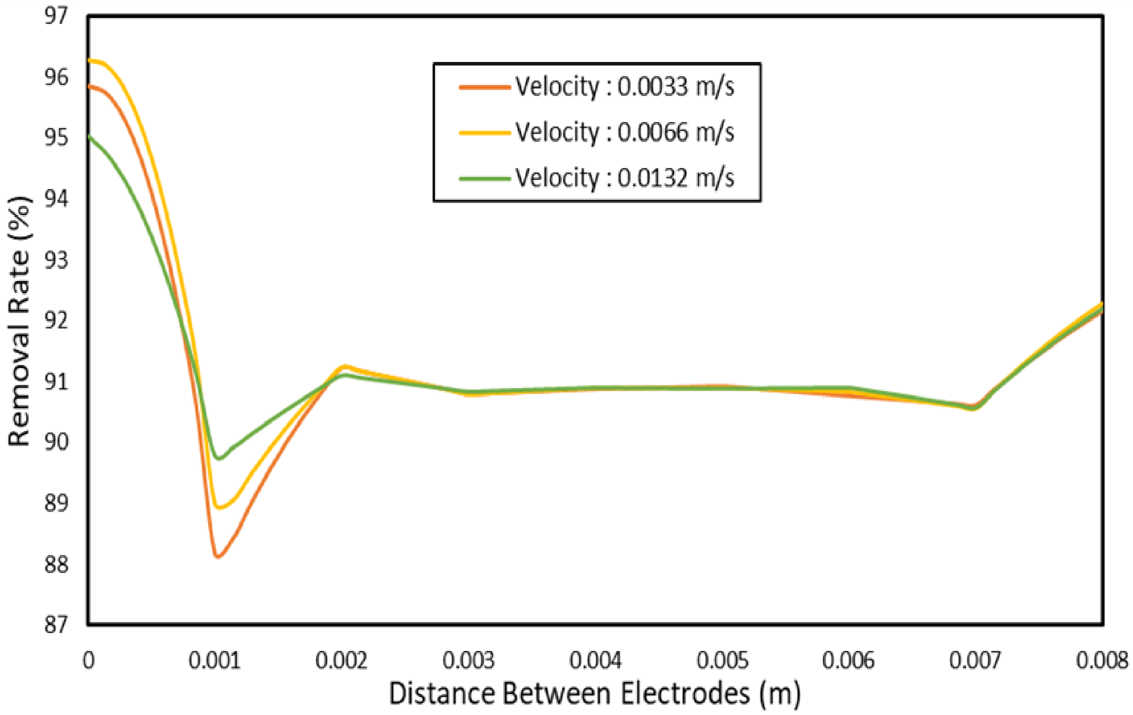

Figure 38. Removal rate of arsenic at location y = 0.05m (velocity = 0.0033, 0.0066, 0.0132 m/s, cell gap = 0.008 m, current = 20 mA, current efficiency = 1, time = 3 mins).

Figure 20 describes the velocity profile of the domain and distribution at 2.5 cm in the y-direction. The maximum velocity is noticed at the center between the electrodes. Figure 24 shows the total concentration of aluminum comparing current efficiencies (100%, 50 %, and 25 %). The current efficiency is the ratio of the actual current density in an electrochemical reaction to the total applied current density. As expected, the total aluminum concentration generated on the anode side increases with increasing current efficiency. There is more solid particle formation on the anode than cathode when looking at difference between total aluminum concentration and soluble aluminum concentration in Figure 25. For the removal rate of arsenic in Figure 26, the current efficiency of 100% generated the highest removal rate of 95.86% on the anode and an overall average of 91.17%. The same occurrence is noticed in Figure 27, Figure 28, and Figure 29 between three currents (5mA, 10 mA, and 20mA). The applied current of 20 mA produced the highest removal rate for arsenic with an average of 91.17%. This explains that for the particular system analyzed, the higher current, the higher the total aluminum concentration generated and the higher the removal rate. However, in most cases in the EC process, increase in current to a certain point will not affect the system's performance anymore and results will remain constant.

