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COLLEGE OF ENGINEERING

A NEW ELECTROCOAGULATION ELECTRODE CONFIGURATION FOR THE
TREATMENT OF SECONDARY TREATED WASTEWATER

BY

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ABSTRACT

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Title: A New Electrocoagulation Electrode Configuration for The Treatment of Secondary Treated Wastewater

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Electrocoagulation (EC) is an effective method that is applied to treat a wide range of wastewater. A new electrode configuration is being proposed with the application of the Dielectrophoresis (DEP) force which is expected to enhance the quality of Treated Sewage Effluent (TSE) using unsymmetrical cylindrical aluminum electrodes with alternating current (AC) power source. In this study, the impacts of power supply type and electrode configuration were examined on both Total Phosphorous (TP) removal efficiency and Chemical Oxygen Demand (COD) reduction. Experiment on three different modules were conducted which are unsymmetrical cylindrical aluminum electrodes with AC power source (AC-DEP), symmetrical cylindrical aluminum electrodes with AC power source (AC) and symmetrical cylindrical aluminum electrodes with DC power source (DC). The simulated results showed that the strength of the DEP force increases as the current density increases. Moreover, the DEP force decreases with distance and reaches minimum magnitude at the outer electrode. AC-DEP electrode configuration with 4.3 mA/cm^2 current density generates sufficient DEP force that significantly enhance the quality of TSE. The experimental results showed that the maximum removal efficiency of TP is 88.3, 68.2 and 46.0 % and COD reduction of 82.4, 66.8 and 43.3 % with electrode mass consumption of 0.02, 0.15 and 2.12 g under running conditions of 4.3 mA/cm^2 current

density and 30 min electrolysis time for AC-DEP, AC and DC, respectively.

ABSTRACT (IN ARABIC)

يعتبر التخثر الكهربائي (Electrocoagulation) وسيلة فعالة لمعالجة نطاق واسع من مياه الصرف الصحي. تم اقتراح تكوين قطب جديد مع تطبيق قوة العزل الكهربائي (Dielectrophoresis) التي من المتوقع أن تعزز جودة المخلفات السائلة المعالجة ثانوياً (Treated Secondary Effluent) باستخدام أقطاب الألومنيوم الاسطوانية غير المتناظرة الموصلة بمولد التيار المتردد. خلال هذه الدراسة، تم فحص تأثير نوع المولد الكهربائي ونوع تكوين الأقطاب على كل من النسبة المئوية لإزالة الفوسفور الكلي (TP) وتقليل المتطلب الكيميائي للأوكسجين (COD). أجريت التجربة على ثلاث وحدات مختلفة وهي أقطاب الألومنيوم الاسطوانية غير المتناظرة الموصلة بمولد التيار المتردد (AC-DEP) وأقطاب الألومنيوم الاسطوانية المتناظرة الموصلة بمولد التيار المتردد (AC) وأقطاب الألومنيوم الاسطوانية المتناظرة الموصلة بمولد التيار المستمر (DC). أظهرت نتائج المحاكاة أن شدة قوة العزل الكهربائي تزداد كلما زادت كثافة التبار الكهربائي. علاوة على ذلك، تتناقص شدة قوة العزل الكهربائي مع زيادة المسافة وتصل الى القيمة الصغرى عند القطب الخارجي. تولد أقطاب الألومنيوم الأسطوانية المتناظرة (AC-DEP) باستخدام كثافة تيار ٤,٣ مل امبير/سم^٢ كمية قوة عزل كهربائي كافية لتعزز بشكل ملحوظ من جودة النفايات السائلة المعالجة ثانوياً. أظهرت نتائج الدراسة التجريبية أن الحد الأقصى لإزالة الفوسفور الكلي هو ٨٨,٣٪، ٦٨,٢٪ و ٤٦,٠٪ بينما الحد الأقصى لتقليل المتطلب الكيميائي للأوكسجين هو ٨٢,٤٪، ٦٦,٨٪ و ٤٣,٣٪ وكمية استهلاك كتلة القطب هي ٠,٠٠٢، ٠,١٥ و ٢,١٢ غرام تحت ظروف التشغيل الآتية ٤,٣ مل امبير/سم^٢ و زمن تفاعل مقداره ٣٠ دقيقة ل AC-DEP، AC و DC، على التوالي.

DEDICATION

This thesis is dedicated to my parents, Mohammad and Enas who taught me to work hard for what I aspire to achieve. This work is also dedicated to my friends who supported me.

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CHAPTER 1: INTRODUCTION

Water is an essential element in earth. A remarkable quantity of water enters in a direct or indirect way in human activities (Cipollina, Micale, & Rizzuti, 2009). There is a shortage in freshwater around the world. Seawater represents 97% of the earth's water and only 3% represents freshwater and from this 3%, only 0.3% is surface water that is available for direct usage (Saeed et al., 2015).

1.1 Wastewater reuse potential in Qatar

Groundwater and desalinated water are the only freshwater resources in Qatar (Jasim, Saththasivam, Loganathan, & Ogunbiyi, 2016). However, the groundwater aquifers depletion and the high Water-Production Cost (WPC) for desalinated water are the major challenges in the management of water systems in Qatar (Jasim et al., 2016). In the GCC region, Qatar has the fastest growing water demand with significant governmental funds on water supply and water desalination (Jasim et al., 2016). The continuous increase in population, industrial and agriculture activities, increases the demand of desalinated water. Therefore, various wastewater treatment technologies have been developed to make wastewater reusable (Jasim et al., 2016; Saleem, Bukhari, & Akram, 2011). Wastewater treatment is more cost-effective and consumes less energy than water desalination plants (Jasim et al., 2016).

Wastewater treatment plants remove the contaminants from wastewater to meet water quality standards. Hence, it can be returned back to the environment and reused for many useful applications and sectors (*Guidelines for Water Reuse*, 2012; Jasim et al., 2016). Treated secondary effluent from wastewater treatment plants contains contaminants, such as TP and COD that has to be removed before water can be reused (Lv, Feng, Liu, & Xie, 2017; Szymański, Morawski, & Mozia, 2018). Thus, tertiary treatments are essential. The main sectors that utilizing the treated wastewater and its

application are (*Guidelines for Water Reuse*, 2012; Jasim et al., 2016):

- 1- Urban: vehicle washing and fire protection.
- 2- Agriculture: food and non-food crops and livestock watering.
- 3- Impoundments: recreation area and snowmaking.
- 4- Environmental: artificial wetlands.
- 5- Industrial: cooling towers and produced water from gas production.
- 6- Groundwater recharge.
- 7- Potable: indirect and direct potable reuse.

In GCC countries, the largest amount of water consumed is in agricultural sector followed by domestic and industrial sectors (Jasim et al., 2016; Sayed & Ayoub, 2014). Due to the improvement of life on all levels, the demand of water continuously increases.

The characteristics of secondary effluent differ slightly depend on the design of the wastewater treatment that was applied, Table 1 shows the characteristics of secondary effluent from various treatment plants:

Table 1. *Characteristics of TSE from Various Treatment Plants*

No.	COD (mg/L)	TP (mg/L)	Reference
1	55	1.32	(He & Xue, 2010)
2	49.7	11.5	(Órpez et al., 2009)
3	100	5.6	(Arbib, Ruiz, Alvarez-Díaz, Garrido-Perez, & Perales, 2013)
4	24.5	0.08	(Xin, Hong-ying, & Jia, 2010)
5	24	0.5	(Yang et al., 2011)

Therefore, the quality of TSE need to be improved in order to reuse it in more sectors and applications (Jasim et al., 2016).

1.2. Wastewater treatment

Due to the improvement of life on all levels, the demand of water continuously increases. In Arab countries, only 60% of the total volume of the generated wastewater and industrial wastewater of 10.85 km³/yr is treated and only one-third of this treated effluent was reused (Abdel-Dayem, Taha, & Choukr-Allah, 2012). Some studies showed that if 80% of the treated wastewater was reused, this could potentially address the water scarcity in Arab region (Abdel-Dayem et al., 2012; Jasim et al., 2016). Therefore, the treatment of wastewater is an appropriate solution for the growing water scarcity problem. The type of wastewater and the degree of treatment are determined

based on the required water quality (Jasim et al., 2016). The direct disposal of wastewater in ocean, seawater and river has strong environmental impacts on marine life as it decreases the amount of dissolved oxygen and it increases algal growth (Cerqueira & Marques, 2012; Oron et al., 2006). Therefore, the improvement of wastewater treatment methods and applications is necessary in order to reduce the environmental issues related to wastewater handling and disposal (Al-Hammad, Abd El-Salam, & Ibrahim, 2014; Oron et al., 2006). The aim of wastewater treatment is to remove the contaminants to meet the end-user requirements. The main constituents of wastewater include either physical, chemical and biological contaminants such as solids, nutrients such as nitrogen and phosphorous, heavy metals, organic matter that cause the depletion of dissolved oxygen, pathogenic organisms, color and odor.

Wastewater treatment consists of multiple unit operations which usually include more than one of treatment category which are physical, chemical and biological treatment categories (Moussa, El-Naas, Nasser, & Al-Marri, 2017).

- Physical treatment:

Physical treatment process targets the removal of solids by physical separation without causing any change in the wastewater characteristics. Examples of physical processes such as screening, filtration and sedimentation.

- Chemical treatment

Chemical treatment process targets the removal of suspended and dissolved contaminants by addition of chemicals. However, these treatment processes are less attractive due to the additional costs and the way for chemicals disposal. Examples of chemical processes such as coagulation and flocculation, ion exchange and adsorption.

- Biological treatment

Biological treatment process targets the biodegradation of organic matters and nutrients from wastewater by utilizing microorganisms. Biological processes can be classified according to the availability of dissolved oxygen present in wastewater. Thus, aerobic process is a process that needs the presence of oxygen, while anaerobic process is carried in the absence of oxygen. Examples of biological unit operations such as rotating biological contractors, trickling filters and activated sludge. In biological treatment process, the removal efficiency of phosphorous is not exceed 30%, and it requires post-treatment to remove the remaining phosphorous. Therefore, biological treatment is not the appropriate solution for the removal of phosphorous from the treated effluent (Bektas, Akbulut, Inan, & Dimoglo, 2004).

Table 2. *Wastewater Treatment Unit Operations (Moussa et al., 2017)*

Unit Operations	
Physical treatment	<ol style="list-style-type: none"> 1. Screening 2. Filtration 3. Sedimentation
Chemical treatment	<ol style="list-style-type: none"> 1. Coagulation and flocculation 2. Electrocoagulation 3. Ion exchange 4. Adsorption
Biological treatment	<ol style="list-style-type: none"> 1. Rotating biological contractors 2. Trickling filters 3. Activated sludge

1.2.1. Chemical Oxygen Demand reduction

Chemical oxygen demand treatment is the degradation of the organic matters present in wastewater. COD is the amount of dissolved oxygen required by microorganisms to degrade the organic wastes aerobically. Organic wastes could be biodegradable or non-biodegradable organic compounds or inorganic compounds and it comes either from natural source or from pollution. According to Federal Clean Water

Act, COD is considered as a conventional pollutant and it is used as an indication of the efficiency of the wastewater treatment process (Moreno-Casillas et al., 2007). Treated effluent standard of Qatar for COD is 50 mg/L (Ashghal, 2005). While the average COD value present in raw wastewater is 500 mg/L as mentioned by Ashghal standards (Ashghal, 2005).

1.2.2. Total Phosphorous treatment

Wastewater with excess amount of phosphorous is responsible for many types of problems. Hence, the concentration of phosphorous in wastewater is critical since at low concentration, it will not affect the human being because it presents in most organisms and cells (Kamaraj, Ganesan, Lakshmi, & Vasudevan, 2013). However, at high concentration it will cause problems. The industrial wastewater with excess phosphate can cause fouling of the pipes (Kamaraj et al., 2013). As well as, this phosphate is one of the natural nutrients that should be removed in order to avoid eutrophication (Tchamango, Nanseu-njiki, Ngameni, Hadjiev, & Darchen, 2010). Eutrophication is the enrichment of water bodies with nutrient that causes the dramatic growth of algae which will cause the depletion of oxygen. Consequently, the depletion of aquatic life. The main sources of phosphorous compounds are croplands and cattle. Each country has a strict limit on the discharging wastes containing phosphorous (Kamaraj et al., 2013). The forms of total phosphorous in wastewater are orthophosphates, condensed phosphates and organic phosphates (Kamaraj et al., 2013). The average TP value present in wastewater is 8 mg/L, while the treated effluent should not exceed 1 mg/L as set by Qatar's standard (Ashghal, 2005).

However, electrocoagulation is preferred over the conventional methods as it is characterized by ease in solid-liquid separation as it produces heavy flocs, high removal efficiency of the pollutants for a wide range of wastewater types with the production of

low sludge wastes, inexpensive, compact facility and no additional supply of chemicals (Aoudj, Khelifa, Drouiche, Hecini, & Hamitouche, 2010; Emamjomeh & Sivakumar, 2009; Hakizimana et al., 2017; Karamati-Niaragh, Moghaddam, Emamjomeh, & Nazlabadi, 2019; Mouedhen, Feki, Wery, & Ayedi, 2008). In addition, multiple studies showed the effectiveness of electrocoagulation as an alternative of disinfection (Ghernaout, Badis, Kellil, & Ghernaout, 2008; Jianga, Graham, Andre, Kelsall, & Brandon, 2002).

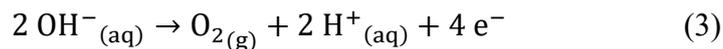
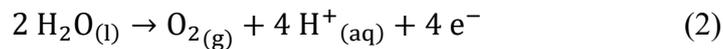
1.2.3. Electrocoagulation treatment process

Electrocoagulation consists of two metal electrodes (known as anode and cathode) that are submerged in wastewater and the electrodes are connected to a power supply for certain electrolysis time. The passing current causes the dissociation of metals on electrodes into metal ions (Didar & Islam, 2017). These metal ions induces three stages which are formation of coagulants, destabilization and neutralization of contaminants and suspended colloids, and aggregation of these destabilized colloids forming flocs that can be settled down by gravity (A. K. Chopra, Sharma, & Kumar, 2011; Eyvaz, Gürbulak, Kara, & Yüksel, 2014; Farhadi, Aminzadeh, Torabian, Khatibikamal, & Alizadeh Fard, 2012; Impa, Nagarajappa, Krishne Gowda, & Manjunath, 2015; Kuokkanen, Kuokkanen, Rämö, & Lassi, 2013; Mohammad Y A Mollah et al., 2004; Ozyonar & Karagozoglu, 2011). In electrocoagulation, the most two commonly used electrode materials are aluminum (Al) and iron (Fe). The advantages of Al and Fe as electrode materials are their availability and non-toxicity as they form non-toxic precipitants (Hakizimana et al., 2017; Safari, Aghdam, & Kariminia, 2016). Destabilization of contaminants and colloidal suspensions can be achieved by the interaction between aluminum ions generated from the oxidation of the anode and the negative colloids present in wastewater resulting in high positive ions

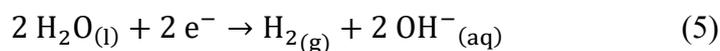
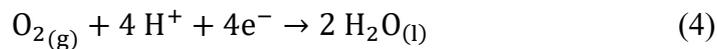
concentration around the negative colloid that tends to reduce the electrical double layer thickness and the electrostatic repulsive force, which eventually forming flocs (Eyvaz et al., 2014). These formed flocs can be removed either by settled down by gravity or deposited on the cathode surface or rise up to the surface by the generated hydrogen gas from the cathode (Canizares, Carmona, Lobato, Martinez, & Rodrigo, 2005; G. Chen, 2004; Eyvaz et al., 2014; Hakizimana et al., 2017).

