ELSEVIER

Contents lists available at ScienceDirect

Journal of Environmental Management

journal homepage: www.elsevier.com/locate/jenvman



Enhancing the electrocoagulation process for harvesting marine microalgae (*Tetraselmis* sp.) using interdigitated electrodes



Wardan A. Khatib ^a, Arslan Ayari ^a, Ahmed T. Yasir ^b, Mohammed Talhami ^a, Probir Das ^c, M. A. Quadir ^c, Alaa H. Hawari ^a, ^{*}

- ^a Department of Civil and Architectural Engineering, Qatar University, P.O. Box 2713, Doha, Qatar
- ^b Department of Chemical Engineering, Qatar University, P.O. Box 2713, Doha, Qatar
- c Algal Technologies Program, Center for Sustainable Development, College of Arts and Sciences, Qatar University, 2713, Doha, Qatar

ARTICLE INFO

Keywords: Marine microalgae Electrocoagulation Dielectrophoresis: harvesting efficiency Electric intensification

ABSTRACT

Marketable value of algal biomass has been increasing in recent years due to its wide range of applications. This study investigates the performance of a novel cylindrical interdigitated electrode array in electrocoagulation for the harvesting of marine microalgae (*Tetraselmis* sp.). The new electrode array is expected to exert a dielectrophoretic (DEP) force which would assist in the harvesting of the microalgae in the electrocoagulation process. Through numerical investigation, the induction of dielectrophoretic force was confirmed in the new electrode array. In this study, 10 min electrolysis time was found to be sufficient to harvest 82.4% microalgae with 1 cm electrode distance and 50 mA/cm² current density. Furthermore, decreasing the electrode distance to 0.5 cm increased the algal harvesting efficiency to 96.18%. Energy analysis showed that the proposed electrode array shows 38% lower specific energy consumption than the conventional flat sheet electrode array.

1. Introduction

The demand of algal biomass has been increasing in recent years due to its wide range of application. Algal biomass is used in different applications such as biochemistry, bioplastic, biofuel, pharmaceutical, nutrition and cosmetic industries (Gouveia, 2011; Matos et al., 2013). Moreover, due to their high nutrient content, algal biomass has been also found to be suitable as livestock feed (Hawari et al., 2020). Algal biomass is harvested from microalgae which are photosynthetic microorganisms that grow in different water bodies including freshwater, seawater and hot springs (Richmond, 2008). The harvesting of microalgae is challenging due to their small size (1–30 μ m diameter) (Jankowska et al., 2017). Microalgae are primarily harvested by centrifugation, sedimentation, coagulation-flocculation, membrane based filtration and electrocoagulation (Zhao et al., 2021). Electrocoagulation has showed an effective microalgal harvesting efficiency at a reduced energy demand (Gao et al., 2010b; Hawari et al., 2020).

Uduman et al. (2011) investigated the harvesting of two different strains of marine microalgae (*Chlorococcum* sp. and *Tetraselmis* sp.) using electrocoagulation (Uduman et al., 2011). Direct current (DC) was applied using a pair of flat sheet stainless steel electrodes. By applying a

voltage of 10 V for 900 s, 99% Tetraselmis sp. and 98% Chlorococcum sp. were harvested. The study also found that high temperature and high salinity of algal broth improves the harvesting efficiency. Vandamme et al. (2011) studied the effect of electrode type on the microalgal harvesting efficiency (Vandamme et al., 2011). In the study, iron (Fe) and aluminium (Al) electrodes were used to harvest Phaeodactylum tricornutum by applying DC. The study showed that after 60 min of electrocoagulation, the harvesting efficiency of the Al electrode array was 20% higher than the Fe electrode array. Zenouzi et al. (2013) found that using Al electrode array reduces the energy consumption by 23% when compared to Fe electrode array (Zenouzi et al., 2013). Vasudevan S. (2011) found that, by applying an alternating current (AC) instead of a direct current (DC) in electrocoagulation, the energy consumption of the electrocoagulation process can be reduced by 58% (Vasudevan et al., 2011). Hawari et al. (2020) studied algal harvesting using electrocoagulation by applying an alternating current in an asymmetrical cylindrical aluminium electrode array (Hawari et al., 2020). In the study, 90.9% algal harvesting efficiency was achieved within 10 min of electrocoagulation by applying 7.1 mA/cm² current. A summary of different studies that have utilized electrocoagulation for the harvesting microalgae can be found in Table 1 of the supplementary document. The