The cell gap between the anode and cathode is also considered as an influential factor in the determination of a high-efficiency EC process. A shown in Figure 30 and Figure 33, as the cell gap between the electrodes decreases, the total aluminum concentration generated on the anode increases. A high total aluminum concentration is produced by the 2-mm cell gap, which generated a higher solid particle formation and a removal rate of 99.17% on the anode and an overall average of 93%.

Comparing the velocities modeled as shown in Figure 36, the velocity of 0.33 cm/s generated a higher total aluminum concentration on the anode, but the velocity of 0.66 cm/s generated a higher solid particle formation, which resulted in a higher removal rate of 96.28% on the anode and an overall average of 91.3%. With the individual optimum parameters determined, two models were simulated to see the effect of removal rate of arsenic. The first model is Model A as shown in Table 7, which comprises each optimum design parameters for the removal rate of arsenic. Model B also shows a different design simulation. Model A produced a removal rate of 93% while Model B produced a removal rate of 91%.

Table 7: Design Parameters for High-Efficiency EC Process

Parameters / Model	Velocity (cm/s)	Current (mA)	Current Efficiency (%)	Cell Gap (mm)	Average Removal Rate of Arsenic (%)
A	0.33	20	100	2	93%
B	0.66	20	100	2	91%

CHAPTER 4: CONCLUSION AND FUTURE WORK

4.1 Conclusion

Many mathematical models were established to simulate the generation and effect of mass transfer of ionic species in EC process. A 0D unsteady-state model was simulated to see the effect of applied current and initial concentration of pollutant. The conclusions show that the 190 mA applied current generated more solid particle formation, which was used to remove the concentration of arsenic. It also showed that in the beginning stages of the removal of arsenic, the lower the pollutant concentration, the faster the removal rate.

A 1D unsteady-state model was simulated to show the effects of diffusion and it can be concluded that the change in diffusion coefficients did not have a major effect on the removal rate of arsenic. However, the total and soluble aluminum concentration between the anode and cathode became more uniform as the diffusivity increases. But since diffusion is an intrinsic component, which cannot be changed by physical means, convective flux is considered to determine the effect and optimum conditions for the EC process.

A 2D unsteady-state model was simulated to determine the optimum operating conditions for a high-efficiency EC process. The main influential factors that affect the EC process are considered for this simulation: current, velocity, current efficiency, and cell gap between the electrodes. The higher the current efficiency, the higher the total aluminum concentration, and the higher the solid particle formation, the higher the removal rate of arsenic. The shortest cell gap produced a higher solid particle formation and removal rate from the total aluminum concentration generated. It was concluded that with the design parameters shown in Table 7 a high-efficiency EC process can be achieved producing a high removal rate of 93% for arsenic. With the knowledge of the controlling parameters and the pollutant, the design of an EC system can be mathematically modeled, and results can be analyzed before any experimental operation is performed.

4.2 Future Work

In wastewater treatment, EC and chemical coagulation are considered to be fast operations for the formation of solid particles. The EC process is now being looked at as a substitute for chemical coagulation for treatment of wastewater. To increase the efficiency of the system and removal of a pollutant, turbulent mixing conditions can be considered, which allows for better mixing during the hydrolysis reaction. The use of different cell design for the multiple cells in the electrocoagulation process serve as a key to increase retention time and produce high removal of pollutant. To avoid or reduce the build-up of oxide film in the electrode, causing passivation, chloride ion can be applied to the wastewater. This paper focused on the removal of the heavy metal arsenic, but there are more sources of wastewater contaminant that affect the environment and need to be tackled. With the help of the numerical modeling of EC, the removal rate of other wastewater parameters can be determined. More importantly, other areas in wastewater such as textile, oily bilge, restaurant, petroleum refinery, biodiesel wastewater, etc., can be explored to treat the high-concentrated wastewater parameters generated in production.

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