QATAR UNIVERSITY

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AN ENHANCED ELECTROCOAGULATION PROCESS FOR THE REMOVAL OF FE AND MN FROM MUNICIPAL WASTEWATER USING DIELECTROPHORESIS (DEP)

BY

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ABSTRACT

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Title: An Enhanced Electrocoagulation Process for the Removal of Fe and Mn from Municipal Wastewater Using Dielectrophoresis (DEP)

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This study evaluates the removal efficiency of Fe and Mn from primary treated municipal wastewater using new electrodes configuration in electrocoagulation (EC) process. The used electrodes configuration generates a dielectrophoretic (DEP) force in an EC process. The effect of the electrolysis time, electrodes spacing and current density on the removal of Fe and Mn was studied. The maximum removal of Fe and Mn were obtained using operational time of 60 min, electrodes spacing of 0.5 cm and an applied current of 800 mA. Under these operating conditions and using the new electrodes configuration, the Fe and Mn removals were 96.8% and 66%, respectively. The main advantage of using the DEP induced electrode configuration was the minimal consumption of the electrodes. The new electrode configuration showed 42% less aluminum content in the reactor compared to the aluminum electrodes without DEP effect. Moreover, the energy consumption at the selected operation conditions was 4.88 kWh/m^3. Additionally, the experimental results were compared with the simulation results of the new electrodes configuration done by COMSOL software.

DEDICATION

I'm dedicating this thesis to my parents, brothers and sisters who were always supporting me to achieve what I have achieved until now. Moreover, it is dedicated to my friends who also supported me and helped me whenever I needed help.

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CHAPTER 1: INTRODUCTION

Water is considered as a mainstay element for humanity survival on earth. It is the key for the living of humans, plants and animals. Even though it's high importance, the fresh water on earth is only 3% that has 0.3% as surface water (Saeed et al., 2015). Moreover, the fresh water is being decreased since any interference with any kind of pollutants or even interference with sea water as the case of many underground aquifers, makes the water unusable by humans. In addition to the shortage of fresh water, the ratio of birth to death in this year is more than 2 (Worldometers.info, 2019) thus a huge imbalance between fresh water demand and consumption is causing a global concern (Ang, Mohammad, Hilal, & Leo, 2015). Because of this fact, researchers started to find alternatives for fresh water to protect the humanity (UNESCO, 2012). Intensive researches were going on the last decades to treat wastewaters and a lot of methods were found to be capable of treating different kinds of wastewaters. Each method has its advantages and disadvantages and each method was found to be in excellent shape of removing a specific type of wastewater while it's ineffective when it comes to treat other kinds of wastewaters.

1.1 Reuse of wastewater in Qatar

In some countries such as Qatar there is no direct source for fresh water other than ground water and desalinated water. One of the major challenges that Qatar is facing is the high cost of the desalinated water and the depletion of ground water aquifers. Qatar has a sharp increase in water demand and it's the fastest in the GCC (Jasim, Saththasivam, Loganathan, Ogunbiyi, & Sarp, 2016). Because of the rapid increase in the industrial activities, agricultural and continuous increase of population, the demand of desalinated water is being increased sharply (Saleem, Bukhari, & Akram, 2011). Therefore, researchers have started to treat wastewater with various technologies

in order to make the treated wastewater reusable (Jasim et al., 2016). Treatment of wastewater was found to be less energy consumable and thus more cost effective than desalinated water (Jasim et al., 2016). Treated wastewater are being utilized mostly by several sectors such as (Agency), 2012; Jasim et al., 2016):

1- Agricultural: livestock watering and most of the crops.
2- Environmental: non-natural wetlands.
3- Urban: fire protection and vehicle washing.
4- Impoundments: snowmaking and recreation area.
5- Industrial: produced water and gas protection.
6- Potable: direct and indirect potable reuse.
7- Groundwater recharge.

In GCC countries, agricultural sector is consuming the largest amount of water then comes the domestic and industrial sectors (El Sayed & Ayoub, 2014; Jasim et al., 2016).

1.2 Municipal wastewater treatment

The aim of treatment of wastewater is to remove the pollutants from the wastewater in order to be within the quality standards so that it can be either dumped into the environment or reused in many applications such as agricultural (Agency), 2012; Jasim et al., 2016). Municipal wastewater is known to be rich with multiple pollutants including heavy metals. Therefore, prior discharging the wastewater into any natural water body, heavy metals must be removed. Heavy metals are considered highly toxic elements that could be absorbed easily by living organisms due to the high solubility in aqueous environment. Heavy metals can be accumulated in the human body due to the contaminated water or food. The growth of heavy metals inside the human body could result in serious health disorder (Babel & Kurniawan, 2004). Heavy metals are usually removed from wastewater using conventional process such as

electrochemical removal, ion exchange and chemical precipitation. Heavy metals removal using conventional processes has some disadvantages such as production of toxic sludge, low removal rates and high energy consumption. Several advanced technologies have studied the efficiency of removing heavy metals from several kinds of wastewaters such as electrocoagulation (EC).

With the continuous improvement of life on all aspects, water demand become a major concern since water is an essential material in almost all inventions. (Abdel-Dayem et al., 2014) have stated that treatment of wastewater process is being applied for only 60% of the total wastewater volume (10.85 km^3/yr) in Arab countries. In addition to that, only one-third of the treated wastewater was reused. Increasing this low amount of reusing treated wastewater to 80% could have mitigate the scarcity of water in Arab region (Jasim et al., 2016). Therefore, to reduce the growing of water scarcity issue, treatment of wastewater was found to be an appropriate solution. Treatment process can be different for each wastewater type or treatment level based on the desired quality of the resulted water (Jasim et al., 2016). Disposal of wastewater directly into river, sea and ocean has extreme environmental impacts on aquatic life as it increases the algal growth and reduces the dissolved oxygen amount (Cerqueira & da Costa Marques, 2012; Oron et al., 2006). Therefore, it is necessary to keep improving the processes used in the treatment of wastewater and their applications to decrease the environmental issues that can be caused by wastewater disposal or handling.

Treatment of wastewater involves several unit operations that mostly includes more than one treatment category. The treatment categories are classified as biological, physical and chemical (An, Huang, Yao, & Zhao, 2017).

- Biological treatment

  In wastewater, Nutrients and biodegradation of organic matter are being

targeted by the biological treatment method by using microorganism. Biological treatment methods are classified in accordance with the amount of oxygen in the wastewater. Therefore, aerobic technique is a method that requires the existence of oxygen. On the other hand, with the absence of oxygen, anaerobic method can be carried out (Nations, 2003). Many processes are considered under the biological unit operations and examples of those are activated sludge, aerated lagoons, rotating biological contractors and trickling filters (Moussa, El-Naas, Nasser, & Al-Marri, 2017). (Bektaş, Akbulut, Inan, & Dimoglo, 2004) have mentioned that biological treatment methods can achieve a maximum of 30% removal efficiency of phosphorus. Thus, to remove the rest of phosphorus, post-treatment methods are required. Therefore, for the removal of phosphorus, biological treatment is not considered an appropriate solution.

- Chemical treatment

In chemical treatment process, adding chemicals to the wastewater are the mainstay component for the removal of dissolved and suspended contaminants. however, they are not widely used due to their additional cost and the disposal of chemicals. Adsorption, ion exchange and coagulation and flocculation are examples of chemical treatment processes (Moussa et al., 2017).

- Physical treatment

Filtration, sedimentation, screening and flotation are the most common examples of physical treatment process. Their main target is the removal of solids out of the wastewater without altering the wastewater characteristics by physical separation (Moussa et al., 2017).

Table 1. Unit Operations of the Wastewater Tratment

Wastewater treatment unit operations	
Biological treatment	1. Activated sludge
	2. Aerated lagoons
	3. Rotating biological contractors
	4. Trickling filters
Chemical treatment	1. Adsorption
	2. Ion exchange
	3. Electrocoagulation
	4. Coagulation and flocculation
Physical treatment	1. Filtration
	2. Sedimentation
	3. Screening
	4. Flotation

1.2.1 Manganese removal

In nature, manganese (Mn) is considered one of the most abundant metals. It is very important for the human body since it activates most of the enzymes. Moreover, it has many other useful applications in the industrial field such as electrical coils, dry battery cells and ceramics. It is also an alloying element that many alloys are consist of. However, taking high amounts of manganese leads to manganese psychosis which is an irreversible neurological disorder (Donaldson, 1987; Sharma, Singh, & Gode, 2007; Takeda, 2003). It is considered as impotence, uncontrollable laughter and sexual excitement (Sharma et al., 2007). It was also reported that it causes 'manganese pneumonia'. In general, most of the metals including manganese cause detrimental effects to the plants if excessive amounts were provided (Nriagu, 1988). Therefore, prior to the disposal of wastewater that contains manganese, it should be treated to eliminate the manganese content from it. Several ways have been reported that can be used for the removal of manganese from wastewater (Buamah, Petrusevski, & Schippers, 2008; Choo et al., 2007; Guan, Ni, Wu, & Lai, 2009; Li et al., 2010;

Newcombe & Dixon, 2006; Okoniewska, Lach, Kacprzak, & Neczaj, 2007; Suzuki, Watanabe, Ozawa, & Ikeda, 1998; Xu et al., 2009). The most familiar used method for the heavy metals removal from wastewaters is precipitation through alkaline solution and then coagulation by $Al_2(SO_4)_3$ or $Fe_2(SO_4)_3$ followed by sedimentation. Even though precipitation method showed reliable results in the industrial wastewater treatment, a secondary pollution might be induced due to the addition of chemical substances. As a result of those disadvantages, electrocoagulation was found to be much more efficient for the treatment of several industrial wastewater in many studies (Heidmann & Calmano, 2008).

1.2.2 Iron removal

Iron species exist in groundwater naturally in different forms such as soluble state e.g. Fe^{2+}, insoluble state Fe^{3+}, organic state and bacterial state (Chaturvedi & Dave, 2012). Furthermore, they are being formed unnaturally by many industries such as steel and mining industries (Ghosh, Solanki, & Purkait, 2008). Although iron was classified as secondary pollutant as it is unharmful material on humans, it is unwanted material; turbidity, discoloration and unpleasant taste are considered part of the aesthetic issues related to the existence of iron in any water (Phadke, 2014). In addition, the water that contains iron has high chances of bacterial growth (Leptothrix and Gallionella are examples of iron bacteria), that stops the plumbing in consequence (Chaturvedi & Dave, 2012; Ityel, 2011). Different methods are being used for the treatment of water that contains iron. The leading method among them is the aeration followed by separation which is used for the treatment of groundwater in bulky water supply systems in order to remove iron species (Degrémont, 1972; Ghosh et al., 2008). Other methods are adsorption on solids like calcium carbonate and activated carbon (Hamdouni et al., 2016; Mejri, Ben Salah, & Tlili, 2015), ion exchange (Vaaramaa &

Lehto, 2003), oxidation followed by filtration (Ellis, Bouchard, & Lantagne, 2000) and electrocoagulation (Ghosh et al., 2008; Hashim, Shaw, Al Khaddar, Pedrola, & Phipps, 2017; Vasudevan, Jayaraj, Lakshmi, & Sozhan, 2009; Vasudevan, Lakshmi, & Sozhan, 2009). In comparison between Electrocoagulation process and the other processes, EC can be easily used and installed, chemicals are not required to be added and it can be used with other methods as coupling or hybrid systems. Furthermore, it has a large treatment capacity and produces lower amounts of total dissolved solids unlike the other processes (Azadi Aghdam, Kariminia, & Safari, 2016; Ghosh, Sinha, & Purkait, 2013; Kamaraj & Vasudevan, 2015; Khandegar & Saroha, 2013).

1.2.3 Electrocoagulation

Electrocoagulation is an electro-chemical process where chemical coagulants are generated by connecting sacrificial anodes to an electric current. It combines electrochemistry, flotation and coagulation. It was used since 1889 in England, and was abandoned soon after due to its high operational cost (Sahu, Mazumdar, & Chaudhari, 2014). Extensive use of industries and new kinds of technologies have brought back the importance of utilizing the EC process since most of the pollutants can be removed by electrocoagulation. Therefore, the treatment of wastewater has become much more complex than previously anticipated (Basha, Selvi, Ramasamy, & Chellammal, 2008; Eyvaz, Kirlaroglu, Aktas, & Yuksel, 2009; Lai & Lin, 2003; Merzouk et al., 2009). In the EC process, a lot of physical and chemical phenomena take place, making it one of the more complicated processes used to treat wastewater. Generally, the electrocoagulation process contains two or more electrodes that forms anode and cathode sequence while they are connected to a power supply that is either direct current (DC) or alternative current (AC) for a certain electrolysis time. Electrocoagulation process starts with the coagulants formation, then suspended matter destabilization and

finally aggregation of particles that settle by gravity as illustrated in Figure 1 (Eyvaz, Gürbulak, Kara, & Yüksel, 2014; Uduman, Bourniquel, Danquah, & Hoadley, 2011).