Irdemez et al. (2006) showed that as the aluminum and iron present in wastewater different precipitates would be formed based on pH. At $\text{pH} < 6.5$, inorganic insoluble compounds will be formed. However, at $\text{pH} > 6.5$, metal hydroxide will be formed (Irdemez, Yildiz, & Tosunoğlu, 2006). The main reactions that takes place in EC in case of aluminum electrodes are as follows (Lee, Lewis, & Ashman, 2013):

Anode reactions:

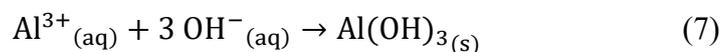


Cathode reactions:



Based on the pH of the electrolyte, the precipitation reaction are as follows:

Acidic condition:



Alkaline condition:



Where $\text{Al}(\text{OH})_{3(\text{s})}$ is an amorphous compound having bulk structure allowing it to settle easily (Aoudj et al., 2010; Eyvaz et al., 2014; Farhadi et al., 2012; Hakizimana et al., 2017; Karamati-Niaragh et al., 2019; Mohammad Y A Mollah et al., 2004; Ozyonar & Karagozoglu, 2011). For settling process, the heavier the flocs the higher the settling velocity (Zodi, Potier, Lopicque, & Leclerc, 2009).

1.2.4. Surface Charge and Destabilization

The sign of the surface charge of most colloids in wastewater is negative (Gregory & Duan, 2003; Neihof & Loeb, 1972). The stability of the colloidal system is as a result of the colloidal particles carry similar charge, therefore they repel each other and remain suspended. There is no net charge of the colloidal system as the negative colloidal particles are balanced by the positive ions from the surrounding medium forming electrical double layer (Ghernaout, Naceur, & Ghernaout, 2011). Colloidal particle has the maximum potential at its surface and its potential decreases through the inner layer as moving away from the surface, where the zeta potential can be measured (Ghernaout et al., 2011). Zeta potential represents the potential difference between the outer layer and the bulk medium. Moreover, it determines the stability of the colloidal system. Therefore, the higher the zeta potential, the larger the electrostatic repulsive force and the more stable of colloidal system (Duman & Tunç, 2009).

The electrical double layer consists of two layers which are inner layer and outer layer. Inner layer is known as stern layer or Helmholtz layer where the positive ions are strongly attached to the surface of the colloidal particles. While the outer layer is known as slipping plane or diffuse layer where the positive ions are loosely attached (Davis, 2010; Ghernaout et al., 2011). The electrical double layer illustrated in Figure 1.

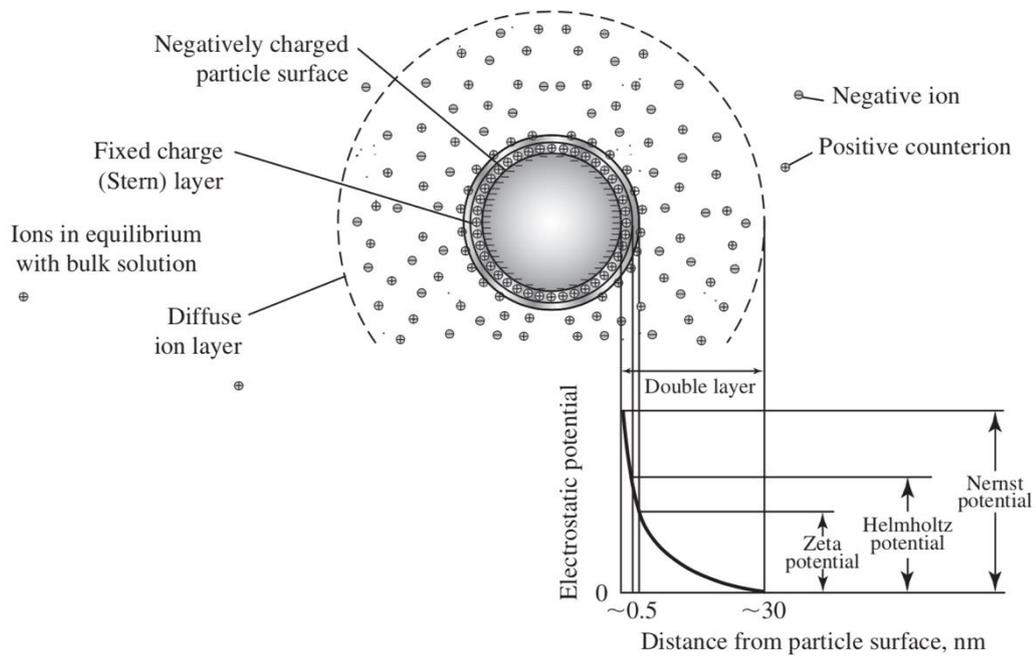


Figure 1. Electrical double layer (Davis, 2010).

Destabilization of contaminants and colloidal suspensions can be achieved by the interaction between aluminum ions generated from the oxidation of the anode and the negative colloids present in wastewater resulting in high positive ions concentration around the negative colloid that tends to reduce the thickness of the electrical double layer and the electrostatic repulsive force, which eventually forming flocs (Eyvaz et al., 2014). These formed flocs can be removed either by settle down by gravity or deposited on the cathode surface or rise up to the surface by the generated hydrogen gas from the cathode (Eyvaz et al., 2014; Hakizimana et al., 2017).

1.3. Dielectrophoresis Force

Dielectrophoresis force is the movement of free particle by dielectric polarization in an inhomogeneous electric field (Hawari, Du, Baune, & Thöming, 2015). Placing a particle suspended in an electrolyte in an inhomogeneous electric field causes a redistribution of the charges inside the particle and the medium at their interface; This redistribution depends on the polarizability of the particle and the medium. Additionally, this distribution leads to a difference in the charge density at each ends of the particle which leads to an induced dipole. Induced dipole can be represented by different forces at each ends of the particle at which the difference between these two forces gives a net force called dielectrophoresis force (Çetin & Li, 2011). Therefore, the dielectrophoresis force applied on spherical particle can be calculated using the following equation:

$$F_{DEP} = 4\pi a^3 \epsilon_0 \epsilon_M \text{re}[\tilde{K}] (\mathbf{E} \cdot \nabla) \mathbf{E} \quad (9)$$

Where, a is the particle radius, ϵ_0 is the permittivity of free space ($8.854 \times 10^{-12} \frac{F}{m}$), ϵ_M is the absolute permittivity of the medium, \mathbf{E} is the field intensity ($\frac{V}{m}$) and $\text{re}[\tilde{K}]$ is the Clausius-Mossotti (CM) factor, which can be calculated by (Hawari et al., 2015):

$$\tilde{K} = \frac{\tilde{\epsilon}_p - \tilde{\epsilon}_M}{\tilde{\epsilon}_p + 2\tilde{\epsilon}_M} \quad (10)$$

$$\tilde{\epsilon} = \epsilon - \frac{j\sigma}{\omega} \quad (11)$$

When AC power supply is used then all permittivity will be replaced with complex permittivity (Çetin & Li, 2011). Where, $\tilde{\epsilon}_p$ is the complex permittivity of the particle, $\tilde{\epsilon}_M$ is the complex permittivity of the medium, $\tilde{\epsilon}$ is the complex permittivity, σ is the conductivity ($\frac{S}{m}$), ω is the angular frequency ($\frac{rad}{s}$), j is the geometric gradient of the square of electric field (\mathbf{E}), which can be calculated by (Hawari et al., 2015):

$$j = \sqrt{-1} \cdot (\mathbf{E} \cdot \nabla)\mathbf{E} = \frac{1}{2} \nabla |\mathbf{E}|^2 \quad (12)$$

Based on the permittivity of the particle and medium, the suspended particles can present different DEP effects in which it can move in different directions, these effects are positive DEP (pDEP) and negative DEP (nDEP). If the permittivity of the particles is higher than the medium, then the particles will be attracted by stronger electric field, and it will present pDEP. Whereas, if the permittivity of the particles is lower than the medium, then the particles will be attracted by weaker electric field, and it will present nDEP (Fei Du, Baune, Kück, & Thöming, 2008; Hawari et al., 2015). The high conductivity of TSE suggests that, the particles in treated sewage effluent (TSE) will exert nDEP because of the permittivity of the suspended will be lower than the medium (TSE) (Hawari et al., 2015). As mentioned in previous study, the DEP force will affect all suspended particles regardless of their electrical charge (F. Du, Hawari, Baune, & Thöming, 2009).

CHAPTER 2: PREVIOUS STUDIES OVER ELECTROCOAGULATION PROCESS

Aoudja et al. (2010) used electrocoagulation for the removal of dyes from textile industrial wastewater with DC power supply. The cathode and anode are made up of aluminum plates. A current density of 18.75 A/m^2 with a treatment period of 60 minutes resulted in a removal percent of 98% of dyes (Aoudj et al., 2010). Asselin et al. (2008) focused in their study on the effect of different arrangement of electrodes and electrode material (aluminum and iron) on the removal of oil and grease, BOD, soluble COD, total COD, total suspended solids, total solids and turbidity from poultry slaughterhouse effluent by means of electrocoagulation with DC power supply. The unit consisted of eight parallel electrode plates connected in two different arrangement once with monopolar electrode system (MP) and the other with bipolar electrode system (BP). Where in the monopolar modes all eight electrodes are connected to the power source, however in the bipolar connection only the two outer electrodes are connected to the power source, while the inner ones are not. They used a current density of 27.273 A/m^2 with different time of application (60 or 90 minutes). The results showed that the steel BP system is better than MP with a removal percent of oil and grease, BOD, soluble COD, total COD, total suspended solids, total solids and turbidity of 90%, 99%, 86%, 50%, 82%, 89%, 64% and 90%, respectively (Asselin, Drogui, Benmoussa, & Blais, 2008). Makwana et al. (2017) targeted in their study the reduction in COD, turbidity and phosphate from anaerobically treated urban wastewater by means of electrocoagulation with DC power supply. The unit consisted of two aluminum cathode and anode plates operated with a current density of 150 A/m^2 for 18 minutes of treatment time which resulted in 98.4% turbidity removal, 70.9% COD reduction and 99% phosphate removal (Makwana & Ahammed, 2017). Saleem et al. (2011) targeted

in their study the removal of turbidity, COD and TSS from municipal wastewater to meet irrigation standards by using electrocoagulation with DC power supply. A setup consists of circular cell with two iron symmetrical sheets that operated with a current density of 247 A/m^2 for 30 minutes of treatment. EC produced an effluent with 91.8% turbidity removal, 77.2% COD reduction and 68.5% TSS removal. The quality of treated wastewater was found to be within the standards for irrigation and plantation (Saleem et al., 2011). Ozyonar et al. (2011) targeted the reduction of COD, turbidity and phosphate from domestic wastewater by electrocoagulation and the effect of electrode material on the performance of the electrocoagulation process. The unit consists of four electrodes that were set in monopolar and parallel in the cell. This study was operated once with four aluminum electrodes and the other with iron. A current density of 100 A/m^2 with only 10 min of treatment time resulted in 75% COD reduction, 98% turbidity removal, and 98% phosphate removal. At the end of this study, the aluminum electrodes were preferred than iron due to higher removal percent were obtained in case of aluminum (Ozyonar & Karagozoglu, 2011). Nguyen et al. (2016) targeted in their study the removal of phosphorous under different conditions such as electrolysis time, conductivity and initial phosphorous concentration from municipal wastewater by means of electrocoagulation. The electrochemical unit consist of two iron electrodes which were placed in concentric cylindrical shapes. It was found that high conductivity ensures efficient removal percent and at the same time it reduces the overall cost and the retention time of the process (D. Duc Nguyen et al., 2016). Durici et al. (2016) studied the removal of phosphate from synthetic wastewater by electrocoagulation using either aluminum or iron electrodes. The unit consists of anode and cathode sheets with the same surface area. A current density of 10 A/m^2 was used with 40 minutes treatment time resulted in 98.9% and 93.5% phosphate removal for

aluminum and iron electrode, respectively (Đuričić, Malinović, & Bijelić, 2016). Chopra and Sharma (2013) studied the effect of different electrodes combination on the removal of turbidity, reduction in COD and BOD from secondary treated sewage by electrocoagulation with DC power supply. The electrocoagulation unit consists of cylindrical reactor and two electrode plates with two different combination (Anode/Cathode: Fe/Al and Al/Fe). A voltage of 12 V was applied for 30 minutes which resulted in 81.5% turbidity removal, 74.3% COD reduction and 70.8% BOD removal for the Al/Fe combination, while for the Fe/Al combination it resulted in 71.1% turbidity removal, 64.9% COD reduction and 61.8% BOD reduction (Ashok Kumar Chopra & Sharma, 2013). Chen et al. (2000) showed the effect of electrode material (aluminum and iron) on the removal of oil and grease from restaurant wastewater by electrocoagulation using DC power supply. The electrochemical unit consists of five electrodes connected in a dipolar electrode system. A current density ranged between 60 to 80 A/m² resulted in a removal percent of 94% of oil and grease (X. Chen, Chen, & Yue, 2000). Kobya et al. (2011) studied the effect of different electrode modes and electrode material (aluminum and iron) on the removal of arsenic from potable water using electrocoagulation. The electrodes are arranged in three different modes: parallel and monopolar (MP-P), series and monopolar (MP-S) and series and bipolar (BP-S). A current density of 2.5 A/m² was applied for different treatment time. MP-S gives the best removal percent of arsenic than the other two arrangements with a removal percent of 94.1% of arsenic for Fe within only 2.5 minutes electrode and 93.5% removal for Al electrode within 4 minutes (Mehmet Kobya, Ulu, Gebologlu, Demirbas, & Oncel, 2011). Ghosh et al. (2008) focused in their study on the effect of different arrangement of electrodes on the removal of fluoride using electrocoagulation. The electrochemical unit in case of monopolar mode consists of two aluminum sheets, while in case of

bipolar mode it consists of four aluminum electrodes. Where only the two outer electrodes are connected to the power source and the inner electrodes will act as a secondary cell. A current density of 625 A/m^2 was applied for 30 minutes resulted in a removal percentage of 90 % of fluoride (Ghosh, Medhi, & Purkait, 2008). Fouad et al. (2009) showed in their study the effect of different arrangements of electrodes on the removal of oil using electrocoagulation. The unit in case of horizontal oriented electrodes consists of array of separated horizontal electrodes while in case of conventional vertical mode it consists of two parallel aluminum sheets. In addition, the inner side of the horizontal electrodes would act as a heat exchanger if necessary. A current density of 80 A/m^2 was applied for 30 minutes. The results showed that the horizontal oriented mode is better than the conventional vertical ones with a removal percent of 99.8% of oil (Fouad, Konsowa, Farag, & Sedahmed, 2009). Murthy and Parmar (2012) studied the effect of electrode material (stainless steel and aluminum) on the removal of mercury from aqueous solutions by means of electrocoagulation. The unit consists of two parallel electrodes. A current density of 177 A/m^2 for 15 minutes of treatment. At the end of study the stainless steel electrodes gave a better percentage removal of mercury than the aluminum electrodes with a percentage of 99.7% and 98% removal of mercury, respectively (Murthy & Parmar, 2012). Franco et al. (2017) focused in their study on the removal of phosphate from wastewater and surface water by means of electrocoagulation. The unit consists of two parallel aluminum electrodes connected to a direct current power supply. The current was applied for 1 hour showed the ability of electrocoagulation insufficient removal of phosphate from both wastewater and surface water with a removal percentage of 99% of phosphate (Franco, Lee, Arbelaez, Cohen, & Kim, 2017). Attour et al. (2014) studied the influence of different operating parameters on the removal of phosphate from a phosphate synthetic

solution by electrocoagulation. The unit consists of two vertical aluminum electrodes connected to a direct current power source. It was found that the same treatment efficiency was obtained with high current density and shorter electrolysis time or with low current density with longer electrolysis time (Attour et al., 2014). Irdemez et al. (2006) studied the effect of different operating parameters on the removal of phosphate from phosphate synthetic solution by means of electrocoagulation using predicted and experimental approach. Taguchi method was used to optimize the removal of phosphate ions under optimum operating parameters. It was found that the percentage removal of phosphate reached 100% and the observed and experimental phosphate removal percent are close to each other. Therefore, the Taguchi model may be adequate to know the effect of operating parameters in the removal percent (Irdemez et al., 2006). Most of the previous studies showed that electrocoagulation could achieve high removal percentages of contaminants from different types of wastewater.