^{*} Corresponding author. Department of Civil and Architectural Engineering, College of Engineering, Qatar University, 2713, Doha, Qatar. *E-mail address:* a.hawari@qu.edu.qa (A.H. Hawari).

Table 1Initial characteristics of the algal broth.

Parameter	Value	Standard method
Temperature (°C)	$\textbf{23.1} \pm \textbf{0.1}$	APHA 2550 Temperature
pН	6.10 ± 0.1	APHA 4500-H b B. Electrometric Method
Conductivity (mS/	62.53 ± 1	APHA 2520 B. Electrical Conductivity
cm)		Method
Zeta potential (mV)	-29.3 ± 2	Particle Size and Zeta Potential Analyzer,
		Malvin

literature survey suggests that despite better harvesting efficiency, harvesting processes using alternating current has not been studied extensively.

In electrocoagulation, sacrificial electrodes are used to destabilize the suspension of algal particles in the aqueous medium. From the sacrificial electrodes, coagulants are formed in the aqueous medium which reduces the zeta potential and neutralizes the surface charge of the algae cells which in turn promotes coalescence (Uduman et al., 2011). Electrocoagulation comprises of three phases. Firstly, through electrolytic oxidation coagulants are released from the electrode. Secondly, floc formation occurs in situ through neutralization of the negative surface charge of microalgae. Finally, big flocs are lifted through the flotation of hydrogen microbubbles that are formed during the reduction reaction (Matos et al., 2013). During electrocoagulation using aluminium, the following electrochemical reaction takes place in the reactor:

Al
$$\to Al^{3+} + 3e^-$$
 (1)

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^-$$
 (2)

$$Al^{3+} + 3H_2O \leftrightarrow Al(OH)_3 + 3H^+$$
 (3)

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^-$$
 (4)

Complete reaction would be:

2Al
$$+6H_2O \leftrightarrow 2Al(OH)_3 + 3H_2$$
 (5)

It was found by (Alkhatib et al., 2020) that the performance of the electrocoagulation process can be further enhanced by inducing dielectrophoretic (DEP) force in the reactor. DEP force is a force that is generated on the dielectric particles in a non-uniform electric field (Hawari et al., 2015). During the application of the electrical current, dielectric polarization of particles takes place in the solution. As a result, a dipole moment is induced on the particles. Due to this induced dipole moment, a net force is generated on the particles which is known as dielectrophoretic force (Çetin and Li, 2011). Two types of DEP forces can affect the suspension of particles: positive DEP (pDEP) and negative DEP (nDEP). When the permittivity of the particles is higher than the aqueous medium, the particles will be attracted by the stronger electric field producing pDEP. Whereas, weaker electric field will attract the particles if their permittivity were lower than the aqueous medium, presenting nDEP (Du et al., 2009b; Çetin and Li, 2011). Type of DEP force exerted on a particle can be classified using the Clausius-Mossotti factor (\tilde{K}) that is calculated using equation (6):

$$K\% = \frac{\widetilde{\varepsilon}_p - \widetilde{\varepsilon}_M}{\widetilde{\varepsilon}_p + 2\widetilde{\varepsilon}_M} \tag{6}$$

$$\widetilde{\varepsilon} = \varepsilon - \frac{j\sigma}{\omega} \tag{7}$$

where, $\tilde{\epsilon}_M$ is the complex permittivity of the medium, $\tilde{\epsilon}_p$ is the complex permittivity of the particles, ϵ is the absolute permittivity, ω is the angular frequency $\left(\frac{rad}{s}\right)$, σ is the conductivity $\frac{S}{m}$, and j is the geometric gradient of the square of the electric field (E) that can be calculated

using equation (8):

$$j = \sqrt{-1} \cdot \left(E \cdot \nabla \right) E = \frac{1}{2} \nabla \left| E \right|^2 \tag{8}$$