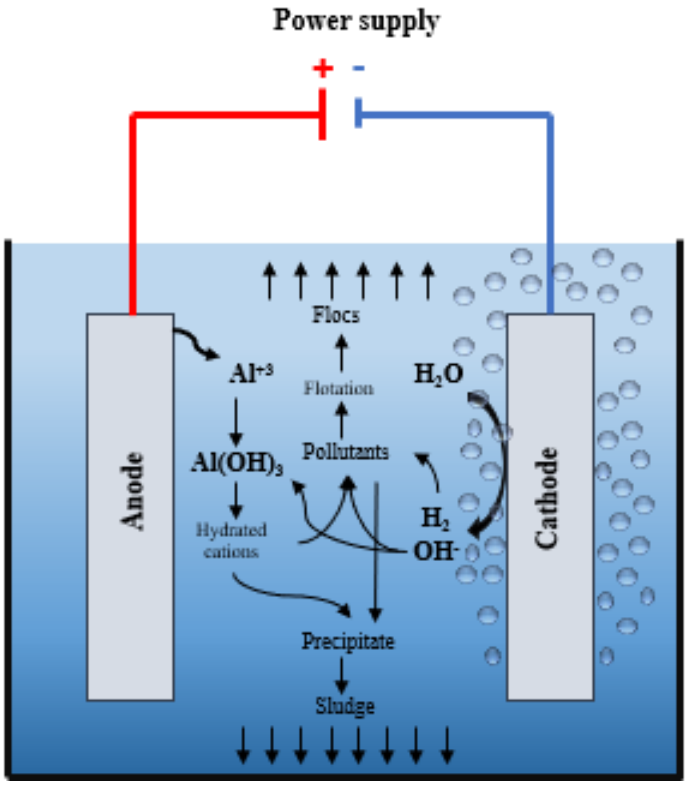


Figure 1. A schematic sketch for the electrocoagulation process.

The sacrificial anodes are usually made of either aluminum or iron (Mahmoud, Farah, & Farrag, 2013). They are widely used because of their high electrodissolution rate, low cost and availability (Feryal Akbal & Kuleyin, 2011; Izquierdo et al., 2010; Ozyonar & Karagozoglu, 2011; Un, Koparal, & Ogutveren, 2013; Vepsäläinen, 2012). (Grubb, Guimaraes, & Valencia, 2000) mentioned that the pH value of the medium controls the precipitants formed as iron and aluminum present in water such as the formation of inorganic insoluble compounds at pH values less than 6.5 while at pH

values more than 6.5, metal hydroxides will be generated. The main reactions that occurs at the aluminum electrodes are:

At anode:

$$\mathrm{Al} \rightarrow Al^{3+} + 3e^{-} \quad (1)$$

$$2H_2O \rightarrow 4H^{+}_{(aq)} + O_{2(g)} + 4e^{-} \quad (2)$$

At cathode:

$$2H_2O + 2e^{-} \rightarrow H_{2(g)} + 2(OH)^{-}_{(aq)} \quad (3)$$

Metal cations (M^{n+}) might be reduced on the cathode surface electrochemically (Heidmann & Calmano, 2008):

$$M^{n+} + ne^{-} \rightarrow nM \quad (4)$$

The production of hydroxide species at the cathode induces the metal ions precipitation by increasing the pH of the wastewater. This occurs in parallel with the precipitation of aluminum hydroxides (Heidmann & Calmano, 2008):

$$M^{n+} + nOH^{-} \rightarrow M(OH)_n \downarrow \quad (5)$$

Since the anode is made of aluminum, Al^{3+} and $Al(OH)^{2+}$ are produced by electrolytic dissolution of the electrode at low pH values. Both of the cationic monomeric species change initially to $Al(OH)_3$ then polymerize to $Al_n(OH)_{3n}$ at appropriate pH values (Das & Nandi, 2019):

$$Al^{3+}_{(aq)} + 3H_2O \rightarrow Al(OH)_3 + 3H^{+}_{(aq)} \quad (6)$$

$$nAl(OH)_3 \rightarrow Al_n(OH)_{3n} \quad (7)$$

Noting that $Al(OH)_3$ amorphous aluminum hydroxide has a bulk structure which allows it to easily settle down (Das & Nandi, 2019; Doggaz, Attour, Le Page Mostefa, Tlili, & Lapicque, 2018; Eyvaz et al., 2014; Farhadi, Aminzadeh, Torabian, Khatibikamal, & Fard, 2012). As for the settling technique, the settling velocity of the flocs is directly proportional to the flocs weight (Zodi, Potier, Lapicque, & Leclerc,

2009). Other ionic species might also be presented in the medium based on the pH values of the solution like dissolved $Al(OH)^{2+}$, $Al(OH)_4^-$, $Al_2(OH)_2^{4+}$ hydroxide complexes. The aluminum hydroxides may remove the pollutants from the wastewater by sorption, electrostatic attraction or co-precipitation followed by coagulation (Heidmann & Calmano, 2008).

1.2.4 Charge of the colloids and destabilization

Most of the colloids in wastewater have a negative surface charge (Duan & Gregory, 2003). Colloidal system stability is achieved by carrying similar charges on the colloidal particles. Therefore, they remain suspended due to repletion effect among them. However, the colloidal system net charge is neutral since the positive ions are balancing the negative colloids in the medium. Consequently, an electrical double layer is formed (Ghernaout, Naceur, & Ghernaout, 2011). At the surface of the colloidal particle lies the maximum potential. Moreover, the potential decreases while moving far from the surface out of the inner layer in which the zeta potential is measured. Zeta potential is the difference in potential between the bulk medium and the outer layer. It also determines the colloidal system stability. Hence, increasing the zeta potential, increases the colloidal system stability and the repulsive force (Duman & Tunç, 2009; Ghernaout et al., 2011).

As shown in figure 2, two layers are forming the electrical double layer which are outer layer and inner layer. The outer layer is called diffuse layer or slipping plane in which the positive ions are slackly attached while the inner layer is called Helmholtz layer or stern layer in which a tight attachment is occurring between the positive ions and the colloidal particles surface (Duman & Tunç, 2009; Ghernaout et al., 2011).

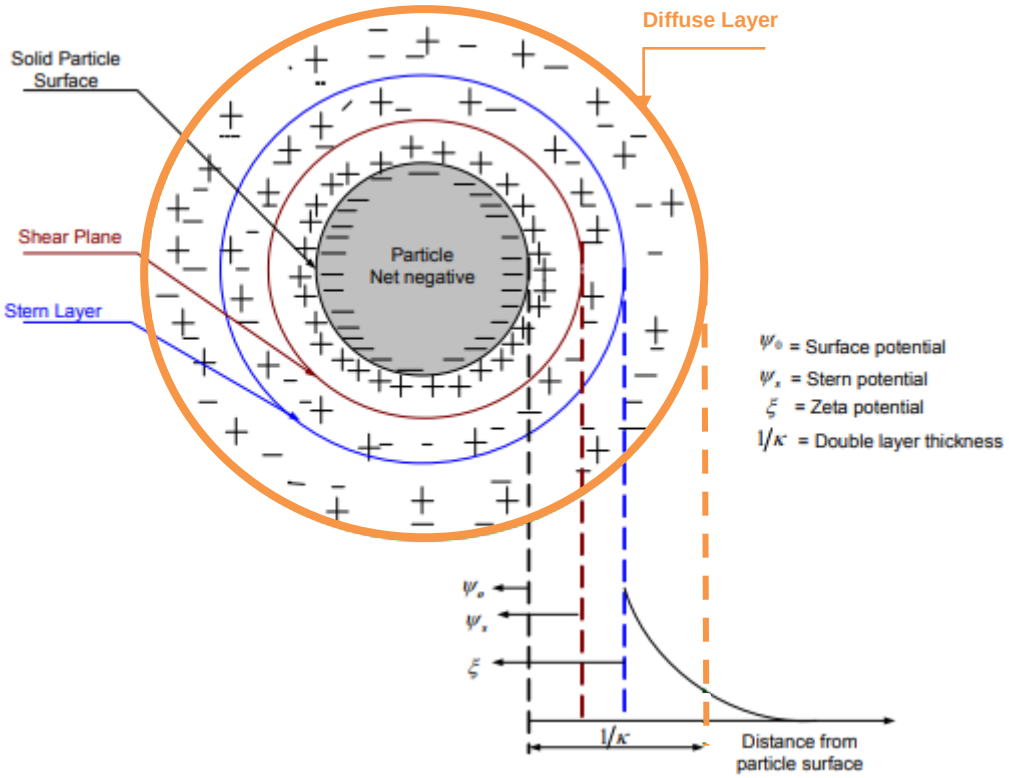


Figure 2. Illustrations of the electrical double layer (Moussa et al., 2017).

Destabilization of suspension colloids and contaminants is a consequence of the interaction between the produced aluminum ions and the presented negative colloids in the wastewater that results in trapping the negative colloids by the high concentration of positive ions. Therefore, the electrostatic repulsive force and the thickness of the electrical double layer tends to be reduced which eventually forms the flocs. The formed flocs are removed by settling down by gravity force or floating on the surface of the wastewater due to the generated hydrogen gas at the cathode (Das & Nandi, 2019; Doggaz et al., 2018; Hashim et al., 2017). Leaving the water in the middle on the container cleaner.

Although electrocoagulation has shown high removal efficiencies of many contaminants, the electrode and energy consumption of EC needs to be improved

further. Recently, there is an increased attention to dielectrophoretic force (DEP) enhanced electrocoagulation (Alkhatib, Hawari, Hafiz, & Benamor, 2020). It was found that inducing DEP force into EC process has enhanced the removal of different kinds of pollutants from different types of wastewater. In addition, inducing DEP force into EC process decreased the energy consumption.

1.3 Dielectrophoretic force

Dielectrophoretic (DEP) force is the motion of free particles in an inhomogeneous electric field by dielectric polarization (Fei Du et al., 2018; Hawari, Du, Baune, & Thöming, 2015; Monfared, Sheikhi, Kasiri, & Mohammadi, 2018). The neutral particles get polarized in an inhomogeneous electric field by Superimposing them. neutral particle makes the particle polarized. Therefore, dipole moment is induced due to the polarization with equal but opposite charges at both sides of the particle. As a result of the difference between the resulting forces on the particles sides and the local electric field in a non-uniform electric field, a net force is generated, and it is known as dielectrophoretic force (FDEP) (Çetin & Li, 2011; Hawari et al., 2015; Monfared et al., 2018). Equation 8 represents the dielectrophoretic force (FDEP) in the case of spherical particle with radius r in a medium that has ε_M relative permittivity (Hawari et al., 2015; Monfared et al., 2018).

$$F_{DEP} = 4\pi r^3 \varepsilon_0 \varepsilon_M \mathrm{Re}\left[\widetilde{K}\right](E \cdot \nabla)E \qquad (8)$$

In which ε_0 is the free space permittivity with a constant value of 8.854×10^{-12} (F/m), $\mathrm{Re}\left[\widetilde{K}\right]$ is the real part of Clausius-Mossotti factor with a value between (-5 to 1) and E is the electric field strength (V/m). Equation 9 is the calculation of $\mathrm{Re}\left[\widetilde{K}\right]$ factor and equation 10 is the calculation of complex permittivity ($\tilde{\varepsilon}$) that should be used to calculate the particle and medium complex permittivity (Hawari et al., 2015; Molla, Masliyah, & Bhattacharjee, 2005; Monfared et al., 2018).

$$\widetilde{K} = \frac{\widetilde{\varepsilon_p} - \widetilde{\varepsilon_M}}{\widetilde{\varepsilon_p} + 2\widetilde{\varepsilon_M}} \quad (9)$$

$$\tilde{\varepsilon} = \varepsilon - \frac{j\sigma}{\omega} \quad (10)$$

In which $\widetilde{\varepsilon_p}$ is the particle complex permittivity, $\widetilde{\varepsilon_M}$ is the suspending medium complex permittivity, ε is the permittivity, $j = \sqrt{-1} \cdot (E \cdot \nabla)E = \frac{1}{2}\nabla|E|^2$ is the geometrical gradient related to the square of the field intensity (E) (Fei Du et al., 2018), ω is the angular frequency (rad/s) and σ is the conductivity (s/m).

Different DEP effects might be presented by the suspended particles where it can move in several directions based on the particle and medium permittivity. These effects are presented as negative DEP (nDEP) and positive DEP (pDEP). nDEP is presented when the particles are attracted by the weaker electric field due to the low permittivity of the particles than the medium. Whereas, the strong electric field attracts the particles that have higher permittivity than the medium and it is called pDEP (F Du, Hawari, Baune, & Thöming, 2009; Hawari et al., 2015; Molla et al., 2005).