Electrocoagulation can be operated with either direct current (DC) or alternating current (AC) power supply. Vasudevan et al. (2012) targeted the removal of cadmium by electrocoagulation using direct current and alternating current power supply. The results showed a removal percentage of cadmium of 98.1% and 97.3% for alternating current and direct current sources, respectively. Also, they characterized electrodes surface by scanning electron microscopy (SEM), and it was found that the surface of electrodes in case of alternating current had smooth microstructure, while the surface of the electrodes in case of the direct current was found to be rough (S. Vasudevan & Lakshmi, 2012). Kamaraj et al. (2013) showed the effects of direct and alternating current on the removal of copper by electrocoagulation. The unit consists of magnesium alloy electrodes sheets. A current density of 0.025 A/dm^2 was used. It was found that a removal percentage of copper of 97.8 and 97.2 % for alternating current and direct

current, respectively (Kamaraj et al., 2013). Eyvaz et al. (2009) aimed to study the effect of direct current (DC) and alternating pulse current (APC) on the removal of two different types of dye from aqueous solution. The unit consists of two pair of aluminum electrodes connected in monopolar parallel mode. A current density of 105 A/m^2 was applied for different treatment time. At the end of this study, it was found that higher removal percent of dyes and TOC were obtained by using APC than DC in shorter electrolysis time (Eyvaz, Kirlaroglu, Aktas, & Yuksel, 2009). All previous studies agreed on adopting alternating current in electrocoagulation process in order to overcome the formation of an impermeable oxide layer on the surface of the cathode and the corrosion that occurs on the surface of the anode due to oxidation when direct current (DC) is used. These phenomena will affect the transport of current and consequently it will affect the treatment process. Therefore, reasonable life of electrode is ensured by using AC power source (Eyvaz et al., 2009; S. Vasudevan & Lakshmi, 2012). The following Table 3 summarizes some of the previous studies:

Table 3. *Previous Studies Related to Electrocoagulation Process*

Types of wastewater	Water pollutants	Operating mode	Electrode material	Source of current	Current density (mA/cm ²)	Removal% of targeted pollutants	Reference
Textile industry	Dyes	Batch	Al	DC power supply	1.875	98%	(Aoudj et al., 2010)
Poultry slaughterhouse effluent	Oil and grease, BOD, soluble COD, total COD and	Batch	Al/Fe (Bipolar/Monopolar)	DC power supply	2.727	Best performance Fe/BP with (90%, 99%, 86%, 50%, 82%, 89%, 64% and 90%, respectively)	(Asselin et al., 2008)

total
suspended
solids

Anaerobically treated urban wastewater	Turbidity, phosphate and COD	Batch	Al	DC power supply	15	98.4%, 70.9% and 99.0%, respectively	(Makwana & Ahammed, 2017)
Municipal wastewater	TSS, turbidity and COD	Batch	Fe	DC power supply	24.7	68.5%, 91.8% and 77.2%, respectively	(Saleem et al., 2011)

Domestic wastewater	COD, turbidity and phosphate	Batch	Al/Fe (Monopolar and parallel)	DC power supply	10	Aluminum is preferred with (75%, 98% and 98%, respectively)	(Ozyonar & Karagozoglu, 2011)
Synthetic wastewater	Phosphate	Batch	Al/Fe	DC power supply	1	98.9% and 93.5% for aluminum and iron electrode, respectively	(Đuričić et al., 2016)
Secondarily treated sewage (STS)	Turbidity, COD and BOD	Batch	(Anode/cathode: Fe/Al and Al/Fe)	DC power supply	12V	Al/Fe is preferred with 81.5%, 74.3% and 70.8%, respectively	(Ashok Kumar Chopra & Sharma, 2013)

Synthetic wastewater	Mercury	Batch	Al/stainless steel	DC power supply	17.7	Stainless steel is preferred with 99.7%	(Murthy & Parmar, 2012)
Drinking water	Fluoride	Batch	Al	DC power supply	62.5	90%	(Ghosh et al., 2008)
Wastewater	Cadmium	Batch	Fe	AC/DC power supply	2	98.1 and 97.3% were achieved for DC and AC power source	(S. Vasudevan & Lakshmi, 2012)
Wastewater	Copper	Batch	Magnesium alloy	AC/DC power supply	0.25	97.8 and 97.2 % were achieved for AC and DC power source	(Kamaraj et al., 2013)

Aqueous solution	TOC and dye	Batch	Al	AC/DC power supply	10.5	RY TOC removal is 57% and dye removal 63% and for DY TOC removal is 89% and dye removal 99.9% for AC power source	(Eyvaz et al., 2009)
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Most of these past studies have been focused on the treatment of highly polluted wastewater, such as restaurant wastewater, poultry slaughterhouse effluent and municipal wastewater using DC power supply, which motivates the study of the effectiveness of EC in post-treating the treated secondary effluent using a novel electrode configuration with AC power source taking into consideration the environmental concerns.

The aim of this study is to enhance TP removal efficiency and COD reduction for TSE by the application of the DEP force. The DEP forces are generated by a novel electrode configuration of unsymmetrical aluminum cylindrical electrodes with alternating current (AC) power source. The impact of electrode configuration and power supply type on TP removal efficiency and COD reduction were evaluated in this combined laboratory scale and simulation study.

CHAPTER 3: EXPERIMENTAL METHODS AND PROCEDURE

Due to the shortage in freshwater around the world especially in middle east, this study was developed for the production of treated water. This study aims to investigate the optimum operating parameters to treat TSE.

3.1. Experimental set-up

Figure 2 shows an electrochemical batch reactor with a capacity of 2.5 L made from Borosilicate Glass. Constant mixing was provided by a magnetic stirrer used at a speed of 4. Two aluminum electrodes are placed in concentric cylinders with radii of 1.25 and 2.25 cm which not completely immersed in TSE. The distance between electrodes is 1 cm. AC-DEP module can be seen in Figure 2(b). The dimensions of the inner aluminum electrode are 2.5 cm internal diameter \times 5 cm high with an effective area of 117.8 cm². The outer aluminum electrode is with the dimensions of 4.5 cm internal diameter \times 15 cm high with an effective area of 551.4 cm². For both AC and DC modules, the experimental setup can be seen in Figure 2(a), the dimensions of the inner aluminum electrode are 2.5 cm internal diameter \times 15 cm high with an effective area of 274.9 cm² and the outer aluminum electrode are with the dimensions of 4.5 cm internal diameter \times 15 cm high with an effective area of 551.4 cm². In both AC-DEP and AC modules, the electrodes are connected to a VARIAC variable transformer (VARIAC, Cleveland, Ohio) to deliver a voltage in a range of (0 to 250 V) at a constant frequency of 50 Hz. Moreover, for the measurements of current and voltage, an oscilloscope (MDO3024, Tektronix, Beaverton, Oregon) was used. The type of wave that was shown in the oscilloscope screen is sine wave. Usually AC power supply generates sine wave with time rely. The time rely concept is to shift the function of the anode and cathode occasionally (Eyvaz et al., 2009). While, for DC module, the electrodes are connected to a DC power supply (GPC-M SERIES). Figure 3 shows the

setup of electrocoagulation used in the lab.

After each run the electrodes are rinsed with water to make sure that the electrodes are free of any contaminants and then both electrodes are dried and weighed. For inner electrode, Mettler Toledo AG204 Analytical Balance is used and for outer electrode, Citizen CY-1003 Precision Balance is used.

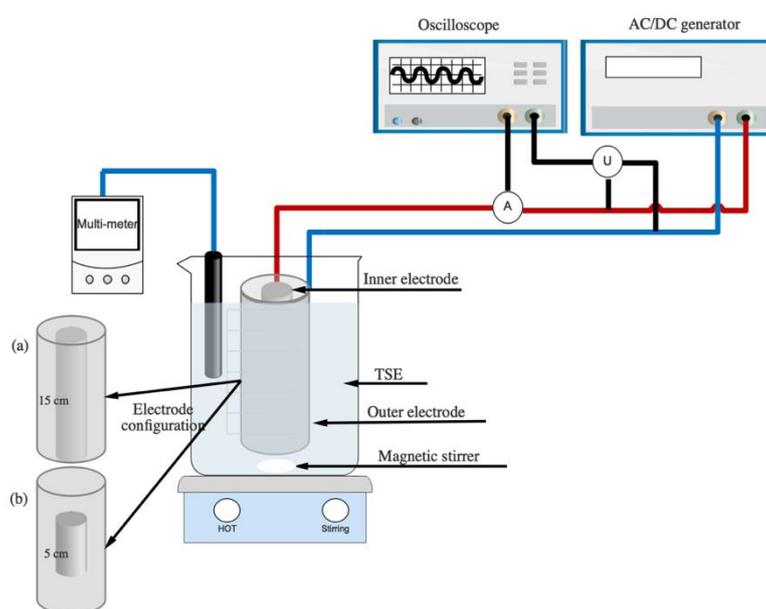


Figure 2. Schematic diagram of electrode configuration with AC/DC power supply.

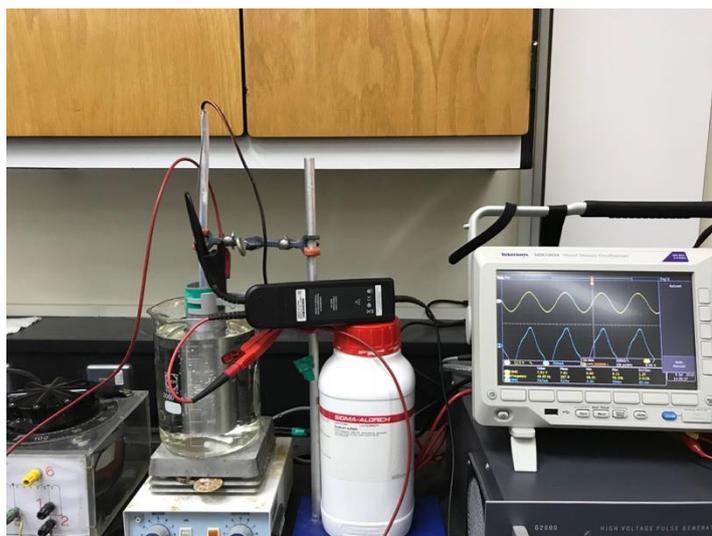


Figure 3. Setup of electrocoagulation system used in the lab.

3.2. Experimental Procedure

For each run, 2130 mL of Treated Sewage Effluent was placed in a beaker. All experiments were done at ambient temperature 20 °C. For the three modules, three different runs were conducted each with different applied current density (0.8, 2.6 and 4.3 mA/cm²) in order to compare between the three modules. The current was applied for 30 minutes electrolysis time with continuous mixing. For each run at specific current density, initial pH, conductivity and voltage were recorded. The samples were taken at three different time, which are 5 minutes, 10 minutes and 30 minutes. For each sample, pH, conductivity, current density and voltage were recorded. The electrodes surface were rinsed and dried after each experiment to make sure that there is nothing attached to the surface. Then the samples were allowed to settle for 24 hours and then analyzed for COD and TP content.

3.3. Experimental Analysis

Table 4. *Standard Methods Used for the Examination of Wastewater Characteristics*

Measured parameter	Standard method
Conductivity (mS/cm)	APHA 2510 B. Conductivity
pH	APHA 4500-H+ B. Electrometric Method
Temperature (°C)	APHA 2550 TEMPERATURE
Turbidity (NTU)	APHA 2130 B. Nephelometric Method
COD (mg/L)	APHA 5220 D. Closed Reflux, Colorimetric Method
TSS (g)	APHA 2540 D. Total Suspended Solids Dried at 103– 105°C
TDS (mg/L)	APHA 2540 C. Total Dissolved Solids Dried at 180°C
TP (mg/L)	1. APHA 4500-P C. Vanadomolybdophosphoric Acid Colorimetric Method 2. APHA 4500-P E. Ascorbic Acid Method
NH ₄ (mg/L)	ASTM D 1426 – 03 Standard Test Methods for Ammonia Nitrogen In Water

After each run, all samples were allowed to settle for 24 hours, then all samples should be passed through a 0.45 μm Millipore filter to remove all suspended solids. Then the filtrate was used for both COD and TP tests.

3.3.1. COD measurement

For COD test, this test is based on standard dichromate digestion method and using Orion AquaMate UV-VIS Spectrophotometer (Waltham, Massachusetts) to estimate COD in post-treated TSE.

After measuring the initial and final COD for each sample, the COD reduction percentage is calculated using the following equation:

$$\% \text{ COD reduction} = \left(\frac{\text{COD}_{\text{initial}} - \text{COD}_{\text{final}}}{\text{COD}_{\text{initial}}} \right) \times 100$$

3.3.2. TP measurement

TP test was done on the basis of digestion method in which all forms of phosphorous converted into dissolved orthophosphate and then using Orion AquaMate UV-VIS Spectrophotometer to estimate the concentration of orthophosphate in the post-treated TSE.

The concentration of TP is calculated using the following equation:

$$\text{TP}_{\text{concentration}} \left(\frac{\text{mg}}{\text{L}} \right) = \frac{\text{measured value} * 0.05 \text{ L}}{0.035 \text{ L}}$$

After calculating the TP concentration then the percentage removal can be calculated using the following equation:

$$\% \text{ TP removal} = \left(\frac{\text{TP}_{\text{initial}} - \text{TP}_{\text{final}}}{\text{TP}_{\text{initial}}} \right) \times 100$$

3.3.3. *Electrode corrosion*

Electrode corrosion were calculated for the three different modules. For each module, after each run the cylindrical aluminum electrodes are rinsed with water to make sure that the electrodes are free of any contaminants and then both electrodes are dried and weighted. After all runs, electrode corrosion was calculated by subtracting the mass of the electrode after a specific run from the mass of the electrode before the same run which expressed in grams.