The complex permittivity is used to replace the absolute permittivity by using alternative current (AC). Finally the DEP force can be calculated using (9) (Hawari et al., 2015):

$$F_{DEP} = 4\pi a^3 \varepsilon_0 \varepsilon_M re \left[\widetilde{\mathbf{K}} \right] (\mathbf{E} \cdot \nabla) \mathbf{E}$$
(9)

This study investigates the performance of a new cylindrical interdigitated electrodes (IDEs) array for electrocoagulation process. The new electrode configuration is designed for easier use in the electrocoagulation reactor. Through numerical analysis, the induction of DEP force in the electrocoagulation process will be demonstrated. Through induction of DEP force, this electrode array is expected to improve algal harvesting efficiency. The impact of current density, electrolysis time and inter-electrode distance on the algal harvesting efficiency of the proposed electrode array will be evaluated. A comparative study will also be performed using a pair of parallel flat plate electrodes with similar electrode area. Analysis of aluminium content in the harvested microalgae and energy consumption will also be investigated.

2. Material and methods

2.1. Microalgal species

In this study, marine microalgae (*Tetraselmis* sp.) was used. *Tetraselmis* sp. are elliptical, spherical, and unicellular microorganisms. Guillard's f/2 solution was used for the growth of algae where all the provided nutrients were of analytical grade. The initial optical density of the collected algae sample was measured at a wavelength of 750 nm using a spectrometer (Orion AquaMate UV-VIS Spectrophotometer Waltham, USA) where the algal broth was found to have an optical density of 0.300. Table 1 summarizes the initial characteristics of the algal broth.

2.2. Numerical analysis

In order to investigate the impact of dielectrophoretic force in the proposed electrocoagulation setup, a numerical model was built using COMSOL Multiphysics 5.5. As seen from equations (8) and (9), the DEP force is directly proportional to the square of the electric field. Thus, the square of the electric field was calculated as an indicator for the DEP force in the two proposed electrode arrays, the EC-DEP array and the parallel plate electrodes (EC). The schematics of the simulated geometry can be seen in Fig. 1. The effect of current density and electrode distance on the square of the electric field was assessed in both geometries. While studying the effect of current density, the electrode distance was kept constant at 1 cm. In this numerical study, the current density varied between 20, 30, 40 and 50 mA/cm². For comparison, the square of the electric field of the EC array was also evaluated at a current density of 50 mA/cm². For analysing the effect of electrode distance, the applied current density was kept constant at 50 mA/cm². For the EC-DEP array, the square of the electric field was evaluated at 0.5, 0.75, 1.00 and 1.25 cm electrode distance. For comparison, the square of the electric field of the EC array was also evaluated at an electrode distance of 0.5 cm. The numerical study was conducted in a two-dimensional model, assuming the length of the cylindrical rods (EC-DEP) and the width of the plates (EC) is infinite. The electric potential was solved at a set of boundary conditions. To solve this problem for the current densities, the quasielectrostatic form was used. The root mean square (rms) of the electric field is calculated using equation (10) (Du et al., 2009a):

$$\mathbf{E} = -\nabla \mathbf{\phi} \tag{10}$$

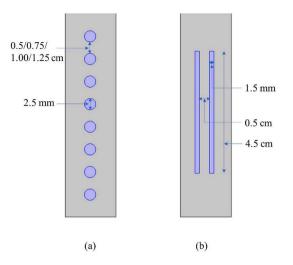


Fig. 1. Schematics of the geometry utilized for numerical study (a) EC-DEP electrode array, (b) EC electrode array.

Here, ϕ is the rms of the electrostatic potential which can be given by the Laplace's equation (11):

$$\nabla^2 \phi = 0 \tag{11}$$

The boundary conditions were fixed for the surface of the charge carrying electrodes:

$$\phi_1 = U_o \tag{12}$$

$$\phi_2 = 0 \tag{13}$$

Here, U_0 is the rms of the oscillating potential drop. To ensure meshindependent results, adaptive mesh refinement has been applied.