CHAPTER 2: ELECTROCOAGULATION PREVIOUS INVESTIGATIONS

Barisci et al. (2016) have experimented the effect of eight different electrodes arrangement to treat greywater from COD that has an initial concentration of 229 mgO_2/L. They have used 4 electrodes that are made of aluminum and iron. The optimum removal of COD was 98% using Al-Fe-Fe-Al electrodes in monopolar parallel (MP-P) arrangement that are connected to DC power supply that generates a current density of 1 mA/cm^2 applied for 30 mins (Barışçı & Turkay, 2016). Kobya et al. (2015) have used EC process to treat can manufacturing wastewater using 4 aluminum electrodes spaced by 1.3 cm and connected to a DC power supply. The optimum run was found to be when following MP-P electrodes arrangement, running time of 40 mins and current density of 2 mA/cm^2. At the optimum run the removal efficiencies of aluminum, zirconium, phosphate, COD and TOC are 99.41%, 99.38%, 99.80%, 72% and 37% from initial concentrations of 125.1 mg/L, 81.2 mg/L, 32.1 mg/L, 850 mg/L and 300 mg/L respectively (Kobya & Demirbas, 2015). Akbal et al. (2011) studied the removal of Cu, Cr and Ni from metal plating wastewater. The highest removal efficiencies of these metals were 100% of Cu, 100% of Cr and 100% of Ni with initial concentrations of 45 mg/L, 44.5 mg/L and 394 mg/L of Cu, Cr and Ni respectively. They were achieved by using 6 electrodes with iron at the anode and aluminum at the cathode arranged in monopolar parallel (MP-P) configuration and connected to DC power supply. The electrolysis time, current density and electrodes spacing were 20 mins, 10 mA/cm^2 and 1cm respectively (F Akbal & Camci, 2011). Al Aji et al. (2012) investigated the efficiency of EC in removing heavy metals from model water using 6 iron electrodes. They have targeted the removal of Mn, Zn, Cu and Ni. At optimum condition, the initial concentrations were 250 mg/L of Mn, Zn, Cu and Ni, electrolysis time 50 min, current density 25 mA/cm^2 generated from DC power supply

and electrodes spacing 0.3 cm. The outcome was 72.6%, 96%, 96% and 96% removal efficiency of Mn, Zn, Cu and Ni (Al Aji, Yavuz, & Koparal, 2012). Shafaei et al. (2011) mentioned that 39.6% removal efficiency of Mn was achieved from synthetic solution by using 2 aluminum electrodes connected to DC power supply in EC process at initial concentration of 22.5 mg/L of Mn. The operating conditions that have resulted in this removal were, 60 min of electrolysis time, 6.2 mA/cm^2 of current density and 1 cm of electrodes spacing (Shafaei, Rezaie, & Nikazar, 2011). Hanay et al. (2011) have proved the efficiency of using electrocoagulation for the treatment of synthetic wastewater. They have targeted the removal of Cu, Zn and Mn. By using 4 aluminum electrodes arranged in monopolar mode, electrolysis time of 35 min, current density of 15 mA/cm^2, electrodes spacing of 0.5 cm and DC power supply, the removal of Cu, Zn and Mn were 100%, 100% and 80-85% respectively (Hanay & Hasar, 2011). Shafaei et al. (2010) studied the effect of aluminum electrocoagulation on the removal of Mn from synthetic wastewater. at initial concentration of 100 mg/L, electrolysis time of 30 min, current density of 6.2 mA/cm^2 generated by DC power supply, electrodes spacing of 1 cm and by using 2 aluminum electrodes, the removal efficiency of Mn was 75% (Shafaei, Rezayee, Arami, & Nikazar, 2010). Das et al. (2020) used 2 aluminum electrodes connected to DC power supply in EC process for the removal of Fe (II) and F from drinking water. The removal efficiencies were 100% and 96% of Fe (II) and F respectively. These removal were occurred at the optimum run where they have used operational time of 60 min, current density of 4.31 mA/cm^2 and electrodes spacing of 1 cm (Das & Nandi, 2020). Doggaz et al. (2018) studied the removal of iron from groundwater using 2 aluminum electrodes in EC process. By running the process for 20 min while using current density of 10 mA/cm^2 generated from DC power supply and electrodes spacing of 1cm, the removal efficiency of iron was more than 97% when the

initial concentration was 25 mg/L (Doggaz et al., 2018). Hashem et al. (2017) investigated the impact of 4 aluminum electrodes in EC process for the removal of iron from drinking water. They have used 20 mg/L of initial concentration of Fe. As for the operation condition they have operated EC for 20 min, used 1.5 mA/cm^2 of current density and 0.5 cm of electrodes spacing. Hence, the obtained removal efficiency of iron was 98.5% (Hashim et al., 2017). Das et al. (2019) experimented the removal of iron from drinking water using 2 aluminum electrodes in EC process. They have reached 98.6% removal efficiency of iron at the optimum condition. The optimal run was achieved at the following parameter: 20 mg/L of initial concentration of iron, 45 min of electrolysis time, 20 mA/cm^2 of current density, 1 cm of electrodes spacing and using DC power supply (Das & Nandi, 2019). Gatsios et al. (2015) have investigated the removal of toxic metals from industrial wastewater using EC process. At optimum condition, they have used 2 iron electrodes spaced by 2 cm and connected to DC power supply. Applying a current density of 14 mA/cm^2 for 90 mins has resulted in removal 100% of Cu, 89% of Mn and 100% of Zn from the initial concentrations which were 5 mg/L of Cu, 5 mg/L of Mn and 10mg/L of Zn (Gatsios, Hahladakis, & Gidarakos, 2015). Table 2 summarizes the previous mentioned work on electrocoagulation.

Table 2. Summary of the Previous Studies on Electrocoagulation.

Feed water	Electrodes no	Electrodes material	Initial concentration (mg/L)	Electrolysis time (min)	Current density (mA/cm^2)	Electrodes spacing (cm)	Removal %	Reference
Greywater	2 sets	Anode: Al Cathode: Al	COD: 229 (mgO_2/L)	30	1	-	98%	(Barışçı & Turkay, 2016)
Can manufacturing	2 sets	Anode: Al Cathode: Al	Al: 125.1 Zr: 81.2 P: 32.1 COD: 850 TOC: 300	40	2	1.3	Al: 99.41% Zr: 99.38% P: 99.8% COD: 72.0% TOC: 37.0%	(Kobya & Demirbas, 2015)
Metal plating	3 sets	Anode: Fe Cathode: Al	Cu: 45 Cr: 44.5 Ni: 394	20	10	1	Cu: 100% Cr: 100% Ni: 100%	(F Akbal & Camci, 2011)
Model	3 sets	Anode: Fe Cathode: Fe	Mn:250 Zn:250 Cu:250 Ni: 250	50	25	0.3	Mn: 72.6% Zn: 96% Cu: 96% Ni: 96%	(Al Aji et al., 2012)
Industrial	1 set	Anode: Al Cathode: Al	Mn: 22.5	60	6.2	1	Mn: 39.6%	(Shafaei, Rezaie, et al., 2011)
Synthetic solution	2 sets	Anode: Al Cathode: Al	Cu: 50-200 Zn: 50-200 Mn: 50-200	35	15	0.5	Cu:100% Zn: 100% Mn: 80-85%	(Hanay & Hasar, 2011)
Synthetic solution	1 set	Anode: Al Cathode: Al	Mn: 100	30	6.2	1	Mn: 75%	(Shafaei et al., 2010)

Feed water	Electrodes no	Electrodes material	Initial concentration (mg/L)	Electrolysis time (min)	Current density (mA/cm^2)	Electrodes spacing (cm)	Removal %	Reference
Drinking water	1 set	Anode: Al Cathode: Al	Fe (II): 20 F: 10	60	4.31	1	Fe (II): 100% F: 96%	(Das & Nandi, 2020)
Groundwater	1 set	Anode: Al Cathode: Al	Fe (II): 25	20	10	1	Fe (II): >97%	(Doggaz et al., 2018)
Drinking water	2 sets	Anode: Al Cathode: Al	Fe (II): 20	20	1.5	0.5	Fe (II): 98.5%	(Hashim et al., 2017)
Drinking water	1 set	Anode: Al Cathode: Al	Fe (II): 20	45	2	1	Fe (II):98.6%	(Das & Nandi, 2019)
Industrial	1 set	Anode: Fe Cathode: Fe	Cu: 5 Mn: 5 Zn: 10	90	14	2	Cu: 100% Mn: 89% Zn: 100%	(Gatsios et al., 2015)

Recently, electrocoagulation has been studied in hybrid system as pretreatment process prior to another process and it was found that it has enhanced the whole system such as enhances the water flux in membrane-based processes. Meanwhile, the other process in the hybrid system was found to be efficient in mitigating the disadvantages of EC process such as reducing operational cost, further removal of pollutants and removal of electrodes metal species from the solution. Al Hawli et al. (2019) have studied the effect of coupling EC with FO to treat produced water. In the EC process, 2 aluminum electrodes connected to DC power supply were used. The removal efficiencies of EC alone were 97%, 91.6%, 5.4%, 91.3% and 97.4% of TSS, turbidity, conductivity, TOC and Oil& Grease respectively, using 10min of electrolysis time and 10mA/cm^2 of current density. They have concluded that coupling EC with FO has further enhanced the removal efficiencies and they were 99%, 98% and 16% of TSS, Turbidity and conductivity respectively (Al Hawli, Benamor, & Hawari, 2019). Sardari et al. (2018) have studied the impact of coupling EC with membrane distillation for the treatment of high salinity hydraulic fracturing produced water (HFPW). They have used 5 aluminum electrodes separated by 0.5 cm and connected to DC power supply. Using current density of 16.5 mA/cm^2 and operational time of 30 s, 91%, 96% and 61% removal efficiencies of TSS, turbidity and TOC respectively were achieved by EC process alone. Moreover, it was mentioned that using EC as pretreatment process has enhanced FO process by 130%. Also, an increase of 21% of water recovery rate was reported for the 24 hours experiment, which reduces the costs of reusing FO and can compensate the use of EC while obtaining better results. (Sardari, Fyfe, Lincicome, & Ranil Wickramasinghe, 2018). Dia et al. (2018) studied the impact of using a hybrid system that consisted of EC and biofiltration (BF) on the treatment of landfill leachates. Electrocoagulation was consisted of hollow cylindrical rod of stainless steel at the

cathode and fully cylindrical rod of pure aluminum at the anode, spaced by 1.55 cm and connected to a DC power supply. The optimum parameters of EC were current density of 8 mA/cm^2 and electrolysis time of 20 min. Electrocoagulation process has obtained removal efficiencies of 37±2%, 15±0%, 60±13%, 82±2.7%, 82±5.5% 95±2.3&, 95±2.6% and -26±23% of COD, TOC, color, turbidity phosphorus, zinc, iron and aluminum respectively. However, after conducting BF process on the pretreated water from EC, the find removal efficiencies were 42±7%, 49±3.3%, -34±26%, 74±15%, -34±87%, -796±425%, 14±63% and 66±27% and 97% of COD, TOC, Color, turbidity, phosphorus, zinc, iron, aluminum and BOD_5 respectively. In addition, they have concluded that the hybrid system can be used to treat the landfill leachates knowing that the cost is lower than most of the other processes with higher removal efficiencies (Dia, Drogui, Buelna, & Dubé, 2018). Hakizimana et al. (2016) illustrated the influence of using electrocoagulation as pretreatment process for reverse osmosis (RO) technique. It was mentioned that using EC as pretreatment process can further reduce the membrane fouling and biofouling of membrane-based desalination processes such as RO. Moreover, EC can replace the ordinary processes which most of the time are being used to pretreat the water prior to the use of reverse osmosis such as chlorination and chemical coagulation. In EC method, two aluminum electrodes and DC-power supply were used. At current density of 5.6 mA cm^{-2}, electrodes spacing of 1cm and running time of 30 min, the removal efficiencies were 57.5 %, 81 % and 10 % of DOC, absorbance and hardness respectively. It was stated that EC has removed the majority of microbial cells, thus it is considered in perfect shape as disinfection process (Hakizimana et al., 2016). Sardari et al. (2018) stated that, treating poultry processing wastewater (PPW) by ultrafiltration only will have a decline in the water flux due to the high amount of fats, oil and grease and total suspended solids (TSS). Therefore,

they have investigated the efficiency of EC prior to ultrafiltration since EC is famous in removing TSS and oil and grease pollutants. In the EC process, 5 aluminum electrodes were used spaced by 9mm, arranged in bipolar series (BP-S) mode and connected to a DC power supply to generate a current density of 0.3 mA/cm^2. At operational time of 5 min and using only EC method, the removal efficiencies were 87%, 94%, 59% and 84% of BOD, FOG, COD and TSS respectively. It was found that by using EC as pretreatment method, the initial flux was 106 $L/(m^2h)$ and dropped to 46 $L/(m^2h)$ in the first day which had reduced to 22 $L/(m^2h)$ after seven days, while in the normal case of using UF alone, the flux reduced by 91% in the first day and dropped to zero after 3 days (Sardari, Askegaard, et al., 2018). Table 3 summarizes the previous mentioned work on using EC as pretreatment process.

Table 3. Summary of the Previous Studies on Using EC as Pretreatment Process.

Hybrid system	Feed water	EC optimum parameters	Pollutants removal efficiency after EC process (%)	Enhancements to the system	Reference
EC-FO	Produced water	Electrodes: 2 Al electrodes Electrolysis time: 10 min Current density: 10 mA/cm^2	Conductivity: 5.4% TSS: 97% Turbidity: 91.6% Oil& grease: 97.4% TOC: 91.3%	Further removal of conductivity to reach: 16% Further removal of TSS to reach: 99% Further removal of turbidity to reach: 98%	(Al Hawli et al., 2019)
EC-MD	High salinity hydraulic fracturing produced water	Electrodes: 5 Al electrodes Electrolysis time: 0.5 min Current density: 16.5 mA/cm^2 Electrodes spacing: 0.5 cm	Turbidity: 96% TSS: 91% TOC: 61%	Water flux enhancement at 60℃: 50%	(Sardari, Fyfe, et al., 2018)

Hybrid system	Feed water	EC optimum parameters	Pollutants removal efficiency after EC process (%)	Enhancements to the system	Reference
EC-BF	Landfill leachates	Electrodes: Al at anode and stainless steel at cathode Electrolysis time: 20 min Current density: 8 mA/cm2 Electrodes spacing: 1.55 cm	COD: 37±2% TOC: 15±0% Ammonia: 6±8.7% Color: 60±13% Turbidity: 82±2.7% P: 82±5.5% Zn: 5±2.3, Fe: 95±2.6% Al: -26±23%	Further removal of COD, TOC, ammonia, color, turbidity, P, Zn, Fe, Al and BOD5 from the feed: 42±7%, 49±3.3%, 99%, -34±26%, 74±15%, -34±87%, -796±425%, 14±63%, 66±27% and 97% respectively	(Dia et al., 2018)
C-RO	Seawater	Electrodes: 2 Al electrodes Electrolysis time: 30 min Current density: 5.6 mA/cm^2 Electrodes spacing: 1 cm	DOC: 57.5% Absorbance: 81% Hardness: 10%	NA	(Hakizimana et al., 2016)

Hybrid system	Feed water	EC optimum parameters	Pollutants removal efficiency after EC process (%)	Enhancements to the system	Reference
EC-UF	Poultry processing wastewater	Electrodes: 5 Al electrodes Electrolysis time: 5 min Current density: 3 mA/cm2 Electrodes spacing: 0.5 cm	FOG: 94% TSS: 84% COD: 59% BOD: 87%	Further removal of FOG, TSS, COD, BOD and Proteins to reach: 100%, 100%, 92%, 98% and 90% respectively Water volume recovery: 30% Reuse of the membrane up to 6 times	(Sardari, Askegaard, et al., 2018)

Although electrocoagulation has shown high removal efficiency for different pollutants from wastewater, the energy and electrode consumption of EC needs to be improved further. Recently, there is an increased attention to the use of dielectrophoretic force (DEP) in electrocoagulation (Alkhatib et al., 2020; Hawari, Alkhatib, Das, Thaher, & Benamor, 2020). Alkhatib et al. studied the impact of using DEP force in EC process for the removal of chemical oxygen demand (COD) and total phosphorus (TP) from secondary treated wastewater (Alkhatib et al., 2020). The removal efficiency of COD and TP enhanced by 18% and 24 %, respectively, using DEP force compared with the regular EC process. Moreover, the electrode corrosion decreased by 87% when using DEP force in the EC system. Hawari et al. studied the impact of inducing DEP force for the enhanced harvesting of marine microalgae (Hawari et al., 2020). The major significance of using the DEP force in the EC process was found in the aluminum content in the harvested biomass which decreased by 52% compared to the conventional symmetrical EC electrodes.