The following three Figures (Figure 4, 5 and 6) show the unsymmetrical inner electrode, the symmetrical inner electrode and the outer electrode.



Figure 4. Unsymmetrical aluminum inner electrode for AC-DEP.



Figure 5. Symmetrical aluminum inner electrode for AC/DC.



Figure 6. Cylindrical aluminum outer electrode.

3.4. Wastewater characterization

An experimental analysis of a real secondary treated wastewater sample that was collected from a wastewater treatment plant in Doha and the characterization of wastewater is summarized in Table 5:

Table 5. *Characterization of Wastewater*

Measured parameter	Value
Conductivity (mS/cm)	5.1
pH	6.9
Temperature (°C)	22.2
Turbidity (NTU)	0.8
COD (mg/L)	206.3
TSS (g)	0
TDS (mg/L)	2816
TP (mg/L)	7.6
NH ₄ (mg/L)	0.5

3.5. Numerical methods

The dimensions of the aluminum cylindrical electrodes that were used in the simulation are shown in Figure 7. The square electric field for the inner and outer electrodes is simulated in order to check the efficiency of the used unsymmetrical

cylindrical electrodes using COMSOL Multiphysics software. The simulation was conducted in two dimensions only, assuming that the height of the electrodes going to infinity. The electric potential was solved at a set of boundary condition in order to calculate the electric field and the DEP force exerted on the particles. The quasi-electrostatic form was used in order to solve this problem, for the applied currents and frequencies. The root mean square (rms) of the electric field is given by:

$$E = -\nabla\varphi \quad (13)$$

Where, φ is the rms of the electrostatic potential which can be given by Laplace's equation (assuming that the medium is liquid only with the absence of any particles and it is homogeneous):

$$\nabla^2\varphi = 0 \quad (14)$$

Fixed boundary conditions are applied on the surface of the charge carrying electrodes which are the inner electrode and outer electrodes:

$$\varphi_1 = \frac{U_0}{2} \quad (15)$$

$$\varphi_2 = -\frac{U_0}{2} \quad (16)$$

Where, U_0 is the rms of the oscillating potential drop.

In order to ensure mesh-independent results, adaptive mesh refinement has been applied.

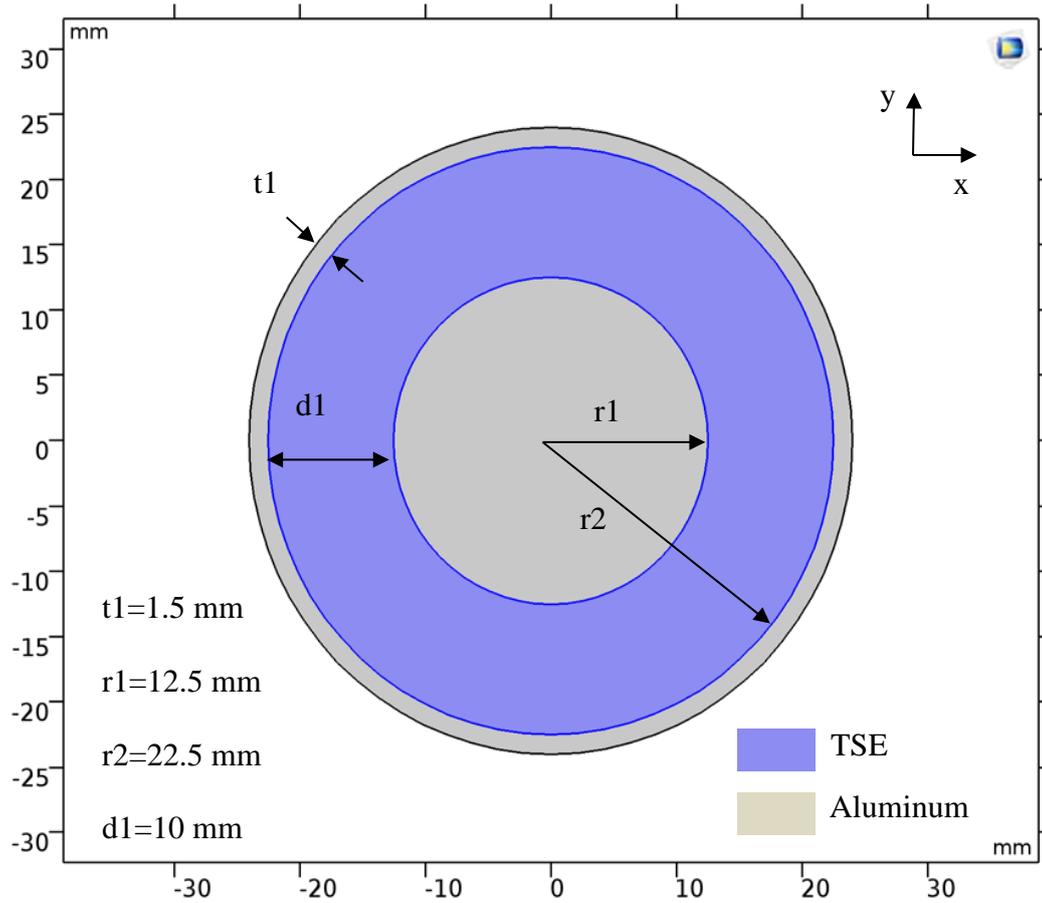


Figure 7. Sketch of the simulated geometrical parameters.

3.6. Error estimation

Electrocoagulation process for each of the three modules and each current density were repeated twice. Error bars represents the standard deviation of a data set (“Interpreting Error Bars,” n.d.). The following equation was used to calculate the standard deviation error bars for each reported data:

$$SD = \sqrt{\frac{\sum_1^N (x_i - \bar{x})^2}{N}}$$

Where, x_i : the measured value, \bar{x} : the average of the two measured value and $N=2$, the

number of replications. Error bars are depicted on all Figures. All standard deviation error bars do not exceed 5%.

CHAPTER 4: RESULTS AND DISCUSSION

The performance for the post-treatment of TSE using three different electrocoagulation modules (AC-DEP, AC and DC) were studied, for TP removal efficiency and COD reduction percent in terms of the impact of specific parameters such as current density and electrolysis time. After that the comparison between the performance of three modules was conducted in terms of the impact of electrode configuration and power supply type.

For each module (DC, AC and AC-DEP) and each current density (0.8, 2.6 and 4.3 mA/cm²), three samples are taken at different electrolysis time (5, 10 and 30 min). Electrocoagulation process for each of the three modules and each current density was repeated for two times. Therefore, the total number of samples for each module are 18 samples at which the total phosphorous removal percent and chemical oxygen demand reduction percent are studied.

4.1. Performance of DC module in the post-treatment of TSE

4.1.1. *Impact of current density*

TP removal and COD reduction percent were studied for symmetrical aluminum electrodes with DC power source. This is achieved to examine the impacts of the variation of current density on the removal of TP and COD reduction.

Figure 8 shows total phosphorous removal efficiency for three different current density (0.8, 2.6 and 4.3 mA/cm²) at three different electrolysis time (5, 10 and 30 min). As can be seen, for 5 min electrolysis time, 0.8 mA/cm² current density has the lowest TP removal efficiency with 7.8 %, while 4.3 mA/cm² current density has the highest TP removal efficiency of 32.9 % and TP removal efficiency for 2.6 mA/cm² current density was 24.2 %. As electrolysis time increases up to 10 min, TP removal percent for 0.8, 2.6 and 4.3 mA/cm² were obtained as 11.2, 26.9 and 40.1 %, respectively.

Finally, TP removal percent for 0.8, 2.6 and 4.3 mA/cm² at 30 min were obtained as 25.3, 33.9 and 46.0 %, respectively. Therefore, at a certain electrolysis time, the total phosphorous removal percent was increasing as current density increases. It was clearly seen that TP removal increases as current density increases due to when more alternating current passes through the inner electrode leads to more dissociation of inner electrode into metal ions which enhances the destabilization mechanism. These result is compatible with most of previous studies as those studies showed a direct relationship between current density and TP removal efficiency at a certain electrolysis time for DC power supply system (Attour et al., 2014; Lacasa, Cañizares, Sáez, Fernández, & Rodrigo, 2011; Shalaby, Nassef, Mubark, & Hussein, 2014; Tran, Drogui, Blais, & Mercier, 2012; Subramanyan Vasudevan et al., 2008).

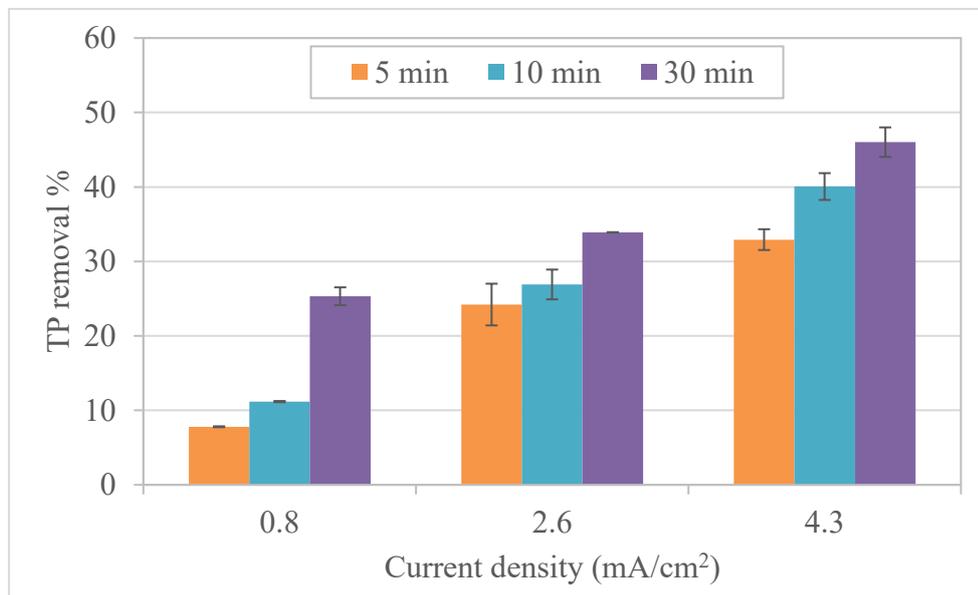


Figure 8 . Impact of current density on TP removal percent at three different electrolysis time for DC power source.

In Figure 9, three different current density were used to study the effect of current density on COD reduction which are 0.8, 2.6 and 4.3 mA/cm². At 5 min electrolysis time, COD reduction percent increases from 18.8 to 27.1 % as current density increases from 0.8 to 4.3 mA/cm². In addition, for 10 min and 30 min electrolysis time same trends were obtained. As for 10 min electrolysis time, COD reduction were obtained as 25.5, 32.9 and 45.1 % for 0.8, 2.6 and 4.3 mA/cm² current density, respectively. Similarly, for 30 min electrolysis time, COD were obtained as 24.2, 26.2 and 43.3 % for 0.8, 2.6 and 4.3 mA/cm² current density, respectively. Therefore, at a certain electrolysis time, COD reduction increases as current density increases. At high current density, more metal ions are generated, and the formation of flocs increases (Aoudj et al., 2010; Makwana & Ahammed, 2017). This result is compatible with (Dehghani, Seresht, & Hashemi, 2014; Ozyonar & Karagozoglu, 2011).

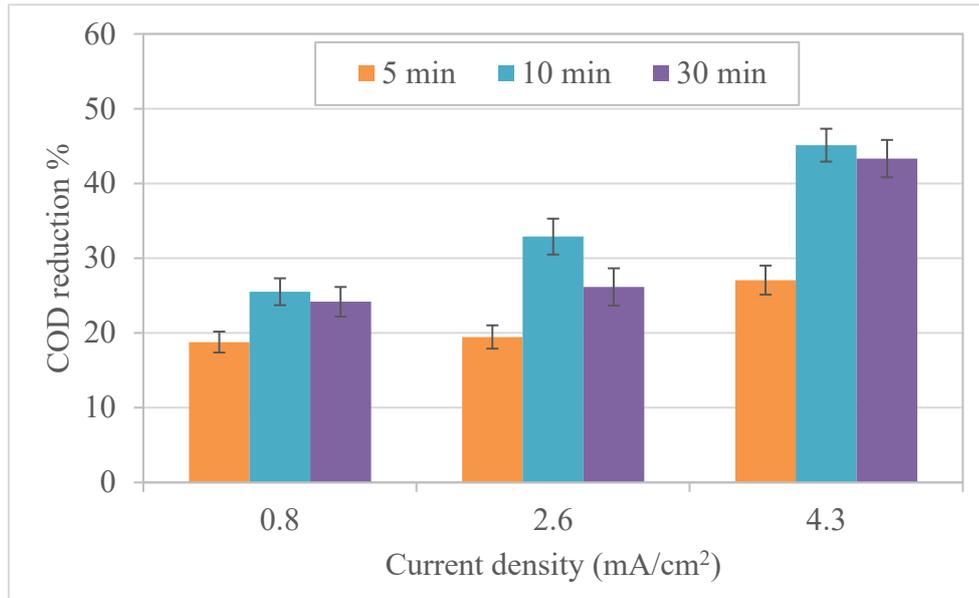


Figure 9. Impact of current density on COD reduction percent at three different electrolysis time for DC power source.

4.1.2. Impact of electrolysis time

The impact of the variation of electrolysis time on TP removal and COD reduction were examined for symmetrical aluminum electrodes with DC power source.

Figure 10 shows the variation of TP removal efficiency versus electrolysis time at three different current density (0.8, 2.6 and 4.3 mA/cm²). As can be seen, at current density of 0.8 mA/cm², TP removal percent was increasing from 7.8 to 25.3% as electrolysis time increased from 5 to 30 min. Similarly, for both remaining applied current density (2.6 and 4.3 mA/cm²), TP removal percent increases as electrolysis time increases. At current density of 2.6 mA/cm², TP removal percent was increasing from 24.2 to 33.9% as electrolysis time increased from 5 to 30 min. Moreover, at maximum applied current density of 4.3 mA/cm², TP removal percent increases from 32.9 to 46.0

% as electrolysis time increases from 5 to 30 min. Therefore, at maximum electrolysis time of 30 min, TP removal efficiency was improved for 0.8 mA/cm² current density by 17.5 %, for 2.6 mA/cm² current density by 9.7 % and for 4.3 mA/cm² current density by 13.1%. For TP removal, at any selected current density, the removal of TP was increasing as the electrolysis time increases. Longer electrolysis time produces more metal hydroxide flocs, so more TP will be adsorbed resulting in improvement of TP removal (Attour et al., 2014; Lacasa et al., 2011; Shalaby et al., 2014; Tian et al., 2018; Tran et al., 2012).