2.3. Experimental setup & procedure

The experimental setup of the electrocoagulation process is shown in Fig. 2. All the experiments were carried out in a graduated beaker with a volume of 1 L. An alternating current (AC) in the electrocoagulation process was provided using a variable transformer (KDGC-1KVA, China). To ensure the homogeneity of the broth in the reactor, continuous mixing was provided at 200 rpm using a magnetic stirrer (DLAB

MS-H280-Pro, China). The samples were collected from the reactor using a peristaltic pump (OMEGA FPU5-MT, Surrey, UK).

In this study, the performance of a cylindrical interdigitated electrode array (IDEs) was evaluated and compared with the performance of a pair of parallel plate electrodes. The IDEs array is composed of 8 interdigitated cylindrical electrodes. The length and diameter of each cylindrical rod were 65 mm and 2.5 mm, respectively, with a total surface area of 40.82 cm². In this paper, this electrode array will be referred to EC-DEP. The two parallel flat sheet aluminium electrodes have an area of 40.82 cm². This electrode array will be referred as EC in this paper. Using these two electrode arrays, the effect of electrolysis time, electrode distance and current density on the algal harvesting process was studied. During the electrocoagulation process, current and voltage were measured using two digital multimeters (Mastech MS8217, USA). After electrocoagulation, samples were collected from the reactor followed by 30 min of settling time. The optical density of the collected samples was then measured. The algal harvesting efficiency (η) was calculated using equation (14):

$$\eta = \left(\frac{OD_0 - OD_t}{OD_0}\right) 100\% \tag{14}$$

Where, OD_0 is the initial optical density of the algal broth and OD_t is the optical density after a prespecified time (t). After each experiment, the electrodes were cleaned before reused using sandpaper to remove any precipitates. Furthermore, the energy consumption C_{energy} (kWh/m³) was calculated using equation (15):

$$C_{energy} = \frac{U \times I \times t}{1000 \times v \times C_i}$$
 (15)

$$R = \frac{\rho L}{A} \tag{16}$$

where U is the voltage (V), I is the applied current (A), t is time (hr) at specific harvesting efficiency (%), C_i is the initial concentration of microalgae and v is the volume of the broth (m³). The resistance (R) of the electrodes was calculated using equation (16):

Here, ρ , L, and A corresponds to resistivity of the material, length and area of cross section of the electrode.

2.4. Aluminium content

In order to measure the aluminium content in the harvested algal biomass, the harvested algae samples were freeze dried for 1 day. 10 mg

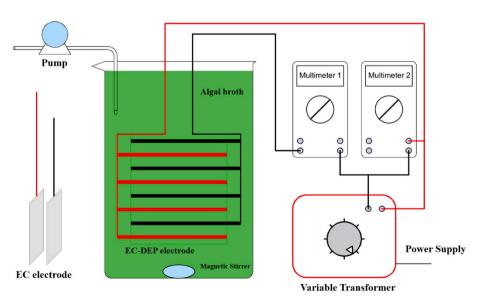


Fig. 2. Schematic diagram of the experimental setup

of the biomass was then digested with 2 mL of concentrated nitric acid in a hydrothermal autoclave reactor (Techinstro, India). The reactor was heated to 150 $^{\circ}\text{C}$ in a furnace for 5 h. Then the reactor was left to cool down to room temperature. The digested samples were filled with 10 mL of distilled water and filtered using a 0.2 μm pore size syringe filter (GD/X Whatman, UK). Finally, the samples were analysed by ICP-OES (Perkin OPTIMA 7300 DV, USA). An industrial machine vision camera (Daheng Imaging MER-112-32U3C, China) was used to study the surface of the electrode arrays before and after the electrocoagulation process. All the experiments were performed in triplicate and the average value was reported. The error bars for each presented value represent the variance of the different measured samples.