According to the findings from our previous studies, the DEP force can enhance the removal efficiency of pollutants from wastewater and decrease the corrosion of electrodes. This study proposes a new electrodes configuration for the removal of Fe and Mn from primary treated municipal wastewater. The electrode configuration is an enhancement of our previous electrode configuration (Hawari et al., 2020). In this study two electrodes with rods will be used. Hence, each electrode is expected to produce a DEP force. The impact of current density, electrode spacing, and electrolysis time were investigated.

CHAPTER 3: MATERIALS AND METHODS

3.1 Characteristics of wastewater samples

Primary treated municipal wastewater was used as the feed solution in the process. The samples were collected from a municipal wastewater treatment plant located in the northern district of Doha, Qatar. The samples were collected after the grit removal stage. The initial concentrations of the pollutants of the collected wastewater samples are summarized in Table 4. The conductivity and pH of samples were measured using OAKTON PCD650 multi-meter. The turbidity was measured using a turbidity meter (Hach 2100p). Heavy metals concentration was measured using ICP-MS (Nexion 300D). As shown in Table 4 the concentration of most heavy metals was below the detection limit. As for Zn, Ni and Cu they also became below the detection limit after experiments. However, Fe and Mn were always detected, and the removal efficiencies were reported.

Table 4. Charachteristics of Primary Treated Municipal Wastewater.

Parameters	Value	Standard method
Temperature (°C)	22.5±0.3	APHA 2550 Temperature
Conductivity (mS/cm)	2.20±0.01	APHA 2510 B. Conductivity
pH	7.35±0.01	APHA 4500-H+ B. Electrometric Method
Turbidity (NTU)	158±2	APHA 2130 B. Nephelometric Method
Fe (mg/L)	0.124±0.001	EPA Method 200.8
Mn (mg/L)	0.118±0.001	
Al (mg/L)	0.038±0.001	
Zn (mg/L)	0.003±0.001	
Ni (mg/L)	0.003±0.001	
Cu(mg/L)	0.001±0.001	
Cd	< dL	
Cr	< dL	
Pb	< dL	
Co	< dL	

3.2 Experimental setup

Two electrode configurations were used in this study. The first configuration used two symmetrical flat sheet aluminum electrodes with dimensions of 7 cm x 5 cm connected to an AC power supply, this configuration will be called AC-EC (Figure 3(A)). The second configuration used two symmetrical aluminum electrodes with rods connected to an AC power supply, this configuration will be called to as AC-DEP (Figure 3(B)). In the AC-DEP configuration seven aluminum rods with a diameter of 2 mm were attached to the 7 cm x 5 cm aluminum sheets. The distance between each rod was 1 cm from center to center with an edge distance of 0.5 cm (Figure 3(B)). Figure 4 shows a schematic sketch of the experimental setup used in this study. A VARIAC transformer was used to generate a frequency of 50 Hz and the AC current with a voltage between 0 – 250 V. A TEKTRONIX oscilloscope device was used to get the current and voltage in the system. The experiments were performed at room

temperature. A magnetic stirrer was used at a stirring speed of 200 rpm to mix the solution in the reactor. After each experiment the samples were left to settle for 1 hour and then stored in the fridge at temperature 4^{o}C before analysis. The electrodes were washed with water and cleaned using sandpaper after each experiment.

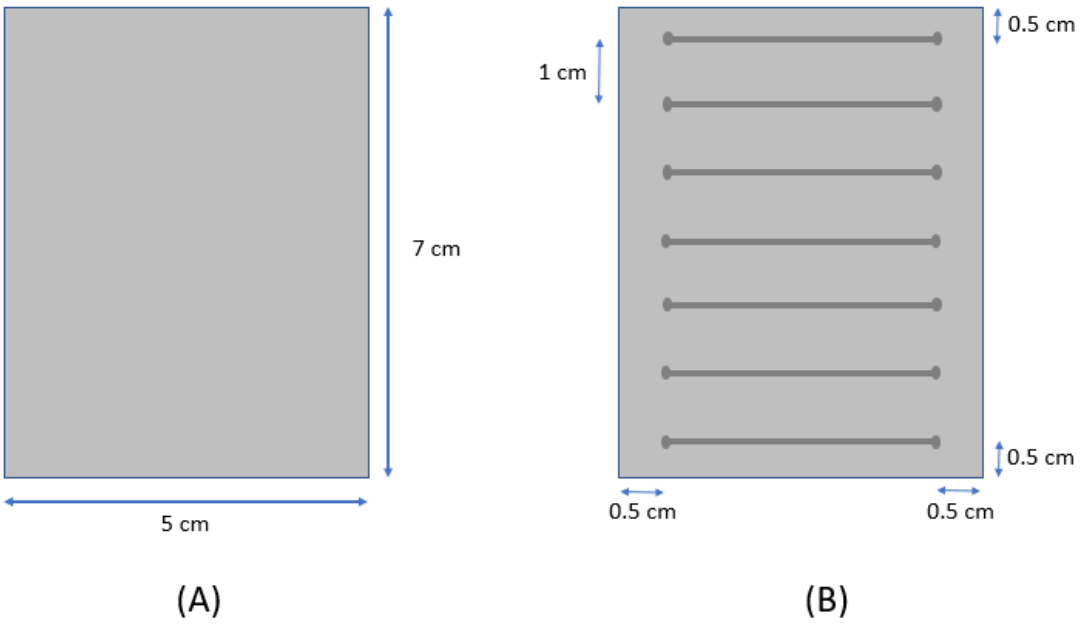


Figure 3. A schematic sketch for the electrodes used in this study (a) electrocoagulation electrode (AC-EC), (b) DEP inducing electrodes (AC-DEP).

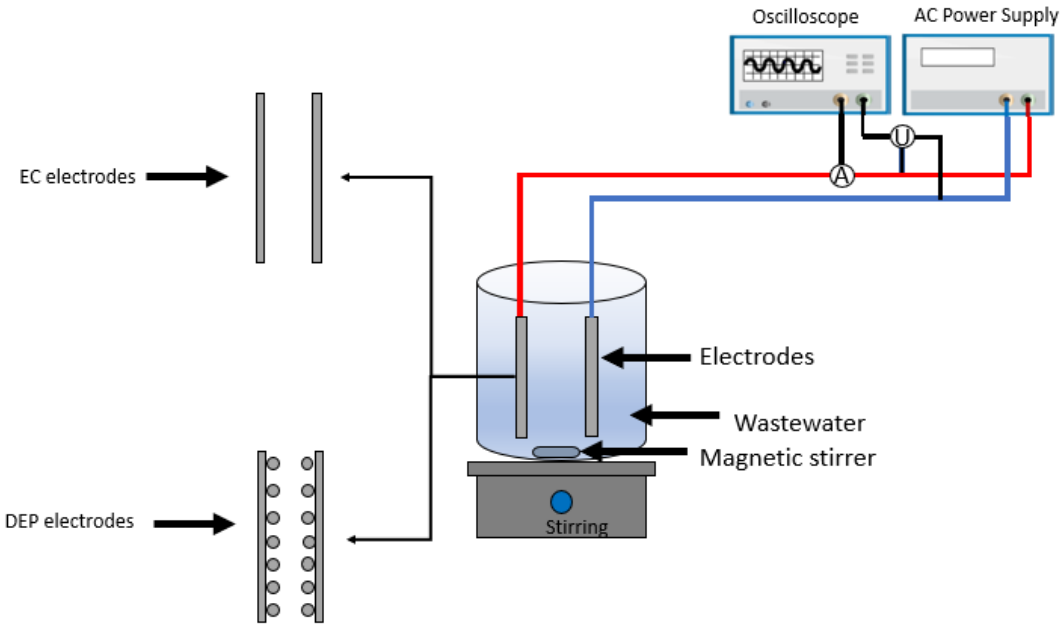


Figure 4. A schematic sketch for the bench scale electrocoagulation setup used in this study.

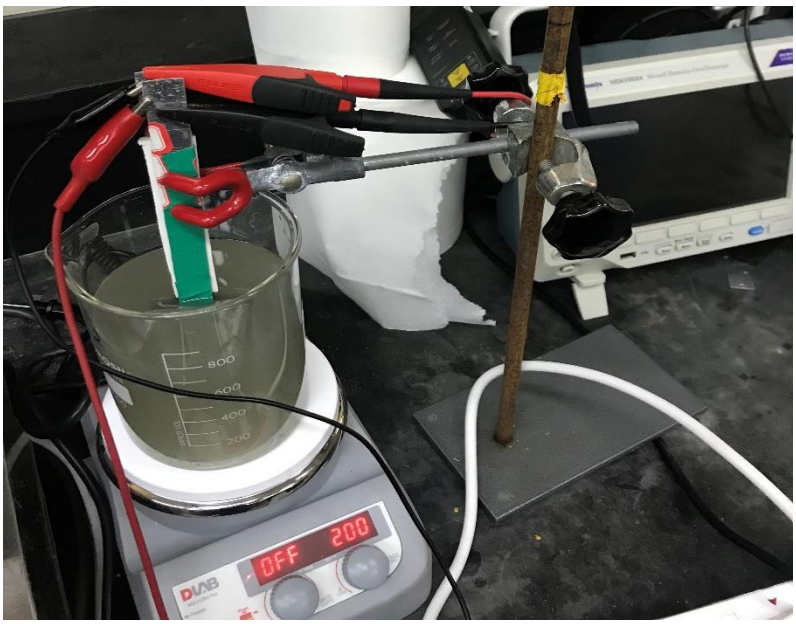


Figure 5. Electrocoagulation system setup used in the lab.

The removal % of heavy metals was calculated using Equation 11.

$$\text{M}\ (\%) = \frac{M_i - M_f}{M_i} \times 100 \quad (11)$$

Where M (%) is heavy metal removal efficiency, M_i is the initial heavy metal concentration in the wastewater (mg/L) and M_f is the final heavy metal concentration in the wastewater (mg/L). The specific energy consumption was calculated using Equation 12.

$$E_s = \frac{\text{U} \times \text{I} \times \text{t}}{\text{V}} \quad (12)$$

Where E_s is the specific energy consumption (Kwh/m^3), U is the electric potential (V), I is the applied current (A), t is the electrolysis time (h) and V is the sample volume (m^3).

3.3 Numerical simulation

DEP inducing electrodes were simulated in COMSOL Multiphysics software. The simulated electrodes are shown in Figure 6, 7 and 8. As noticed form Equation 13, the DEP force is directly related to the electric field. Therefore, the electric field squared was simulated in the model as an indicator for the DEP force.

$$\text{E} = -\nabla\varphi \quad (13)$$

In which, φ is the root mean square (rms) related to the electrostatic potential that can be provided by Laplace's equation. The boundary conditions that were applied on the electrodes surface are fixed.

$$\Phi_1 = \text{U}_\text{o} \quad (14)$$

$$\varphi_2 = 0 \quad (15)$$

U_0 is the oscillating potential drop rms. The simulated medium was primary treated wastewater. The model was made of two dimensions, while assuming the width of the electrode is almost infinity.