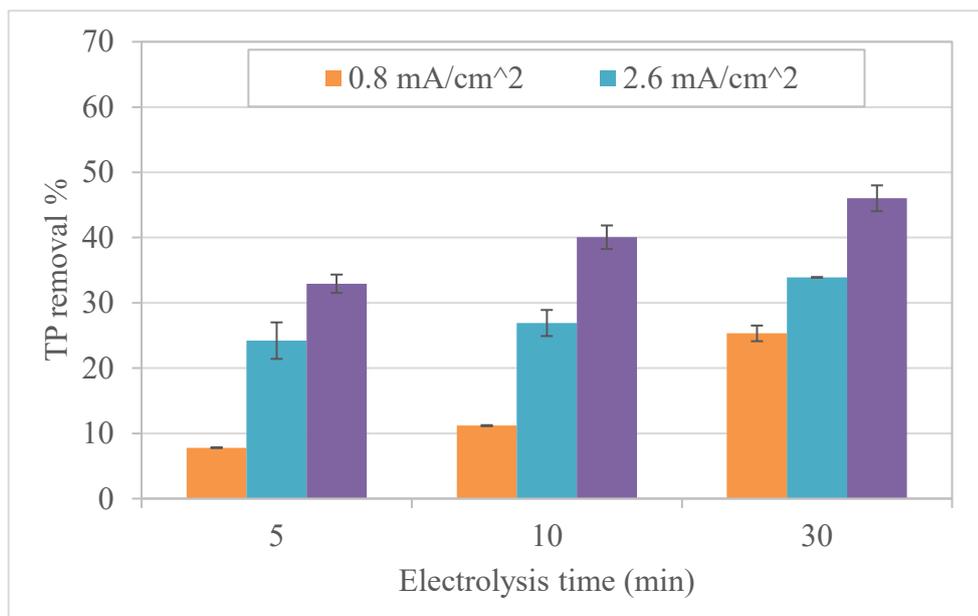


Figure 10. Impact of electrolysis time on TP removal percent at three different current density for DC power source.

Figure 11 shows the effect of the variation of electrolysis time on COD reduction percent at three different current density (0.8, 2.6 and 4.3 mA/cm²). It can be clearly seen that at 0.8 mA/cm² current density, COD reduction were obtained as 18.8, 25.5 and 24.2 % for 5, 10 and 30 min electrolysis time, respectively. Similar trends were achieved for both 2.6 and 4.3 mA/cm² current density. At a current density of 2.6 mA/cm², the maximum COD reduction percent was achieved at 10 min electrolysis time of 32.9 %, while the lowest COD reduction was achieved at 5 min electrolysis time of 19.5 %. For 4.3 mA/cm² current density, the sample that was taken at 10 min electrolysis time is achieving the highest COD reduction of 45.1% while the sample that was taken at 5 min electrolysis time is achieving the lowest COD reduction of 27.1 %. Therefore, at a certain current density, COD reduction percent increases as electrolysis time increases reaching a peak value at 10 min electrolysis time then it tends to decrease with electrolysis time due to the cathode passivation phenomena which affects the transfer of electrons, eventually the treatment efficiency. This result is compatible with (Eyvaz et al., 2009; Zaroual, Azzi, Saib, & Chainet, 2006).

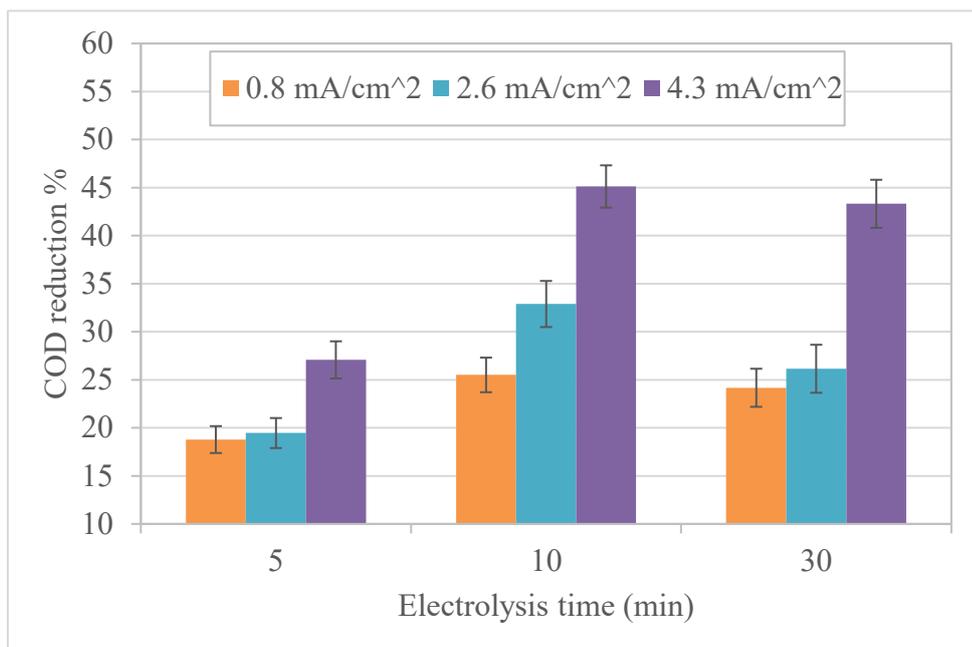


Figure 11. Impact of electrolysis time on COD reduction percent at three different current density for DC power source.

4.2. Performance of AC module in the post-treatment of TSE

The impact of current density and electrolysis time on TP removal percent and COD reduction percent were studied on this subsection using symmetrical cylindrical aluminum electrodes with AC power supply.

4.2.1. Impact of current density

Figure 12 shows the variation of TP removal percent versus current density at three electrolysis time. For 5 min electrolysis time, the enhancement in TP removal percent for 0.8 mA/cm² current density relative to 4.3 mA/cm² is by 17.6%, for 10 min is by 25.7% and for 30 min is by 24.4%. Therefore, at a certain electrolysis time, TP removal percent increases as current density increases. All previous studies for electrocoagulation using AC power supply show a direct relationship between the

removal percent of the pollutants and current density (Garcia-segura, Maesia, Eiband, Melo, & Martínez-Huitle, 2017; D. Duc Nguyen et al., 2016; Dinh Duc Nguyen, Kim, & Yoon, 2014).

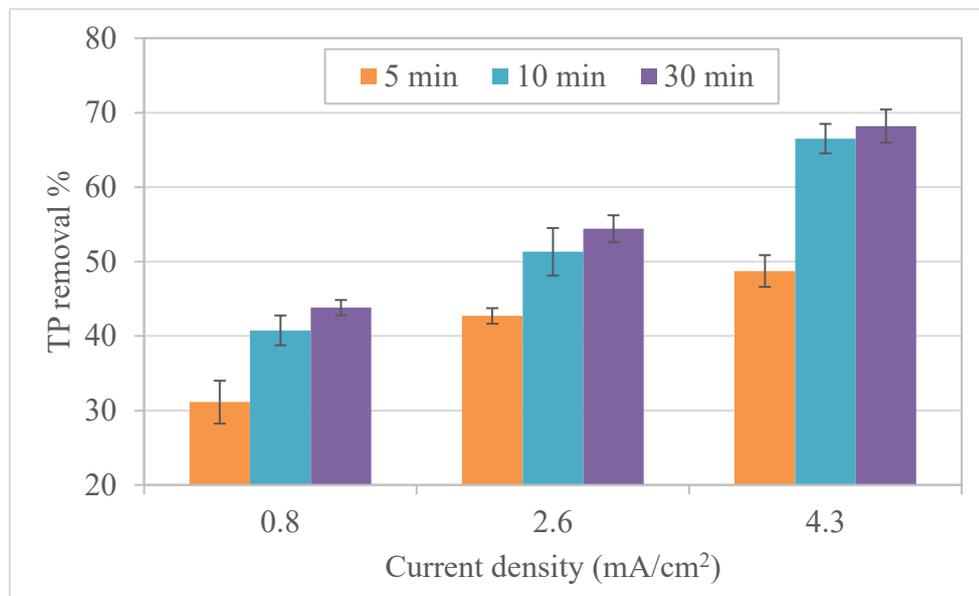


Figure 12. Impact of current density on TP removal percent at three different electrolysis time for AC power source.

In Figure 13, the impact of three different current density (0.8, 2.6 and 4.3 mA/cm²) was examined on COD reduction percent at three different electrolysis time (5, 10 and 30 min). Similar trends to TP removal percent were achieved in COD reduction percent in terms of the variation of current density. As can be clearly seen, at any specific electrolysis time, a current density of 4.3 mA/cm² has the highest COD reduction percent over the other current density (0.8 and 2.6 mA/cm²). Moreover, the enhancement in COD reduction using 4.3 mA/cm² current density relative to 0.8

mA/cm² current density, for 5 min is by 10.8%, for 10 min is by 16.7% and for 30 min is by 14.4%. In general, for AC power source COD reduction increases as current density increases at any certain electrolysis time (Dinh Duc Nguyen et al., 2014).

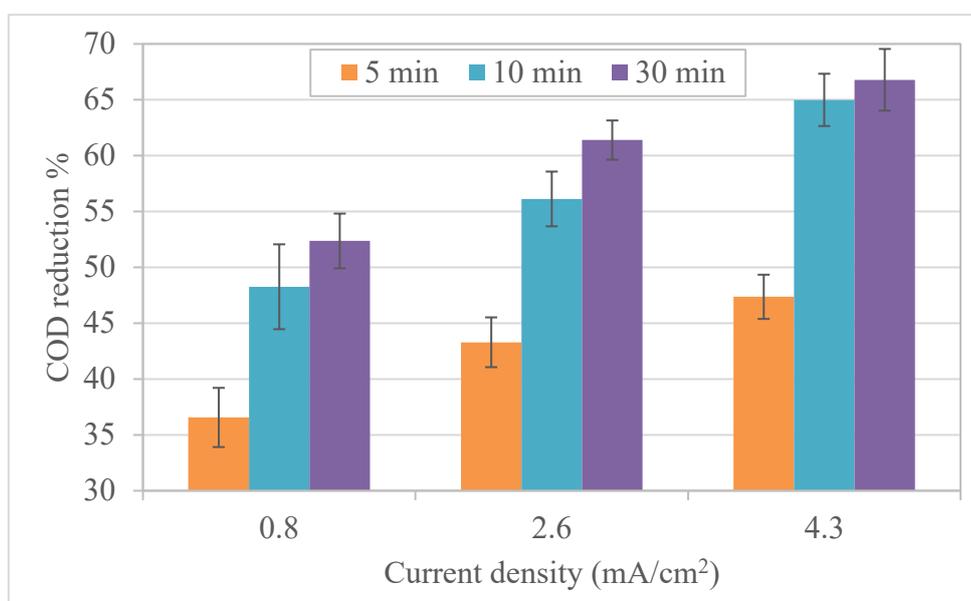


Figure 13. Impact of current density on COD reduction percent at three different electrolysis time for AC power source.

4.2.2. Impact of electrolysis time

In Figure 14, three different electrolysis time (5, 10 and 30 min) were used to examine the impact of electrolysis time on TP removal percent at three different applied current density (0.8, 2.6 and 4.3 mA/cm²). As can be seen, for 0.8 mA/cm² current density, the TP removal percent increases from 31.1% to 43.8% as the electrolysis time increases from 5 min to 30 min. Similarly, for 2.6 mA/cm² current density TP removal efficiency increases from 42.7% to 54.4% and for a current density of 4.3 mA/cm², TP

removal efficiency increases from 48.7% to 68.2%. Therefore, at any specific current density, as electrolysis time increases TP removal percent increases. Most of the previous study shows a direct relationship between electrolysis time and TP removal percent for AC power supply (D. Duc Nguyen et al., 2016; Dinh Duc Nguyen et al., 2014).

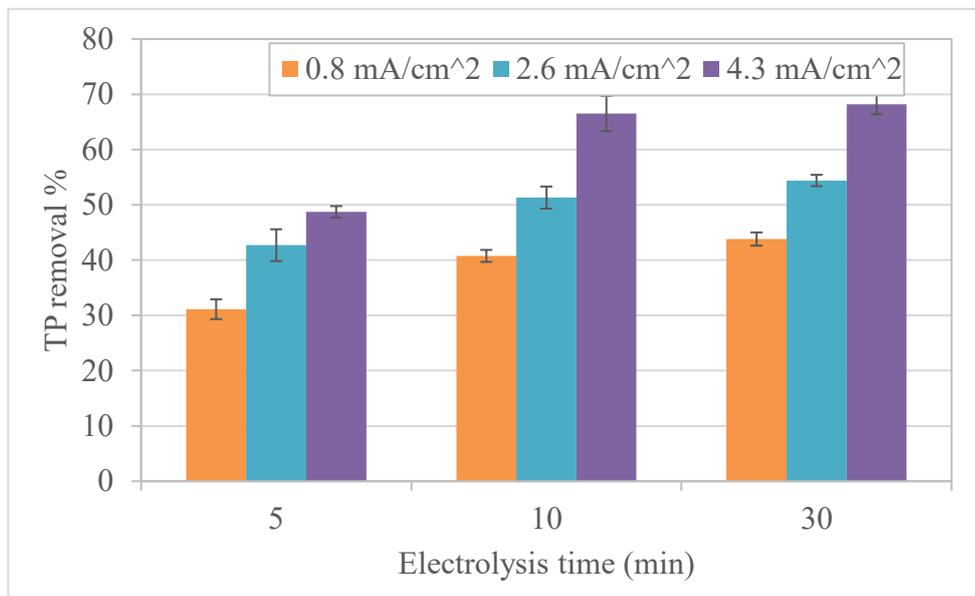


Figure 14. Impact of electrolysis time on TP removal percent at three different current density for AC power source.

Figure 15 illustrates the relationship between electrolysis time and COD reduction at three different current density (0.8, 2.6 and 4.3 mA/cm²). As can be seen, at minimum current density 0.8 mA/cm², COD reduction percent increases from 36.6% to 52.4% as electrolysis time increases from 5 min to 30 min. Similarly, at 2.6 mA/cm² current density, COD reduction percent increases from 43.3% to 61.4% as electrolysis

time increases from 5 min to 30 min. As well as, at maximum current density of 4.3 mA/cm², COD reduction percent increases from 47.4% to 66.8%. Therefore, at a certain current density, COD reduction increases with electrolysis time. Unlike DC power supply, where COD reduction decreases after a certain electrolysis time due to the formation of oxide layer on the surface of cathode which affects the transfer of electrons, eventually the treatment efficiency (Eyvaz et al., 2014, 2009; Xuhui et al., 2008). For AC power supply, most previous studies agreed on a direct relation between COD reduction and electrolysis time (Dinh Duc Nguyen et al., 2014).