3. Results and discussion

3.1. Numerical analysis

In the numerical study, the effect of current density and electrode distance on the square of the electric field was evaluated. The effect of current density was studied at an electrode distance of 1 cm. For the ECDEP array, the studied current densities were 20, 30, 40 and 50 mA/cm². To compare the results with the EC array, the square of the electric field in the EC array was evaluated at 50 mA/cm² current density. Fig. 3 shows the effect of current density on the square of the electric field distribution for the studied electrode arrays. As seen from Fig. 3, for the EC-DEP electrode array, the maximum squared electric field intensity of $1\times 10^9~{\rm V}^2/{\rm m}^3$ was found at the surface of the electrodes and the intensity decreased with decreasing current density. Whereas for the EC electrode array, the squared electric field intensity was found only around the top and bottom edges of the plates.

The effect of electrode distance on the square of the electric field was evaluated with constant current density of 50 mA/cm². For the EC-DEP array, the studied distance between the electrodes was 0.5, 0.75, 1.00 and 1.25 cm. For comparison, the EC array was analysed with 0.5 cm electrode distance. Fig. 4 shows the square of the electric field distribution for the studied electrode arrays. As seen in Fig. 4, the maximum squared electric field intensity of $4\times10^9~V^2/m^3$ is observed at the surface of the electrodes in the EC-DEP array. However, as the electrode distance increases, the area of maximum squared electric field intensity around the surface of the electrode decreased (Fig. 4 (a), (b), (c) and (d)). Furthermore, the numerical study on the EC array illustrated that

the squared electric field intensity was found only around the top and bottom edges of the plates (Fig. 4 (e)). Hence, indicating minimal DEP force distribution is the EC array setup compared to the EC-DEP array.

3.2. Impact of electrolysis time

The impact of electrolysis time on the algal harvesting efficiency was studied using EC and EC-DEP arrays. The electrode distance and applied current density were kept constant at 1 cm and 50 mA/cm², respectively. Fig. 5 shows the effect of electrolysis time on the algal harvesting efficiency. As seen in Fig. 5, after 1 min, the algal harvesting efficiency for both electrode arrays was 6.89%. After 20 min of electrocoagulation, the harvesting efficiency reached 94.5% for both electrode arrays. For both electrode arrays, the harvesting efficiency increased with time. Application of current for longer duration will result in dissociation of further Al^{3+} ions from the sacrificial electrodes (as seen in equation (1)) (Hawari et al., 2020). Between a pH of 5 and 7, these Al³⁺ ions will react with OH⁻ ions in water and form Al(OH)₃ (Arain et al., 2015). Al(OH)₃ will neutralize the surface charge of the microalgae, which will cause the reduction in the electrostatic repulsion between the microalgae particles which will allow the Van der Wall's force to dominate. As a result, coalescence of the suspended microalgae will be promoted. Simultaneously, H2 gas will also be produced from the electrolytic reduction reaction at the anode (as seen in equation (4)). Formation of H2 and consumption of $\mathrm{OH^-}$ by $\mathrm{Al^{3+}}$ would reduce the pH of the algal broth. Reduction of the pH of the algal broth below 5 would prevent the formation of Al(OH)₃, as indicated by the Pourbaix diagram of Aluminium (Arain et al., 2015). During this study, a pH of 5.32 and 5.57 was recorded after 20 min of electrocoagulation using the EC-DEP and EC electrode array, respectively. Although electrocoagulation of both electrode arrays resulted in similar harvesting efficiency after 20 min of operation, EC-DEP electrode array reached 82.4% algal harvesting efficiency within 10 min. Whereas the EC electrode array reached 59.9% algal harvesting efficiency after 10 min. The EC-DEP array showed rapid coagulation rate due to the added dielectrophoretic effect which is induced due to the non-uniform electric fields created by the interdigitated cylindrical electrodes (Du et al., 2009a). The presence of an intense DEP force in the EC-DEP array was confirmed by the numerical study in section 3.1. The DEP force will promote collision among microalgae and assist the van der Waals's force in promoting coagulation (Hawari et al.,