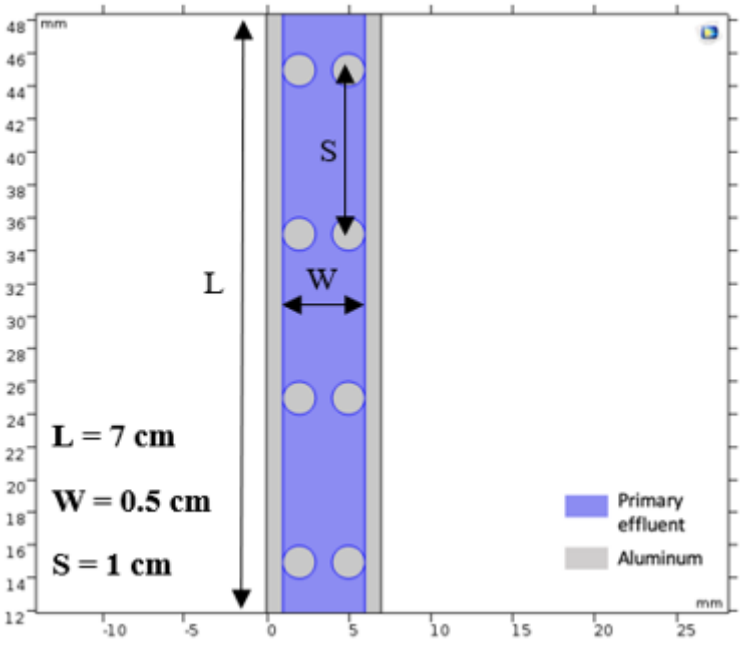


Figure 6. Illustration for the geometrical parameters of the simulated electrodes using electrodes spacing of 0.5 cm.

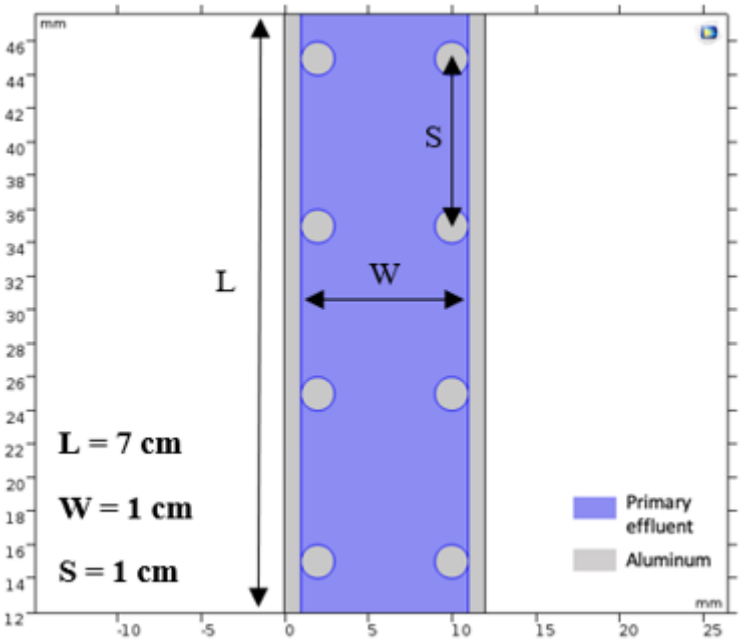


Figure 7. Illustration for the geometrical parameters of the simulated electrodes using electrodes spacing of 1 cm.

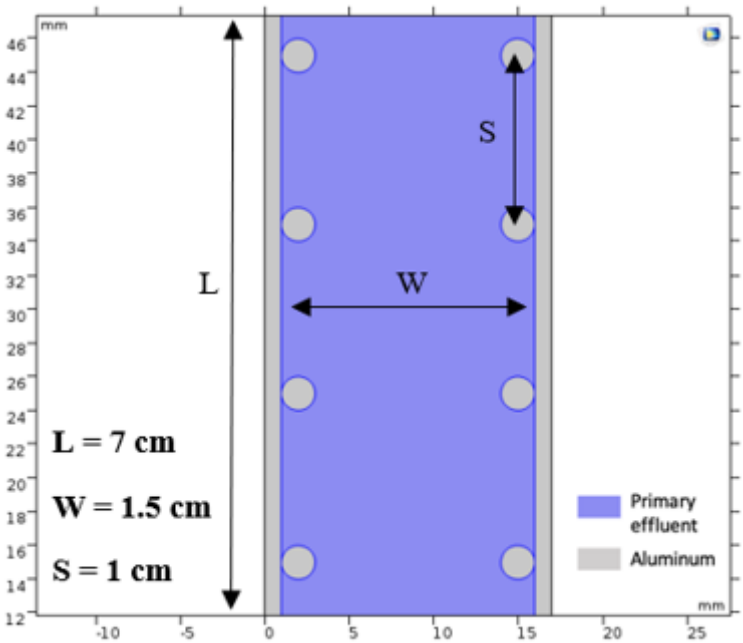


Figure 8. Illustration for the geometrical parameters of the simulated electrodes using electrodes spacing of 1.5 cm.

3.4 Error estimation

All the experimental runs were repeated for three times. The reported result is the average of the experimental trials. The Error shown represents the standard deviation of the results. All the error bars of the standard deviation did not exceed 3%.

CHAPTER 4: RESULTS AND DISCUSSION

4.1 Numerical simulation

4.1.1 Effect of electrodes spacing

The effect of electrodes spacing on the DEP force was evaluated using three different electrodes spacing (i.e. 0.5 cm, 1 cm and 1.5 cm) at an applied current of 600 mA. Figure 9, 10 and 11 show that the DEP force presented by the electric field squared (∇E^2) was the highest when the spacing between the electrodes was 0.5 cm compared to 1 cm and 1.5 cm. At an electrodes' spacing of 0.5 cm the maximum ∇E^2 was 1.2 x 10^{11} v^2/m^3 at the electrode surface and the minimum ∇E^2 was at midpoint with a value of 4.0 x 10^{10} v^2/m^3. At an electrodes' spacing of 1.0 cm the maximum ∇E^2 was 7.0 x 10^9 v^2/m^3 at the electrode surface and went down to zero at a distance 2 mm away from the electrode surface. It was also noticed that a zone with no DEP effect was present. The zone extended for a length of 2 mm. At an electrodes' spacing of 1.5 cm the maximum ∇E^2 was 3.4 x 10^9 v^2/m^3 at the electrode surface and went down to zero at a distance 3 mm away from the electrode surface. It was also noticed that a zone with no DEP effect was present. The zone extended for a length of 5 mm. The results are as expected because at electrodes spacing of 0.5 cm the force from one rod has reached the force from the opposite rod which results in mitigate the zero force in between and increasing the force at the rods. Unlike the other cases of electrodes spacing 1 cm and 1.5 cm.

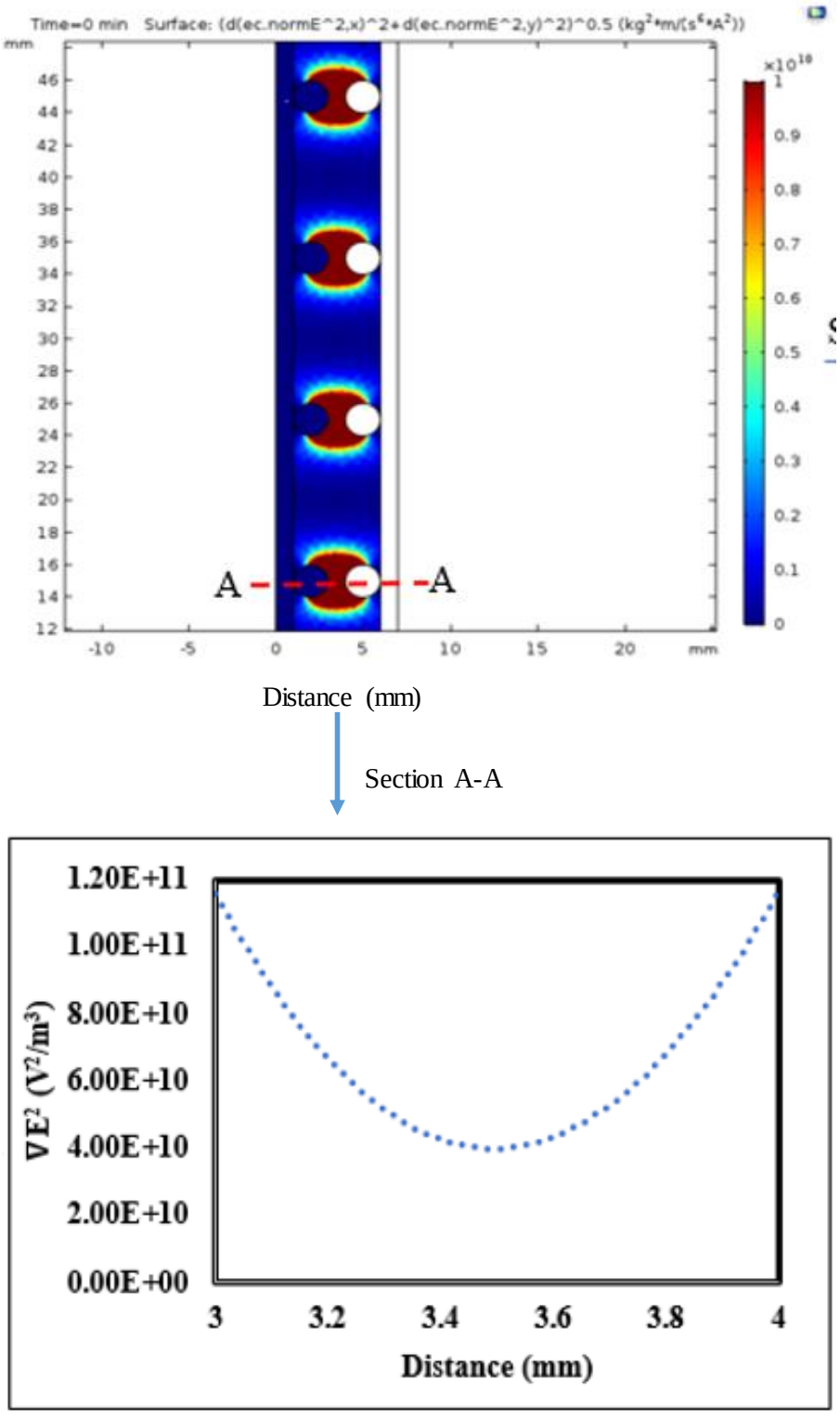


Figure 9. DEP force field ($\nabla|E|^2$) using current of 600 mA and electrodes spacing of 0.5 cm.

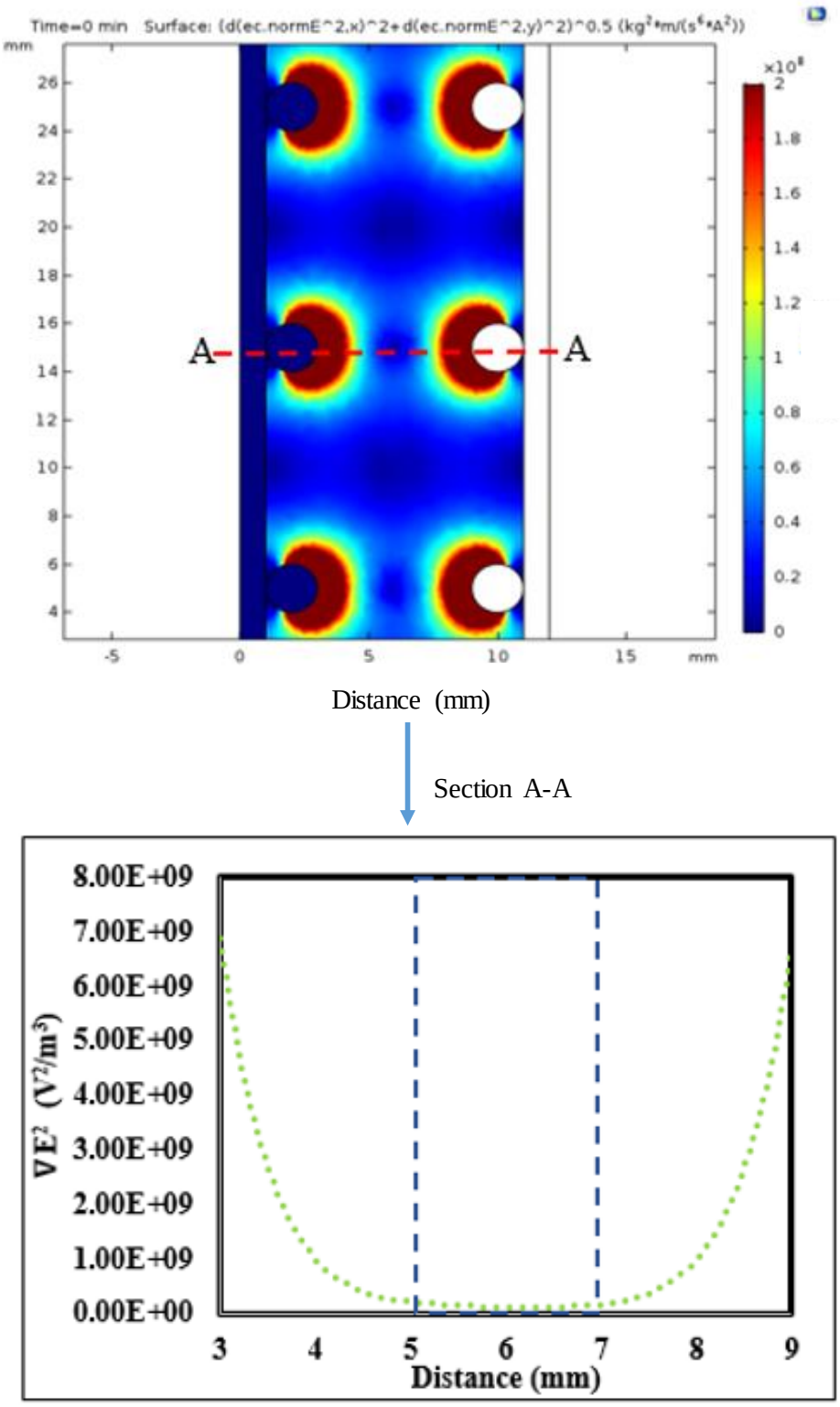


Figure 10. DEP force field ($\nabla|E|^2$) using current of 600 mA and electrodes spacing of 1 cm.