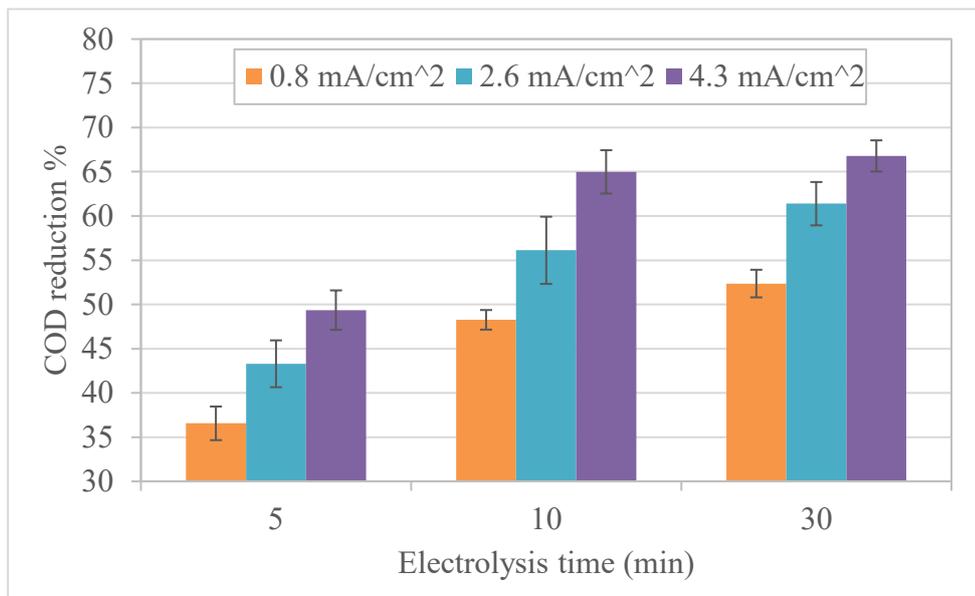


Figure 15. Impact of electrolysis time on COD reduction percent at three different current density for AC power source.

4.3. Performance of AC-DEP module in the post-treatment of TSE

At this subsection, the impact of current density and electrolysis time were studied on TP removal efficiency and COD reduction percent using unsymmetrical aluminum electrodes with AC power source with the application of a new force called Dielectrophoresis (DEP) force.

4.3.1. *Impact of current density*

Figure 16 shows the impact of three different current density (0.8, 2.6 and 4.3 mA/cm²) on TP removal percent at three different electrolysis time (5, 10 and 30 min). Similar trends to AC and DC module were observed in AC-DEP module, where TP removal percent increases as current density increases at any certain electrolysis time. The improvement in TP removal using AC-DEP configuration for 5 min electrolysis time by increasing current density from 0.8 mA/cm² to 4.3 mA/cm² is by 24.6%, for 10 min electrolysis time is by 35% and for 30 min electrolysis time is by 42.8%. Therefore, as the applied current density increases, the magnitude of the DEP force increases (Hawari et al., 2015; Larbi, Du, Baune, Thöming, & Hawari, 2018), hence enhancing the removal efficiency of the pollutants.

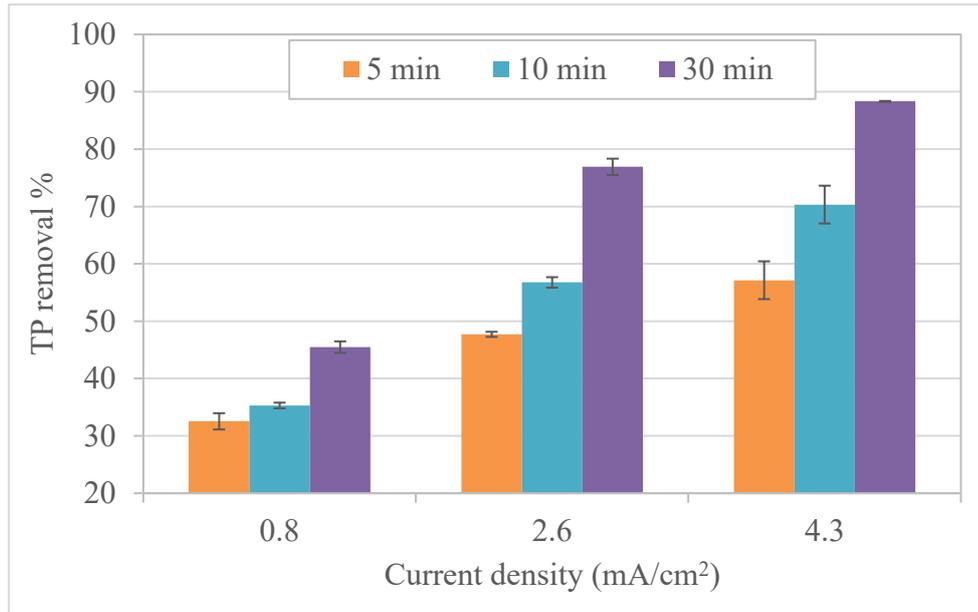


Figure 16. Impact of current density on TP removal percent at three different electrolysis time for AC-DEP power source.

In Figure 17, three different current density were applied in order to study the impact of the variation of current density on the COD reduction percent at three different electrolysis time. Similar to TP removals where at any specific electrolysis time, COD reduction increases as current density increases. Therefore, at 5 min electrolysis time, COD reduction percent increases from 38.5% to 55.4% as current density increases from 0.8 mA/cm² to 4.3 mA/cm², at 10 min electrolysis time COD reduction increases from 42.2% to 68.8% as current density increases from 0.8 mA/cm² to 4.3 mA/cm² and for 30 min electrolysis time COD reduction increases from 55.0% to 82.4% as current density increases from 0.8 mA/cm² to 4.3 mA/cm². Therefore, the improvement in COD reduction percent by increasing current density from 0.8 to 4.3 mA/cm² at 5 min electrolysis time is by 16.9%, for 10 min electrolysis time is by 26.6%

and for 30 min electrolysis time is by 27.4%. At any specific electrolysis time, COD reduction percent increases with current density as the magnitude of the DEP force increases (Hawari et al., 2015).

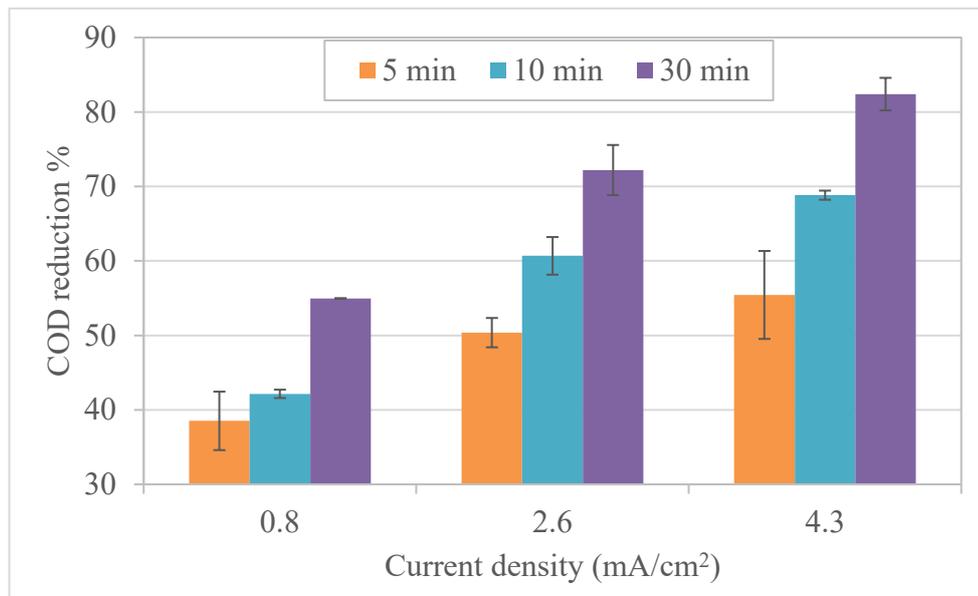


Figure 17. Impact of current density on COD reduction percent at three different electrolysis time for AC-DEP power source.

4.3.2. Impact of electrolysis time

Figure 18 shows the effect of electrolysis time (5, 10 and 30 min) on TP removal efficiency at three different current density (0.8, 2.6 and 4.3 mA/cm²). At a current density of 0.8 mA/cm² TP removal percent increases up to 45.5% as 30 minutes have passed. Similar trends were obtained for both current densities of 2.6 mA/cm² and 4.3 mA/cm². At 2.6 mA/cm² current density TP removal percent increases from 47.7% to 76.9% as electrolysis time increases from 5 min to 30 min at which the improvement

percent is by 29.2%. Also, at a current density of 4.3 mA/cm^2 TP removal percent increases from 57.1% to 88.3% as treatment time increases from 5 min to 30 min at which the improvement percent is by 31.2%. This improvement in the removals is due to the generation of a permanent force which is called the DEP force that continuously pushes the pollutants away from the inner electrode resulting in larger flocs that can be settle down by gravity (Hawari et al., 2015).

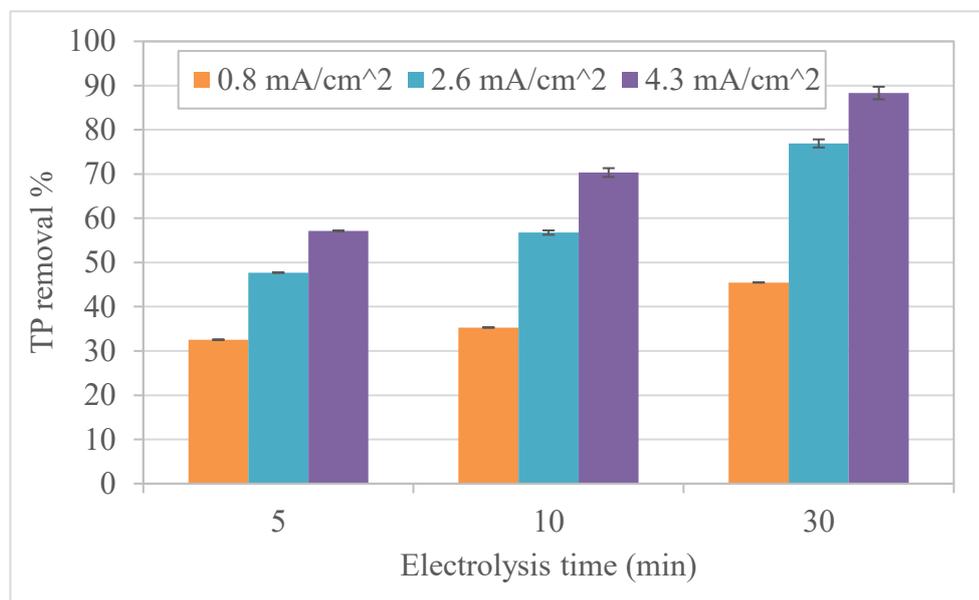


Figure 18. Impact of electrolysis time on TP removal percent at three different current density for AC-DEP power source.

Figure 19 illustrates the relationship between COD reduction and electrolysis time (5, 10 and 30 min) at three different current density (0.8 , 2.6 and 4.3 mA/cm^2). As can be clearly seen that at a current density of 0.8 mA/cm^2 COD reduction increases from 38.5% to 55.0% as electrolysis time increases from 5 min to 30 min. Similarly,

for a current density of 2.6 mA/cm² COD reduction increases from 50.4% to 72.2% as electrolysis time increases from 5 min to 30 min and for a current density of 4.3 mA/cm² COD reduction increases from 55.4% to 82.4% as electrolysis time increases from 5 min to 30 min. Similar to AC electrode configuration COD reduction percent does not decrease after it peaked at a specific electrolysis time; as mentioned before the advantages of AC power supply is that it deter the cathode passivation phenomena, eventually it ensure longer electrode life (Eyvaz et al., 2014, 2009; Garcia-segura et al., 2017; Holt, Barton, & Mitchell, 2006).

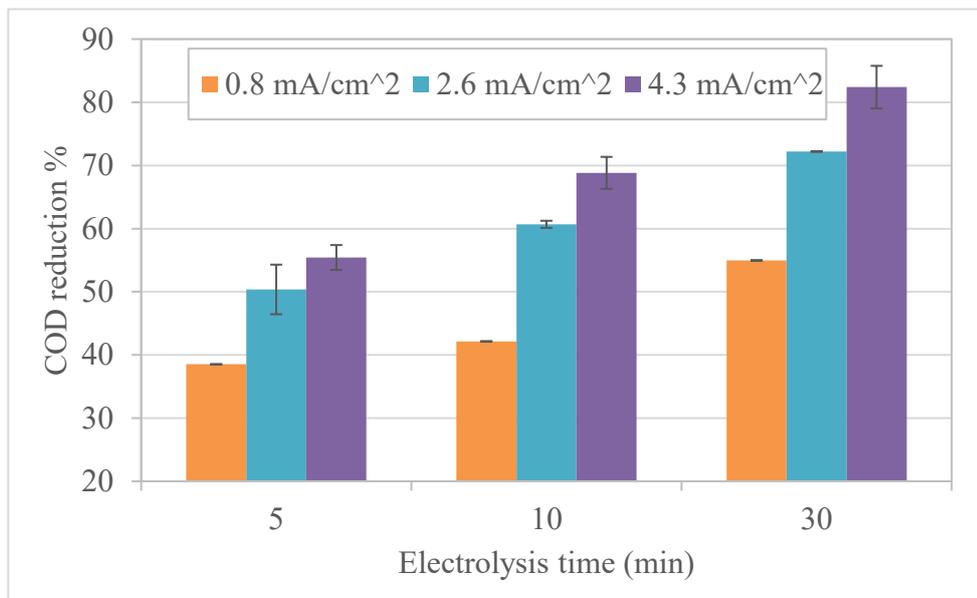


Figure 19. Impact of electrolysis time on COD reduction percent at three different current density for AC-DEP power source.

4.4. Comparison

4.4.1. *Impact of power supply system*

4.4.1.1. *Impact of current density*

Current density and electrolysis time play a key role in the performance of electrocoagulation process. Current density determines the required rate of coagulant dosage and bubble production (Didar & Islam, 2017; Omwene & Kobya, 2018; Sharma, Can, Hammed, Nawarathna, & Simsek, 2018). Figure 20 illustrates the impact of current density on (a) TP removal and (b) COD reduction for alternating and direct current power supply at 30 minutes electrolysis time. Generally, TP removal efficiency and COD reduction percent were increasing as the current density increases for both AC and DC power supply. At 30 min electrolysis time, the maximum TP removal and COD reduction for AC and DC were obtained at 4.3 mA/cm² current density. The reason behind that is as the current density increases, more alternating current passes through the inner electrode leads to more dissociation of inner electrode into metal ions (Al³⁺) according to Faraday's law. More dissociation of inner electrode surface boosts the formation of various large metal hydroxides flocs at which the total phosphorous, organic matter and inorganic matter can adsorb that can settle down by gravity leading to increase in TP removal efficiency and COD reduction (Bayramoglu, Kobya, Can, & Sozbir, 2004; Bazrefshan, Moein, Mostafapour, & Nakhaie, 2012; Đuričić et al., 2016; M. Kobya & Delipinar, 2008; Shalaby et al., 2014). Therefore, these results agreed with the most of previous studies as those studies showed a direct relationship of current density with TP removal efficiency and COD reduction (Bayramoglu et al., 2004; Bazrefshan et al., 2012; S. Chen et al., 2014; Farhadi et al., 2012; M. Kobya & Delipinar, 2008; Omwene & Kobya, 2018; Sahu, Mazumdar, & Chaudhari, 2014; Shalaby et al., 2014).

The enhancement in TP removal efficiency using AC relative to DC for 4.3 mA/cm² current density is by 22.2 %. While the enhancement in COD reduction percent is by 23.5 %. Regarding the high removals that obtained by AC than DC is that in AC power source, the function of the anode and cathode is shifted occasionally which will decrease the load in the function of each electrode that will help in the enhancement of the metal hydroxides formation at which contaminants adsorbed (Eyvaz et al., 2009).