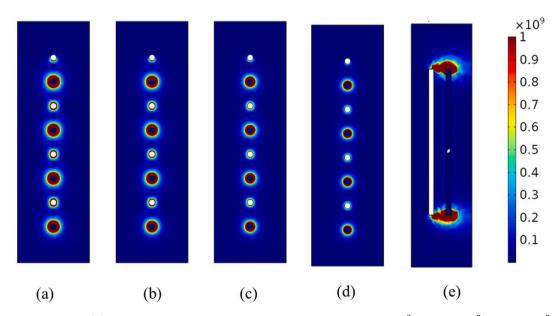


Fig. 3. Square of the electric field ($\nabla |E|^2$) distribution for EC-DEP module for current density of (a) 50 mA/cm², (b) 40 mA/cm², (c) 30 mA/cm² & (d) 20 mA/cm² and for EC module with current density of (e) 50 mA/cm².

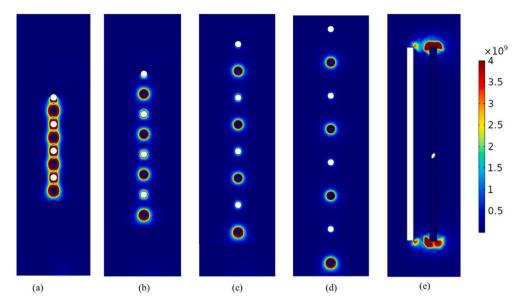


Fig. 4. Square of the electric field ($\nabla |E|^2$) distribution for EC-DEP module with electrode distance of (a) 0.5 cm, (b) 0.75 cm, (c) 1.00 cm & (d) 1.25 cm and for EC module with electrode distance of (e) 0.5 cm.

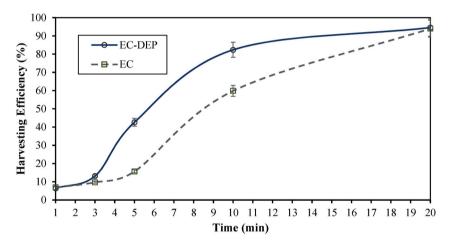


Fig. 5. Effect of electrolysis time on harvesting efficiency (1 cm electrode distance, 50 mA/cm²).

3.3. Impact of current density

The impact of current density on the harvesting efficiency of microalgae was studied for both EC and EC-DEP arrays. During electrocoagulation, the electrode distance was maintained at 1 cm and the electrocoagulation process was carried out for 10 min. The studied current densities were 20, 30, 40 and 50 mA/cm². Fig. 6 shows the effect of current density on the algal harvesting efficiency. As seen in Fig. 6, using the EC array, 24.9%, 39.1%, 47.2% and 59.9% algal harvesting efficiency was obtained after applying 20, 30, 40 and 50 mA/cm² current density, respectively. Whereas, using the EC-DEP module, 54.2%, 76.3%, 85.9% and 88.3% algal harvesting efficiency was obtained after applying 20, 30, 40 and 50 mA/cm² current density, respectively. For both EC-DEP and EC electrode arrays, increasing the current density increases the algal harvesting efficiency. Increasing the current density increases the production rate of Al³⁺ in the reactor (Gao et al., 2010a). Production of more Al³⁺ at a higher current density will promote the formation of Al(OH)3 which would enhance the electrocoagulation process. It can be also seen from Fig. 6 that the enhancement of the harvesting efficiency of the EC-DEP electrode array compared to the EC electrode array was 29.4%, 37.2%, 38.7% and 28.4% at 20, 30, 40 and 50 mA/cm² applied current density, respectively. This is because, along