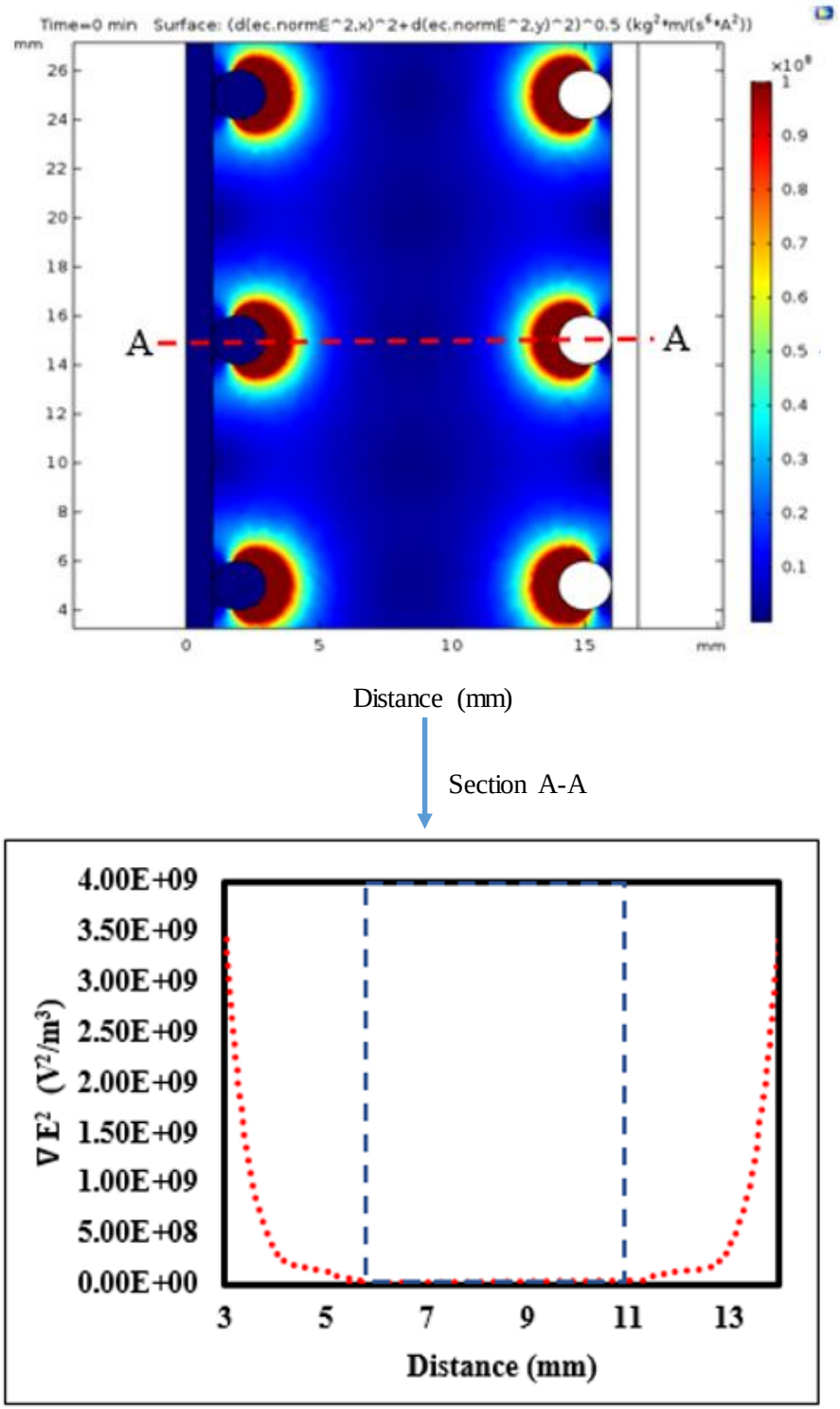


Figure 11. DEP force field ($\nabla|E|^2$) using current of 600 mA and electrodes spacing of 1.5 cm.

4.1.2 Effect of current density

The effect of current density on the DEP force field was evaluated using four applied currents 200 mA, 400 mA, 600 mA and 800 mA with a current density of 5.71, 11.43, 17.14 and 22.86 mA/cm^2, respectively. The electrode spacing was fixed at 0.5 cm. As shown in Figure 12, 13, 14 and 15, the DEP force increased as the applied current increased. The DEP force field was minimal when using an applied current of 200 mA. However, the DEP force field affected larger area using an applied current of 400 mA and became significant when using an applied current of 600 mA and 800 mA.

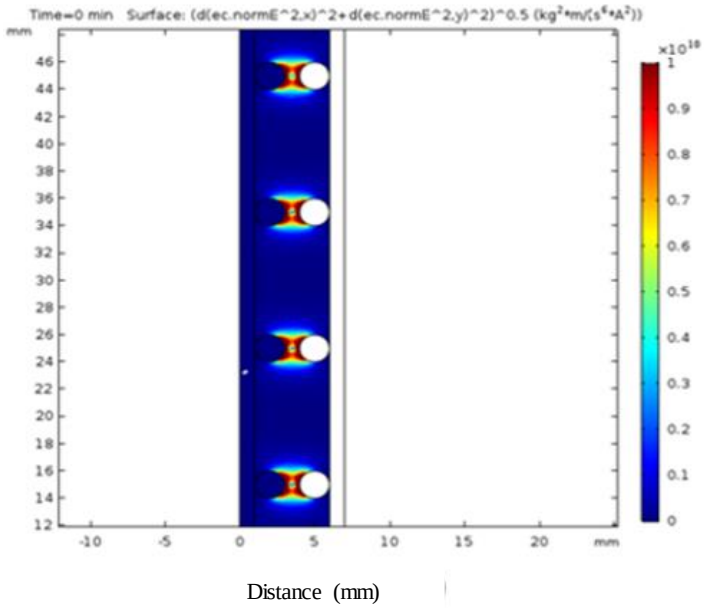


Figure 12. DEP force field ($\nabla|E|^2$) using electrode spacing of 0.5 cm and applied current of 200 mA.

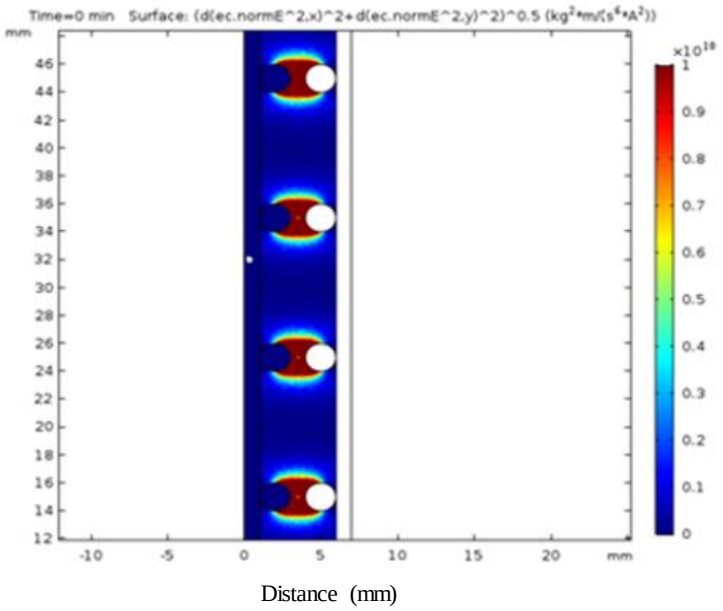


Figure 13. DEP force field ($\nabla|E|^2$) using electrode spacing of 0.5 cm and applied current of 400 mA.

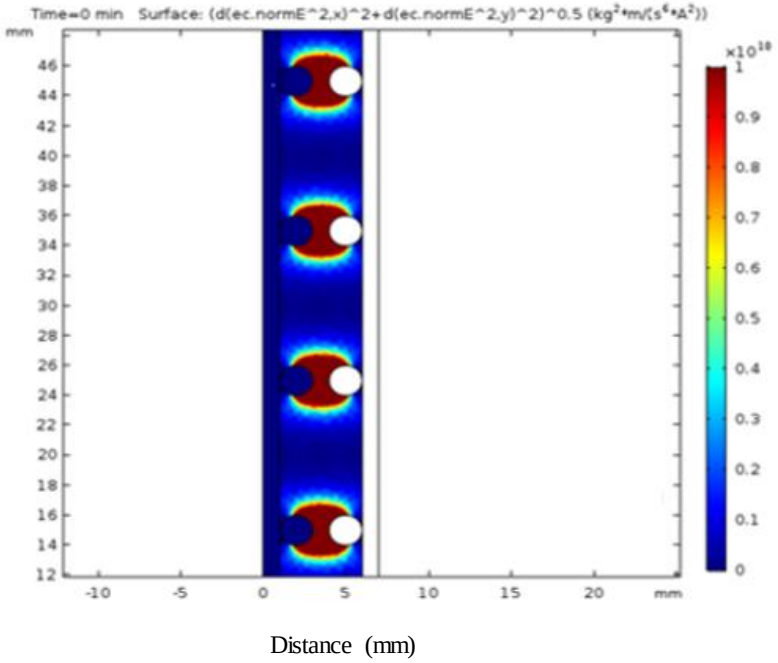


Figure 14. DEP force field ($\nabla|E|^2$) using electrode spacing of 0.5 cm and applied current of 600 mA.

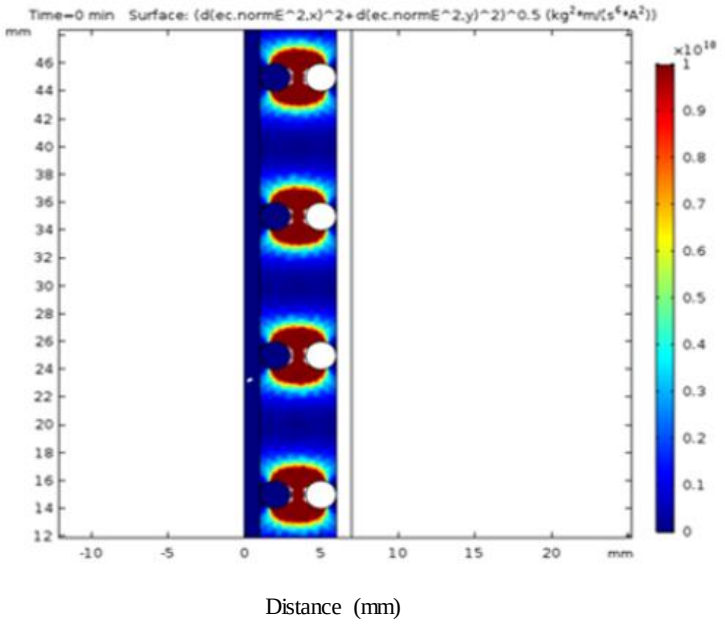


Figure 15. DEP force field ($\nabla|E|^2$) using electrode spacing of 0.5 cm and applied current of 800 mA.

Figure 16 shows the effect of the applied current on the squared electric field. The electric field squared (∇E^2) at the surface of the electrodes was almost 1.5 x 10^{10} v^2/m^3 using an applied current of 200 mA. ∇E^2 at the surface of the electrodes increased significantly by 70% using an applied current of 400 mA. As the applied current increased to 600 mA, ∇E^2 increased by 36% and further enhanced by 54% at an applied current of 800 mA.

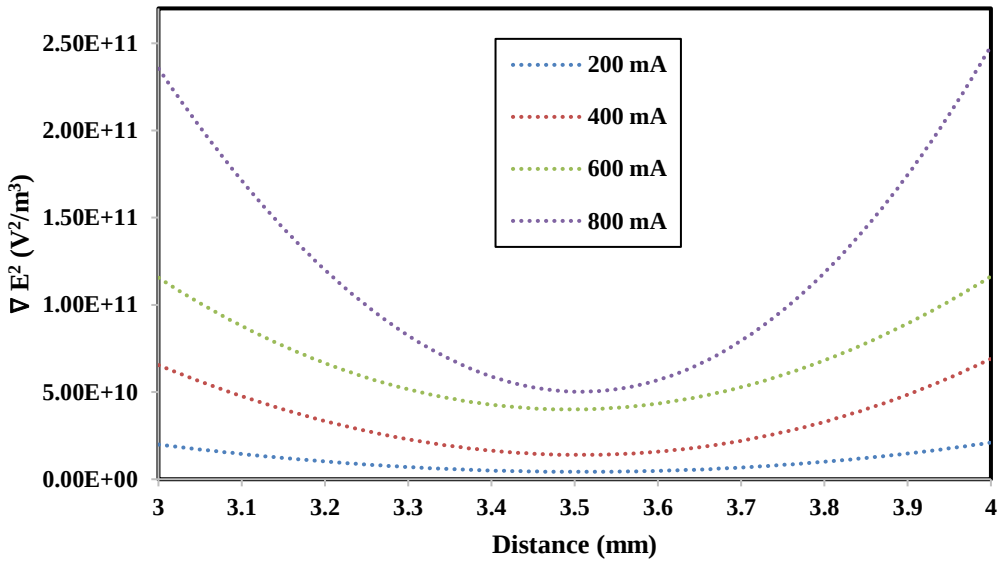


Figure 16. Electric filed squared ($\nabla|E|^2$) using electrode spacing of 0.5 cm and different applied currents 200 mA, 400 mA, 600 mA and 800 mA.