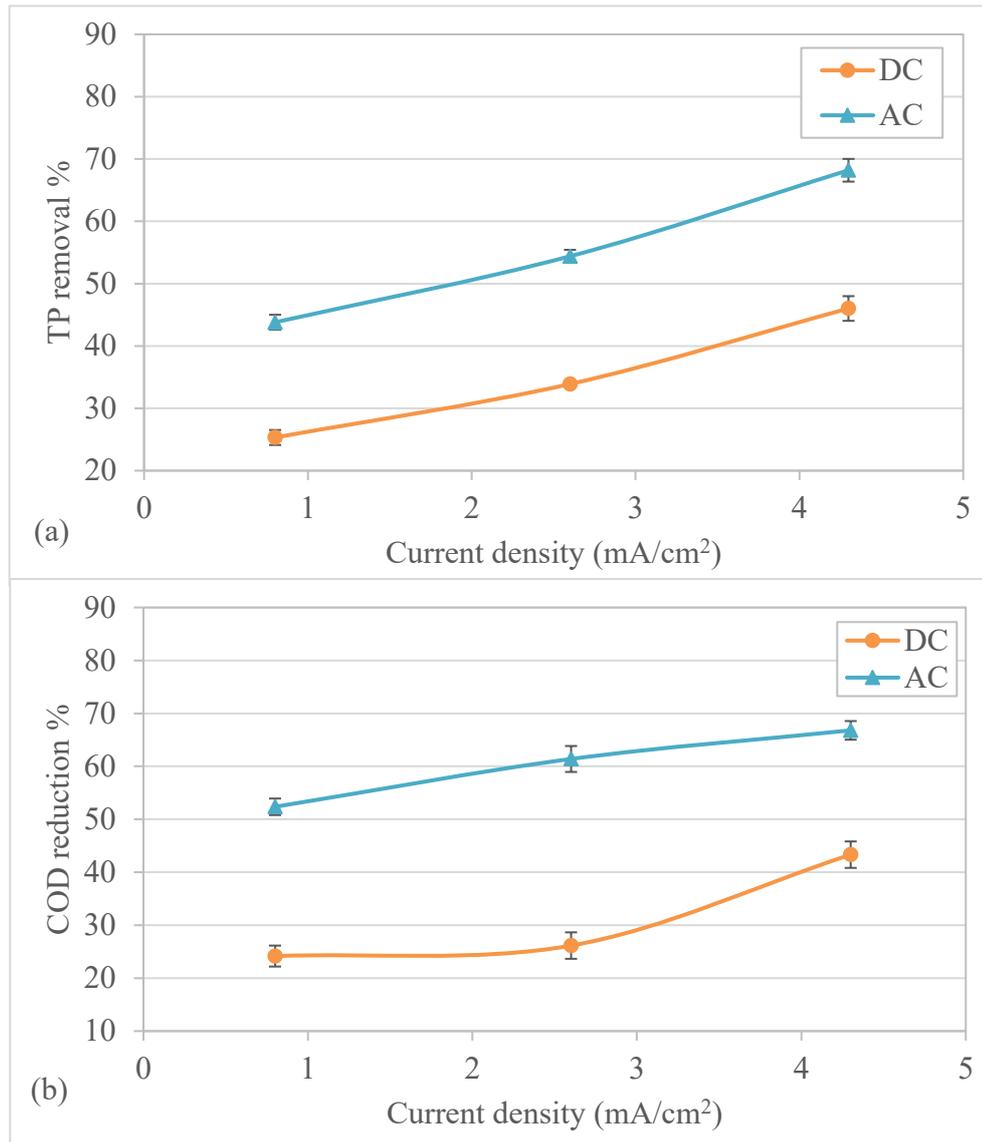


Figure 20. Effect of current density on (a) TP removal percent and (b) COD reduction percent at 30 min electrolysis time.

4.4.1.2. Impact of electrolysis time

Figure 21 shows the impact of electrolysis time on (a) TP removal efficiency and (b) COD reduction in terms of AC and DC power supply at 4.3 mA/cm² current density over 30 minutes electrolysis time. As can be seen, there are a major difference

in the initial removals of TP and COD at 5 min electrolysis time between AC and DC where AC has higher initial removals than DC. This short period of time is insufficient for the formation of metal hydroxides, in which TP removal efficiency and COD reduction for DC at 5 min are only 32.9 and 27.1 %, respectively while for AC 48.7 and 47.4 % removals were obtained, respectively. The high removals obtained by AC power supply system is due to the frequent change of polarity between inner and outer electrodes which enhance the formation of metal hydroxides at which the pollutants can adsorbed. Up to 10 min, both TP removal and COD reduction increase with time. The maximum TP removal of 68.2 and 46.0 % were obtained at 30 min electrolysis time for AC and DC, respectively and the maximum COD reduction of 66.8 and 45.1 % were obtained for AC at 30 min and DC at 10 min, respectively. Since, COD reduction with DC module after it reaches the peak at 10 min, it tends to decrease. The reason may be due to pH of wastewater since at different pH different products form at which organic wastes adsorbed and it may generate soluble products that will remain in the solution and cause COD value to increase, this result is compatible with (Eyvaz et al., 2009; Moreno-Casillas et al., 2007). Moreover, it could be due to after certain electrolysis time, cathode passivation occurred that is an impermeable oxide layer formed on the cathode surface which decrease the transfer of ions from anode to cathode and eventually it affects the dissociation of anode leading to decrease in the formation of metal hydroxides (Comninellis & Chen, 2010; Eyvaz et al., 2014, 2009; Khandegar & Saroha, 2013). The obtained results showed that cathode passivation occurs only in case of DC power supply which is compatible with (Eyvaz et al., 2009; Xuhui et al., 2008).

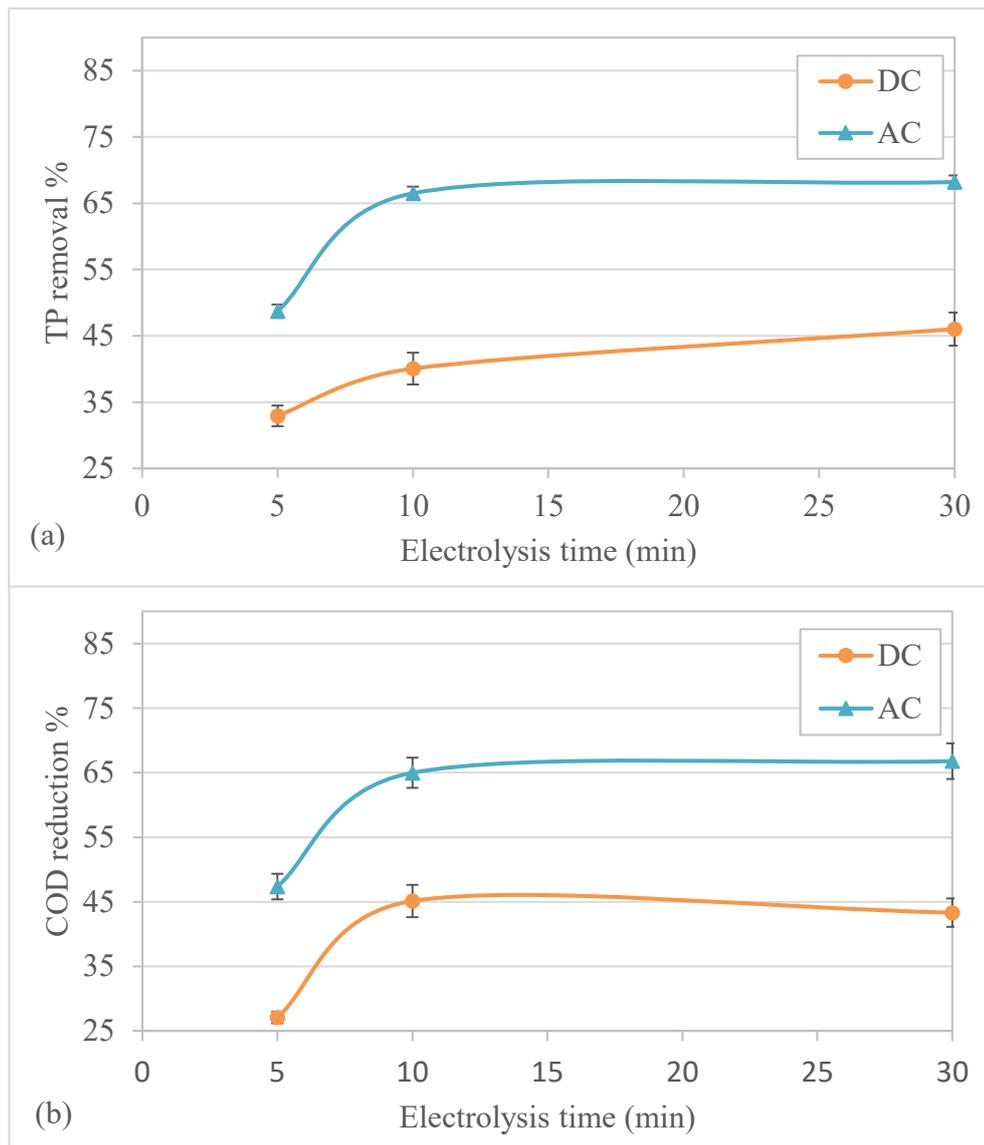


Figure 21. Effect of electrolysis time on (a) TP removal percent and (b) COD reduction percent at 4.3 mA/cm² current density.

4.4.2. *Impact of electrode configuration*

4.4.2.1. *Simulation results*

The DEP force has a direct relationship with electric field squared as shown in equation (9). Therefore, the simulation of the electric field squared was developed in order to examine the effect of current density on the DEP force field distribution as can be seen in Figure 22.

The simulated results showed that the strength of the generated DEP force was increasing towards the surface of the inner electrode having a maximum value at the surface of the inner electrode for all applied current density as can be shown in Figure 23.

Moreover, the results showed that the electrode configuration with 4.3 mA/cm² current density generated the highest maximum DEP force. The strength of electric field squared distribution followed the declining order of 4.3 mA/cm², 2.6 mA/cm² and 0.8 mA/cm² current density as can be seen in Figure 23. Therefore, as the applied current density increases, the strength of the DEP force increases (Hawari et al., 2015; Larbi et al., 2018). As can be seen by Figure 23, the largest effective working area of the DEP force is for electrode configuration with 4.3 mA/cm². Therefore, from the simulated results, it is expected that the electrode configuration with 4.3 mA/cm² current density has the best removal enhancement.

This simulation has proven that the DEP force works at low current density; however, it becomes more important and effective with increasing the applied current density, hence higher applied current density can generate more DEP forces which push more particles away from the inner electrode hence preventing the accumulation of the flocs on the electrode surface and forming larger agglomerates that settle down by gravity, consequently, enhances the removal efficiency.

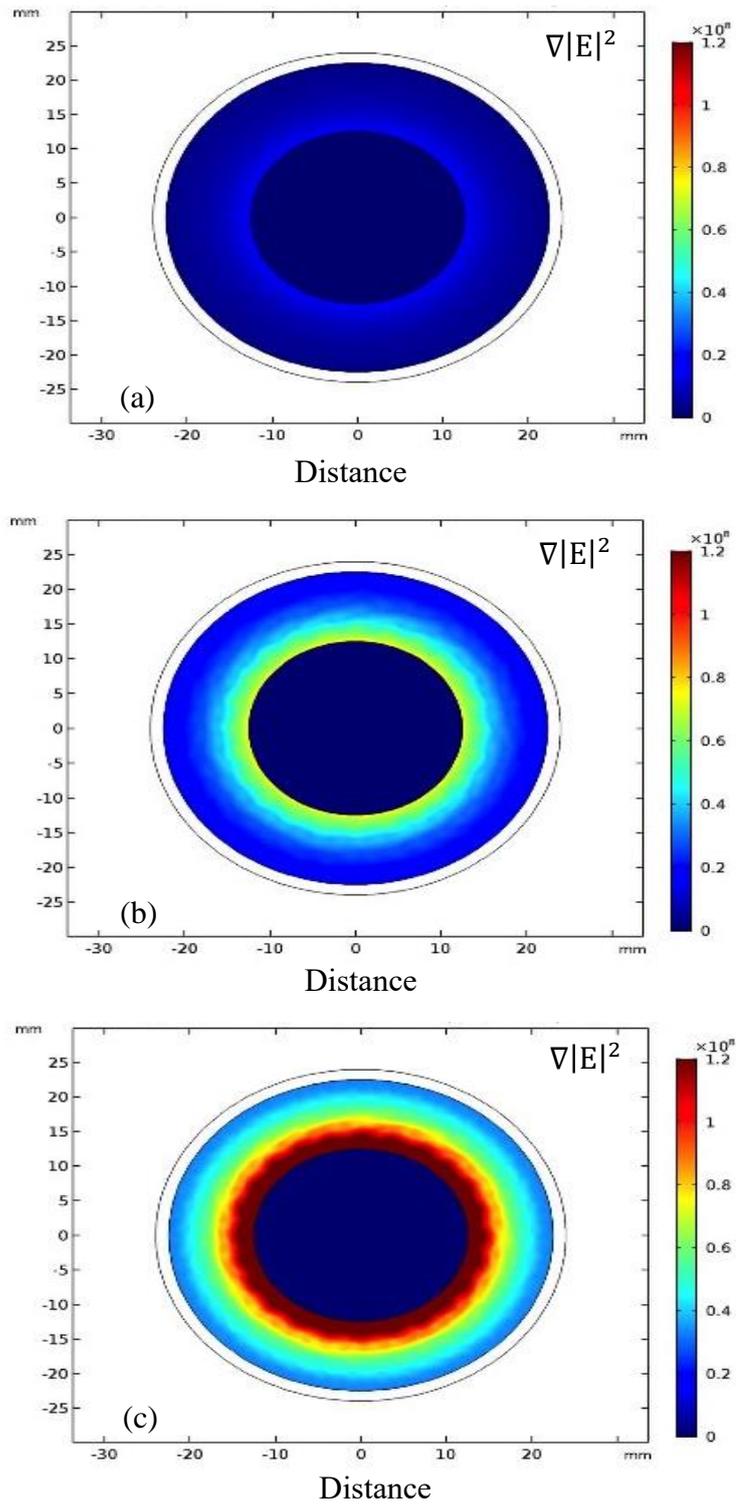


Figure 22. The DEP force field distribution defined as $(\nabla|E|^2)$ for three different applied current density of (a) 0.8 mA/cm^2 (b) 2.6 mA/cm^2 and (c) 4.3 mA/cm^2 .