4.2 Experimental study

4.2.1 Effect of electrodes spacing

To evaluate the impact of electrode spacing, the applied current was fixed at 600 mA and the electrolysis time was 30 minutes. As shown in Figure 17 (a) and (b), the Fe and Mn removal % decreased as the electrode spacing increased using both (AC-EC) and (AC-DEP) configurations. The Fe removal % using (AC-DEP) was slightly higher than (AC-EC). At an electrode distance of 0.5 cm, the removal % of Fe using (AC-EC) and (AC-DEP) were 78.8% and 80.3%, respectively. The Fe removal % decreased by 12.5% as the electrode spacing increased to 1 cm using (AC-EC) and (AC-DEP). The Fe removal % decreased by 9.1% as the electrode spacing increased to 1.5 cm using (AC-EC) and (AC-DEP). The Mn removal % using (AC-DEP) was slightly higher than (AC-EC). At an electrode distance of 0.5 cm, the removal % of Mn using (AC-EC) and (AC-DEP) were 28.8% and 29.6%, respectively. The removal % of Mn

decreased by 19.9% as the electrode spacing increased to from 0.5 cm to 1 cm using both (AC-EC) and (AC-DEP). The Mn removal % decreased by 4.9% as the electrode spacing increased from 1 cm to 1.5 cm using both (AC-EC) and (AC-DEP). The maximum removal % of Fe and Mn was obtained using an electrode spacing of 0.5 cm. The removal % of Fe and Mn was lower at higher electrodes spacing due to the increase in the electrical resistance in the solution. Higher resistance would reduce the dissolution of coagulants from the electrodes thus reduces the removal efficiency of pollutants in the system (Mohora et al., 2012; Sahu et al., 2014). In addition, the enhancement of the removal percentage of Fe and Mn in the (AC-DEP) configuration compared to the (AC-EC) configuration could be due to the effect of the DEP force. The DEP force is expected to enhance the interaction between particles and the formation of flocs, hence, enhance the removal percentage of Fe and Mn. As the spacing between the electrodes decreases, the DEP force increases, and higher removal efficiencies of Fe and Mn were obtained. These results are compatible with the simulation results where it was found that the DEP force was the highest at an electrodes spacing of 0.5 cm and as the distance between the electrodes increased the DEP force decreased. In addition, from the simulation studies it was found that at electrodes' distance of 1 cm and 1.5 cm a zone of no DEP effect existed.

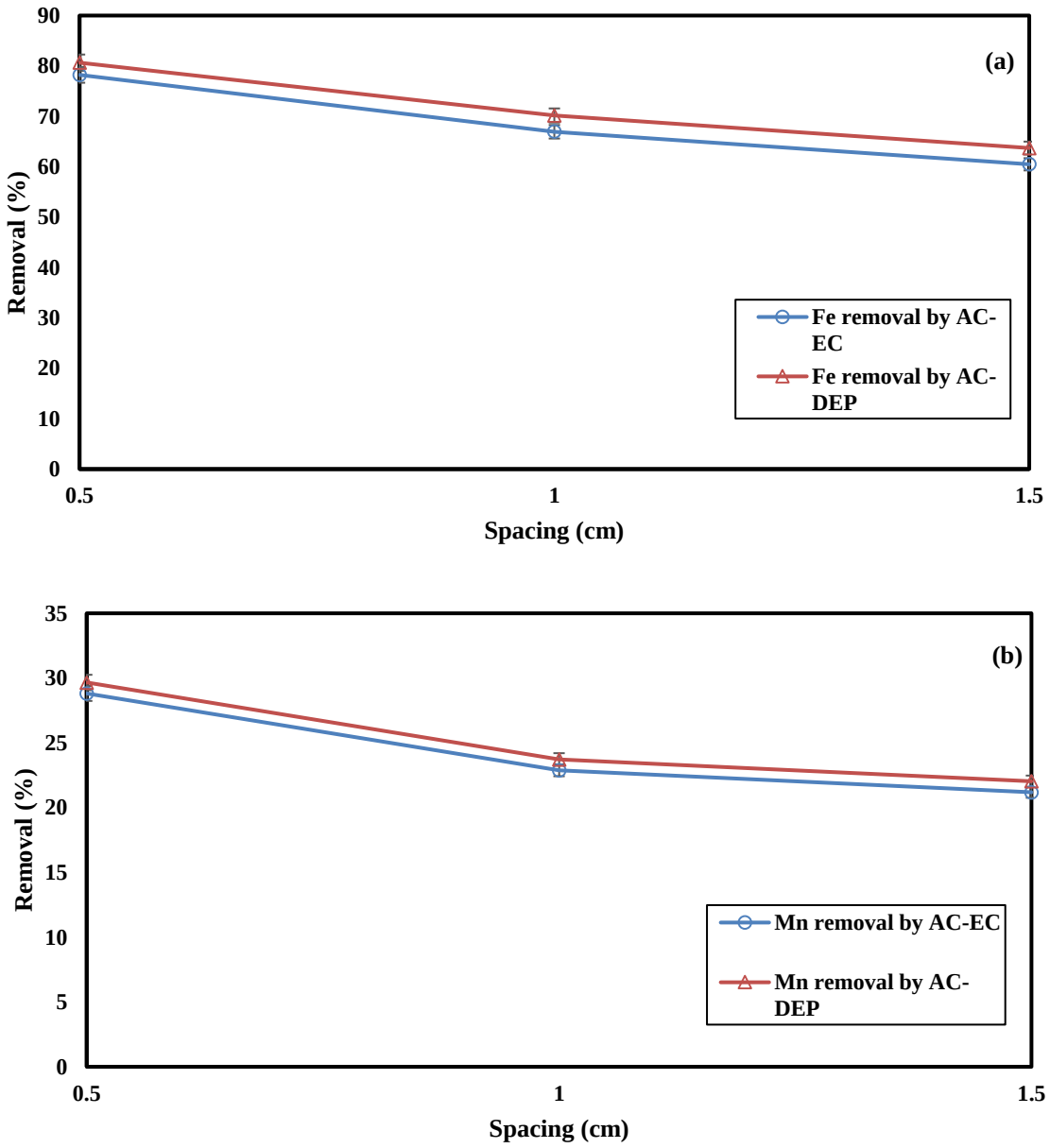


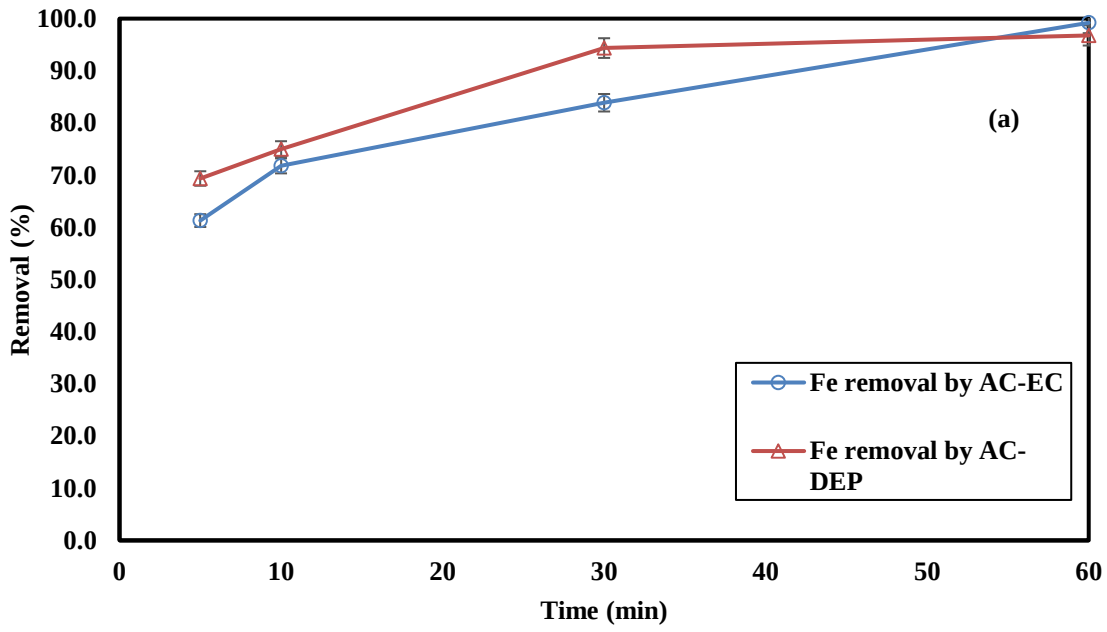
Figure 17. The removal % of Fe and Mn using variable electrode spacing, electrolysis time of 30 min and applied current 600 mA (a) Fe removal % (b) Mn removal %.

4.2.2 Effect of electrolysis time

An electrolysis time of 5, 10, 30 and 60 minutes was investigated. The applied current and the electrode spacing were fixed at 800 mA and 0.5 cm, respectively. As shown in Figure 18 (a), the Fe removal % increased as the electrolysis time increased

using both (AC-EC) and (AC-DEP) configurations. The removal percentage of Fe using (AC-DEP) was higher than (AC-EC) where at an electrolysis time of 5 minutes, the removal % of Fe using (AC-EC) and (AC-DEP) were 61.3% and 69.3%, respectively. The Fe removal % increased by 14.7% (AC-EC) and 3.4% (AC-DEP) as the electrolysis time increased to 10 min. The Fe removal % increased by 14.5% (AC-EC) and 21.2% (AC-DEP) as the electrolysis time increased to 30 min. At an electrolysis time of 60 minutes, the Fe removal % increased by 15.4% using (AC-EC), while the Fe removal % remained almost constant using (AC-DEP). As shown in Figure 18 (b), the Mn removal % increased as the electrolysis time increased using both (AC-EC) and (AC-DEP) modes. At an electrolysis time of 5 minutes, the removal % of Mn using (AC-EC) and (AC-DEP) were almost the same with a value of 5%. As the electrolysis time increased to 10 minutes, the Mn removal % increased by 35.5% (AC-EC) and 56.8% (AC-DEP). At an electrolysis time of 30 minutes, the Mn removal % increased significantly by almost 80% using both (AC-EC) and (AC-DEP) configurations. At electrolysis time 60 min, the Mn removal % increased by 30.1% using (AC-EC) and 13.2% using (AC-DEP). It can be seen that the electrolysis time has a major influence on the removal efficiency of Fe and Mn in the EC process. The electrolysis time will affect the amount of metal ions produced from the elctrodes (Daneshvar, Oladegaragoze, & Djafarzadeh, 2006). As the electrolysis time increases more ions will be produced and hence higher removal efficiencies are expected (Daneshvar et al., 2006). It can be seen from Figure 18 (a) and (b) that the removal efficiency of Fe was higher than the removal efficiency of Mn. The higher removal efficiency of Fe could be attributed to the lower solubility of the formed iron hydroxides. The formation of manganese and iron hydroxides and their precipitation play a dominant role in the removal mechanism of the corresponding metallic ions (Adhoum, Monser, Bellakhal,

& Belgaied, 2004; Shafaei, Pajootan, Nikazar, & Arami, 2011; Shafaei et al., 2010). The solubility constants (K_{sp}) of manganese and iron hydroxides at 25°C are 1.9×10^{-13} and 2.0×10^{-15}, respectively. It can be also noticed from Figure 18 (a) and (b) that at an electrolysis time of 60 minutes the removal efficiency of Fe and Mn in the (AC-DEP) configuration was less than that in the (AC-EC) configuration. This could be due to the fact that after a long period of DEP force application, the force could break the already formed flocs and hence reduce the removal efficiency.



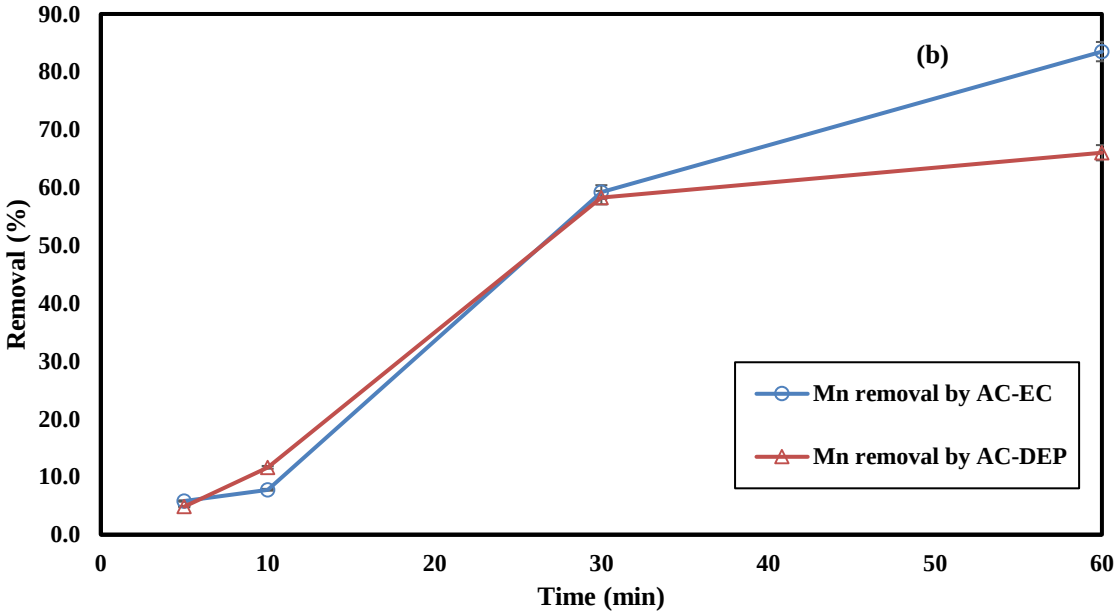


Figure 18. The removal % of Fe and Mn using variable electrolysis time, electrode spacing 0.5 cm and applied current 800 mA (a) Fe removal % (b) Mn removal %.

4.2.3 Effect of applied current

Four different applied currents were tested: 200, 400, 600 and 800 mA corresponding to current densities of 5.71, 11.43, 17.14 and 22.86 mA/cm^2, respectively. The electrolysis time was fixed at 30 min and the electrode spacing was 0.5 cm. As shown in Figure 19, the removal % of Fe and Mn increased as the applied current increased. When using an applied current density of 5.71 mA/cm^2 the Fe removal % was around 28% using both (AC-EC) and (AC-DEP). The Fe removal % increased by 46.2% and 54.8% using an applied current density of 11.43 mA/cm^2 compared to an applied current density of 5.71 mA/cm^2 in (AC-EC) and (AC-DEP) configurations, respectively. The Fe removal % increased to around 80 % using an applied current density of 17.14 mA/cm^2 in both (AC-EC) and (AC-DEP) configurations. The maximum removal % of Fe was 94.3% obtained using an applied

current density of 22.86 mA/cm^2 in (AC-DEP) configuration. The removal % of Mn was 4.2% using a current density of 5.71 mA/cm^2 in (AC-EC) configuration. The Mn removal % increased by 72.3% and 93.3% using a current density of 11.43 mA/cm^2 compared to a current density of 5.71 mA/cm^2 in (AC-EC) and (AC-DEP) configurations, respectively. The Mn removal % increased to almost 30 % using a current density of 17.14 mA/cm^2 in both (AC-EC) and (AC-DEP) configurations. The maximum removal % of Mn was 63.6% obtained using a current density of 22.86 mA/cm^2 in both (AC-EC) and (AC-DEP) configurations. The production of Al^{3+} ions increases as the current density increases due to the enhanced dissolution of the aluminum electrodes (Aoudj, Khelifa, Drouiche, Hecini, & Hamitouche, 2010; Gao et al., 2010). Therefore, the amount of trapped heavy metals increased due to the increased amount of coagulants. In addition, higher current density means higher DEP force as the DEP force is directly proportional with the applied voltage. Higher DEP force means higher interaction between particles and hence more floc formation and higher removal efficiencies.

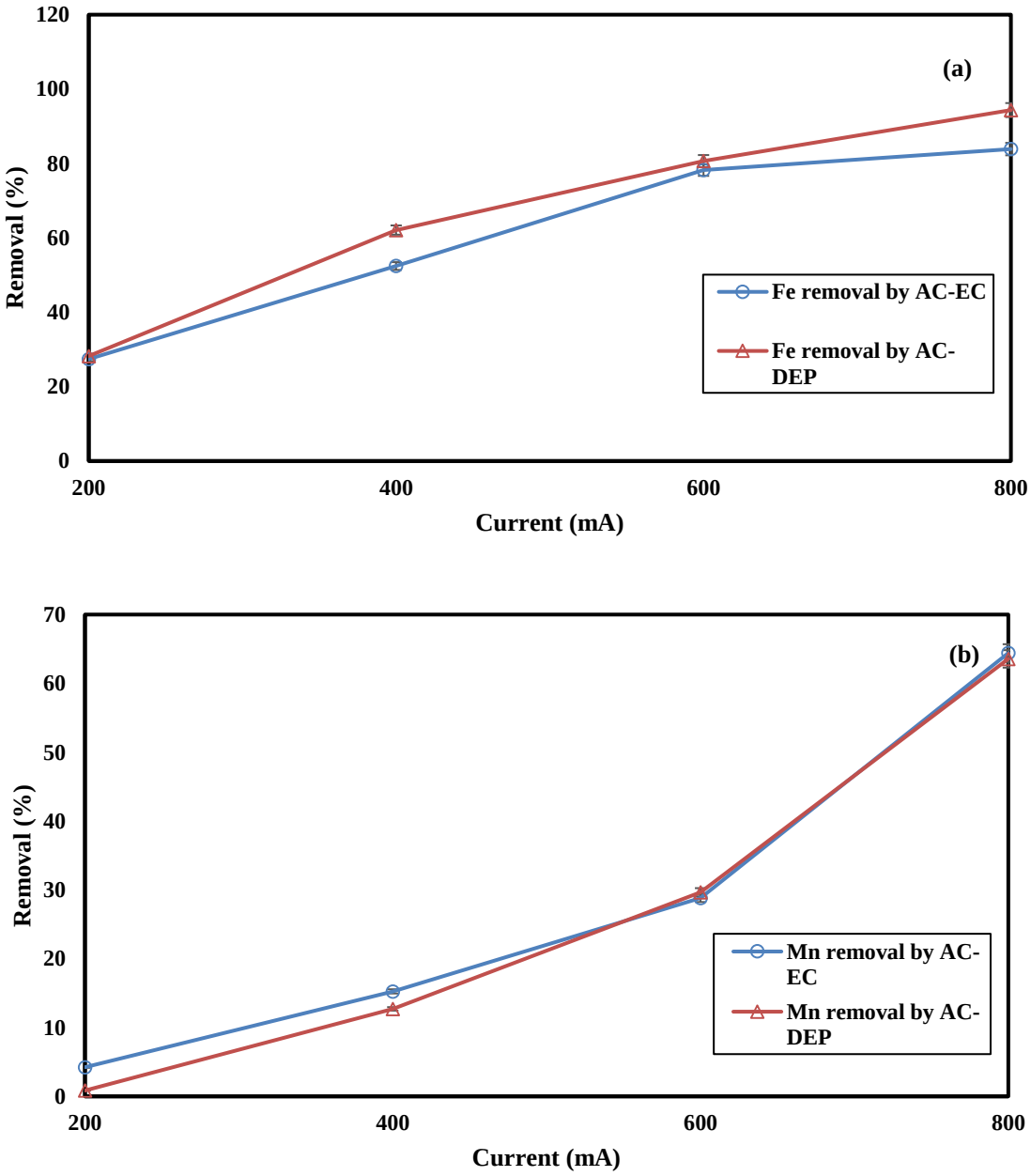


Figure 19. The removal % of Fe and Mn using variable applied current, electrolysis time 30 min and electrode spacing 0.5 cm (a) Fe removal % (b) Mn removal %.

The main advantage of using the DEP inducing electrodes lies not only in the enhanced removal efficiency but in the amount of consumed Al in the EC process. Figure 20 shows the aluminum content in the reactor with respect to the applied current.

The aluminum content increased as the applied current increased using both (AC-EC) and (AC-DEP) configurations. However, the aluminum content obtained using the (AC-DEP) configuration was much lower than the (AC-EC) configuration. At an applied current of 400 mA, the aluminum content using (AC-DEP) was almost 31% less than the (AC-EC). As the current density increased the difference in aluminum content also increased. The aluminum content using (AC-DEP) was almost 42% less than the (AC-EC) at an applied current of 800 mA.

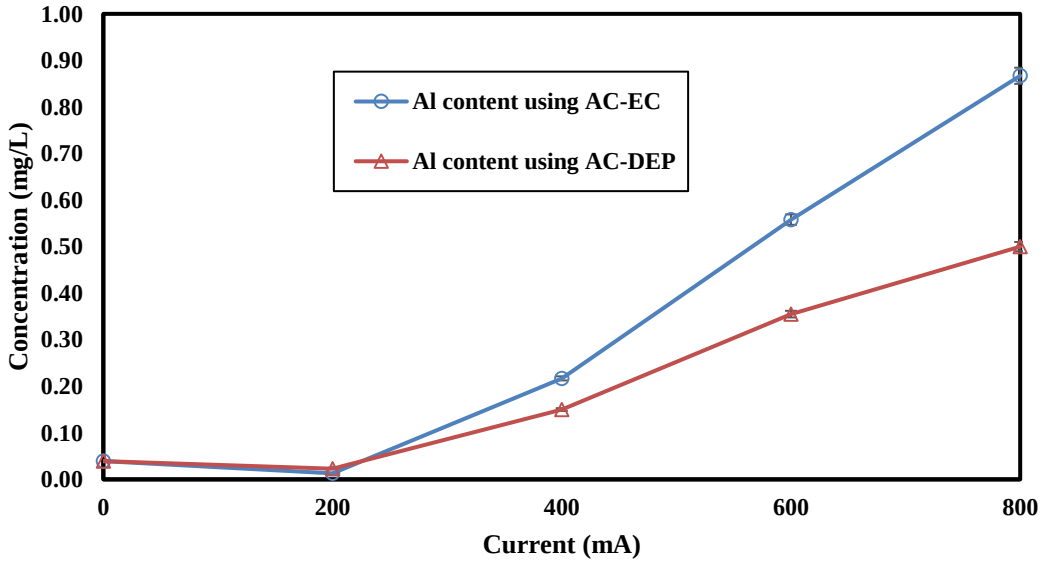


Figure 20. Aluminum content in the reactor using AC-EC and AC-DEP with 0.5 cm electrodes spacing and an operational time of 30 min.

4.2.4 Energy consumption

The energy consumption of the system was evaluated for the four different applied currents (i.e. 200, 400, 600 and 800 mA). The electrode spacing and electrolysis time were fixed at 0.5 cm and 30 min, respectively. As shown in Figure 21, the energy

consumption increased as the applied current increased. When using an applied current of 600 mA, the energy consumption of in the (AC-DEP) was 2% lower than (AC-EC) and at an applied current of 800 mA the energy consumption obtained using the (AC-DEP) was 3% lower than (AC-EC). The energy consumption was almost the same using (AC-EC) and (AC-DEP) configurations. The minimal differences in the energy consumption between the two configurations are due to the differences in the resistance in each system. The shape of the electrodes configuration affected the resistance in the system and thereby affected the amount of consumed energy.

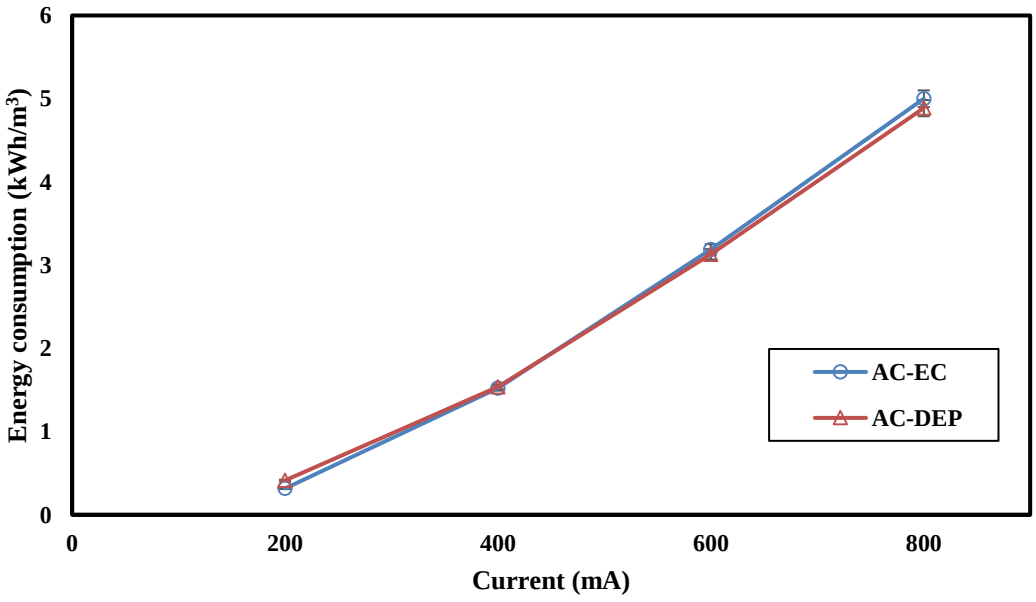


Figure 21. (AC-EC) and (AC-DEP) energy consumption using 30 min of operational time and 0.5 cm electrodes spacing on four different currents which are: 200, 400, 600 and 800 mA.

CHAPTER 5: CONCLUSION

In this study a new electrocoagulation (EC) electrode configuration has been investigated for the removal of Fe and Mn from primary treated municipal wastewater. The effect of electrolysis time, electrodes spacing and current density on the removal of Fe and Mn was investigated. The experimental results showed the following:

- As the electrolysis time increased, the removal of Fe and Mn increased. As the electrolysis time increases more ions will be produced and hence higher removal efficiencies are expected. The maximum removal of Fe and Mn were obtained at an electrolysis time of 60 mins. The removal of Fe was 99.2% using (AC-EC) and 96.8% using (AC-DEP). The removal of Mn was 83.5% and 66% using (AC-EC) and (AC-DEP), respectively.
- As the distance between the electrodes decreased, the Fe and Mn removal increased. This is due to the decrease in the electrical resistance in the solution. Lower resistance would increase the dissolution of coagulants from the electrodes thus increases the removal efficiency of pollutants in the system. The maximum Fe and Mn removal was observed at an electrode distance of 0.5 cm.
- As the applied current increased, the removal efficiency increased. The production of Al^{3+} ions increases as the current density increases due to the enhanced dissolution of the aluminum electrodes. Therefore, the amount of trapped heavy metals increased due to the increased amount of coagulants. The maximum Fe and Mn removal was obtained at applied current of 800 mA.
- The main advantage of using the new DEP inducing electrodes lies not only in the enhanced removal efficiency but in the amount of consumed Al in the EC process. The aluminum content increased as the applied current increased using both (AC-EC) and (AC-DEP) configurations. However, the aluminum content

obtained using the (AC-DEP) configuration was much lower than the (AC-EC) configuration. The aluminum content using (AC-DEP) was almost 42% less than the (AC-EC) at an applied current of 800 mA.

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