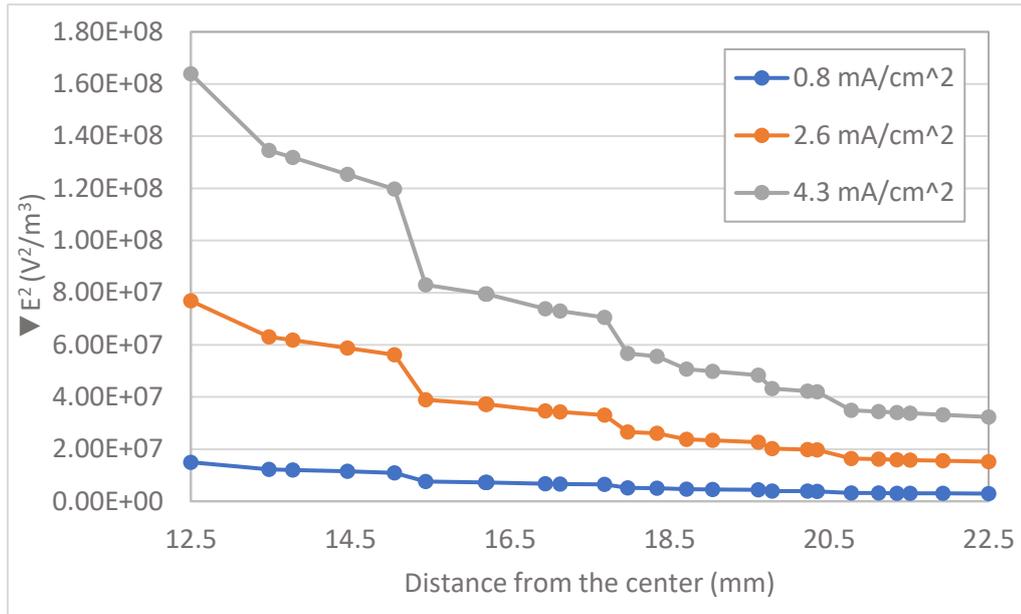


Figure 23. Electric field squared distribution between the inner and outer electrodes for different applied current density.

4.4.2.2. Experimental results

Figure 24 presents the impact of electrode configuration on (a) TP removal efficiency and (b) COD reduction percent over 30 min electrolysis time and for three different applied current density. As can be seen, at low current density (0.8 mA/cm²), the removals obtained by AC-DEP are similar to AC. Due to the much lower DEP force generated at low current density where this force is ineffective to help in a significant enhancement process (see Figure 22(a)). However, at higher current density (2.6 and 4.3 mA/cm²), the generated DEP force is large enough to push the particles away from the inner electrode surface enhancing the formation of larger agglomerates at which the contaminants adsorb and eventually settle down by gravity (see Figure 22(b) and (c)). In addition, this force will affect all suspended particles regardless of their electrical

charge by the induced dipole moment (F. Du et al., 2009; Hawari et al., 2015).

Moreover, for AC electrode configuration for a specific current density most of the TP removal and COD reduction occurs within the first 10 min electrolysis time (see Figure 24(a) and (b)), after that there will be no significant improvement in the treatment process with time. Because of initially the particles are colloid at high frequency, and after that there will be a reduction in the number of coagulants leading to reduce in the adsorption capacity and collision frequency (Fouad et al., 2009). However, for AC-DEP electrode configuration for a specific current density TP removal and COD reduction continuously increase with time. This is due to in conventional electrocoagulation process, agglomeration of suspended particles occurs, but the generated DEP force, which is a permanent force that is not affected by external factors, boost the coagulation process resulting in a continuous generation of the larger agglomeration of suspended particles at which more contaminants attached (Hawari et al., 2015). This agglomerates still exert nDEP effect, but with a higher magnitude, that continuously pushing it away from the inner electrode and eventually removed down by gravity as this generated DEP force decreases with distance in the direction of the outer electrode (see Figure 25).

The enhancement in TP removal efficiency using AC-DEP relative to AC at 30 min electrolysis time for 4.3, 2.6 and 0.8 mA/cm² current density are by 20.1, 22.5 and 1.7 %, respectively. While the enhancement in COD reduction percent are by 15.6, 10.8 and 2.6 %, respectively.

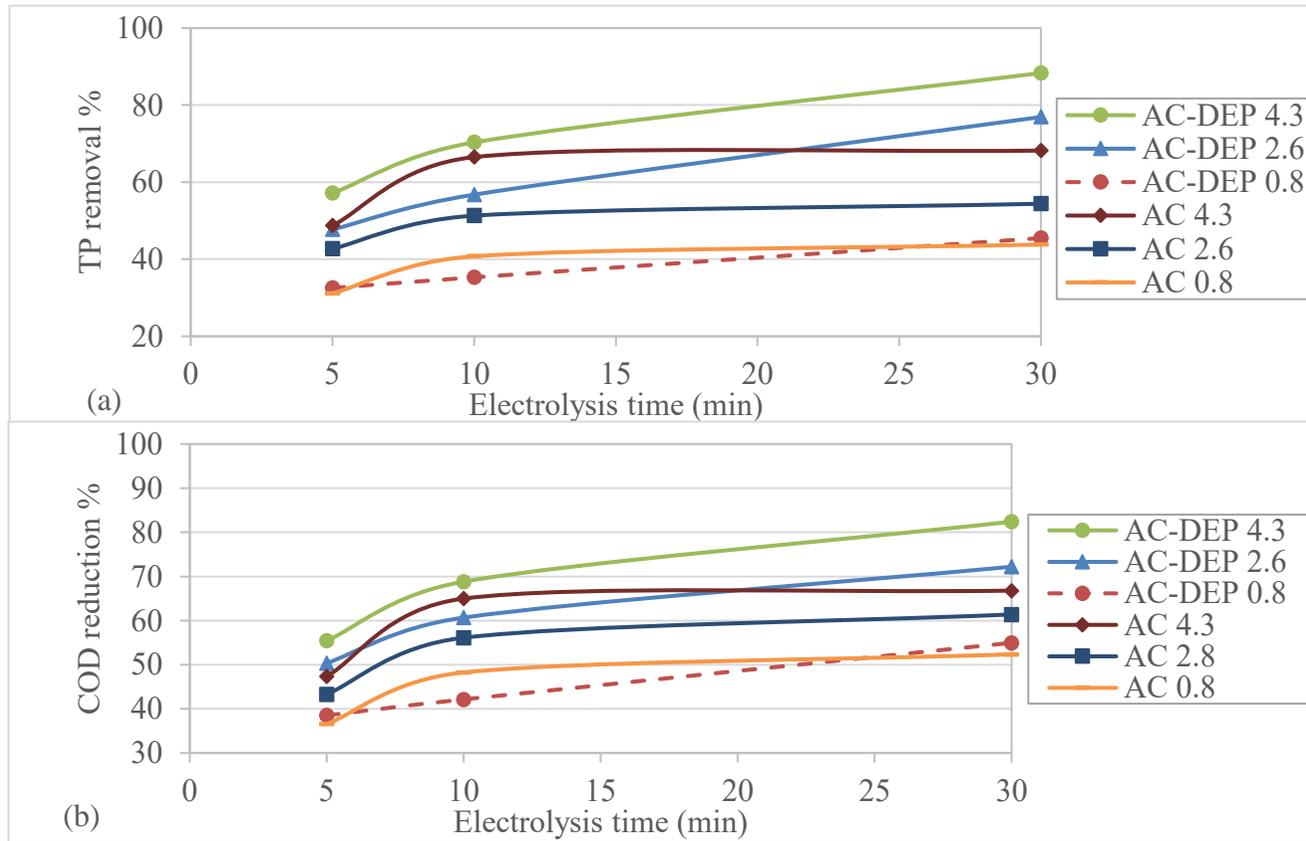


Figure 24. Impact of electrode configuration on (a) TP removal percent and (b) COD reduction percent.

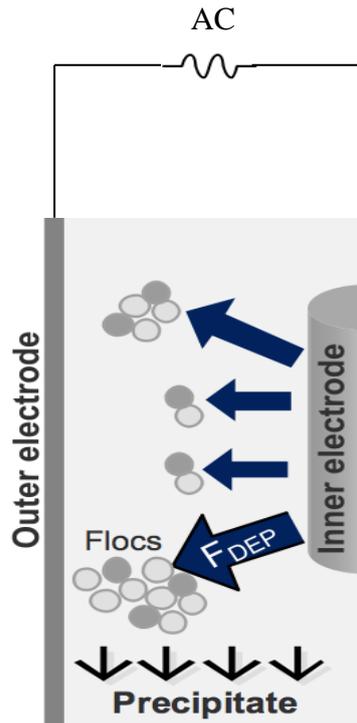


Figure 25. Schematic description of the enhancement of the formation of flocs using the DEP force.

4.5. Electrode corrosion

The dissociation of anode into metal ions causes the corrosion of the electrode material. Electrode corrosion gives an indication of the life of the electrode. Therefore, electrode corrosion play a key role in the viability of the electrocoagulation process (Ghosh et al., 2008). Electrode corrosion can be calculated by subtracting the mass of the electrode after a specific run from the mass of the electrode before the same run (Ghosh et al., 2008). Figure 26 presents the influence of electrode configuration and power supply system on electrode corrosion at 30 min electrolysis time and 4.3 mA/cm^2 current density. Electrode mass consumptions for AC-DEP, AC and DC modules are 0.02, 0.15 and 2,12 g, respectively. As can be seen, both electrode configuration that

working with AC power supply have very small total weight loss relative to DC electrode configuration this result agreed with all previous studies which agreed on the advantage of adopting AC power supply in electrocoagulation process to overcome the cathode passivation and anode oxidation phenomena that may occur in the case of DC power supply. Therefore, the application of AC power source ensures reasonable electrode life (Eyvaz et al., 2009; M Yousuf A Mollah, Schennach, Parga, & Cocke, 2001; S. Vasudevan & Lakshmi, 2012).

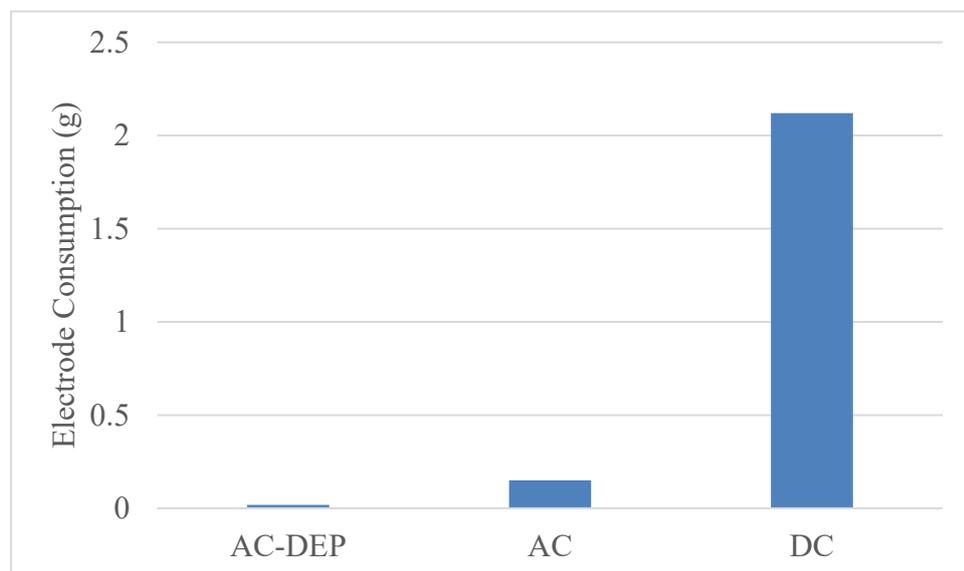


Figure 26. Effect of electrode configuration on electrode corrosion at 30 min electrolysis time and 4.3 mA/cm² current density.

CHAPTER 5: CONCLUSION AND RECOMMENDATIONS

Electrocoagulation process is an effective process for the treatment of a wide range of wastewater with high removal percent of pollutants. EC characterized by being simple, reliable, cost-effective and environmentally friendly process. However, most previous studies have been focused on the treatment of highly polluted wastewater by EC using DC power source, which encouraged this study to explore the effectiveness of EC in post-treating the treated secondary effluent using a new electrocoagulation electrode configuration with AC power supply. The main aim of this study is to compare the effectiveness in post-treatment of secondary treated effluent using three different EC modules, which are symmetrical aluminum electrode using DC power supply, symmetrical aluminum electrode using AC power supply and unsymmetrical aluminum electrodes using AC power source.

In DC module, COD reduction increases until a certain electrolysis time after that it decreases due to the accumulation of the pollutants on the electrode material unlike AC at which COD reduction increases over electrolysis time.

The simulation results showed that as applied current density increases, more DEP forces can be generated which pushes more particles away from the inner electrode forming more agglomerates at which contaminants adsorb. Moreover, it showed that the DEP force decreases with distance towards the outer electrodes hence the agglomerates settle down by gravity.

The experimental results showed that at low current density the removals for AC-DEP are similar to AC while at high current density, AC-DEP electrode configuration obtained higher TP removal and COD reduction than AC electrode configuration. This result agreed with the obtained simulation results. Moreover, it showed that the removals by AC-DEP continuously increase with time unlike AC.

Electrode corrosion was the highest for symmetrical aluminum electrode using DC power source over symmetrical aluminum electrode using AC power source and unsymmetrical aluminum electrode using AC power source.

This study shows its effectiveness in terms of cost by using an unsymmetrical electrodes configuration for the treatment process in which the inner electrode is smaller than the outer electrode producing a notable material reduction.

As EC with unsymmetrical electrode configuration shows its effectiveness in terms of being simple, reliable and cost-effective with high quality effluent, hence as a future work, the analysis should be upgraded to include the continuous mode operation for pilot scale EC system to test its effectiveness in replacing the wastewater tertiary treatment unit. As well as, it is recommended to apply high current density for AC-DEP module to get sufficient generated DEP forces that enhances the treatment process.

To conclude, this study shows that the effectiveness of electrocoagulation process can be enhanced by the application of the DEP force in the post-treatment of TSE.

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APPENDIX: ADDITIONAL RESULTS

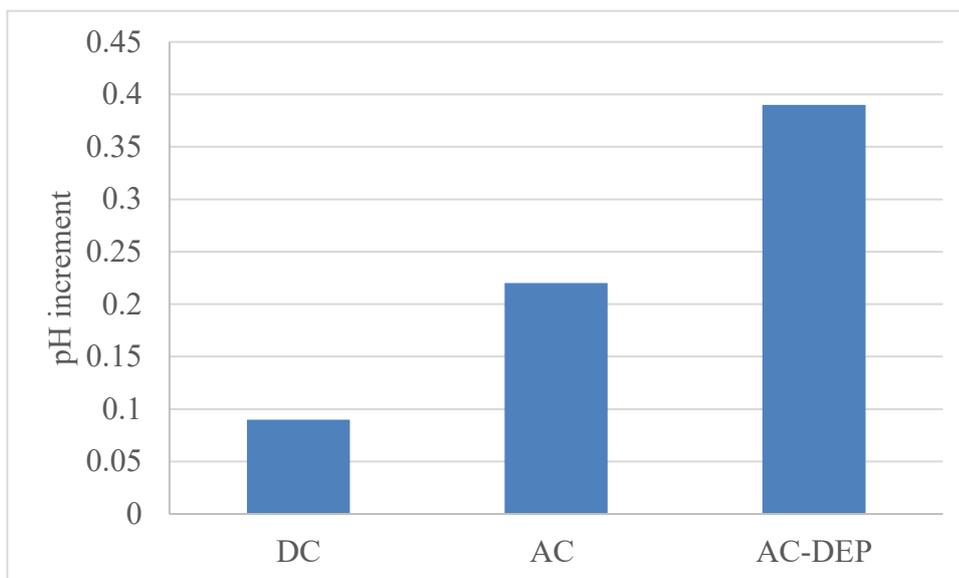


Figure 27. pH increment for DC, AC and AC-DEP electrode configuration at 4.3 mA/cm² and 30 min electrolysis time.