with the van der Waals's force, the proposed EC-DEP electrode array induces additional dielectrophoretic force in the electrocoagulation process that improves collision between the microalgae and enhances coagulation. In this study, the DEP force exerted on the microalgae during electrocoagulation is negative DEP (nDEP) because the permittivity of the microalgae is lower than the permittivity of the algal broth (Hawari et al., 2015). The direction of the nDEP force is towards the region of low electric field from the region of high electric field. Thus, the nDEP will push the microalgae particles away from the surface of the electrodes. This will not only enhance the electrocoagulation process, but also reduce accumulation of microalgae on the electrodes (Hawari et al., 2020). The presence of DEP in the electrocoagulation process was confirmed in the numerical study shown in Fig. 3, which also suggested that increasing the current density will increase the dielectrophoretic force. In addition to the DEP force effect in the EC-DEP electrode array, it was found that the high electric field intensity in the EC-DEP array was recurring between the electrodes. While in the EC electrode array it was found that only the top and bottom edges of the plate electrodes showed high electric field intensity while most of the area of the electrode lacks high electric field intensity as shown in Fig. 7. Fig. 7 shows the electric field intensity at the surface of the EC and the EC-DEP electrode arrays. As seen from Fig. 7 (a), for the EC-DEP array, the highest electric field

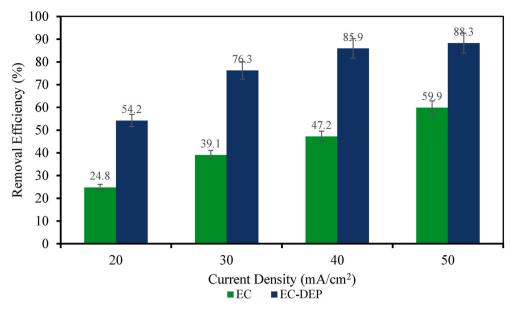


Fig. 6. Effect of applied current on harvesting efficiency (1 cm electrode distance, 10 min electrolysis time).

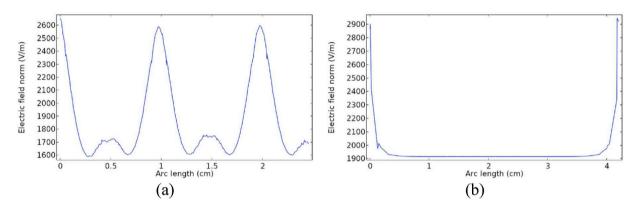


Fig. 7. Electric field intensity at the surface of (a) EC-DEP electrode array and (b) EC electrode array. (electrode distance of 0.50 cm and current density of 50 mA/cm², arc length = distance from the top edge (for EC array) and distance from the top electrode (EC-DEP array).

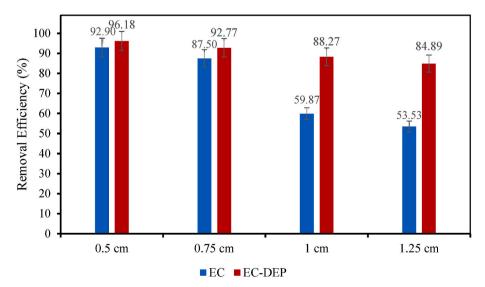


Fig. 8. Effect of electrode distance on algal harvesting efficiency (10 min, 50 mA/cm²).

intensity was 2900 V/m observed at the surface of the electrodes connected with the power source. On the other hand, at the surface of the grounded electrodes, an electric field intensity of 1700 V/m was observed. Whereas Fig. 7 (b) shows that for the EC electrode array, the electric field intensity at the top and bottom edges of the electrode was 2900 V/m. Away from the edges, the electric field intensity remains constant at 1900 V/m. The recurrence of high electric field intensity in the proposed EC-DEP electrode array would result in more production of aluminium in the electrocoagulation process which will enhance the harvesting efficiency. The more production of aluminium using the EC-DEP array compared to the EC array was confirmed in the amount of aluminium in the harvested microalgae. The amount of aluminium in the harvested algae is explained further in section 3.5.

3.4. Impact of electrode distance

The impact of electrode distance on the algal harvesting efficiency was studied for both EC and EC-DEP arrays. The studied electrode distances were 0.5, 0.75, 1.00 and 1.25 cm. The applied current density and electrolysis time of the electrocoagulation process were kept constant at 50 mA/cm² and 10 min, respectively. Fig. 8 presents the impact of electrode distance on the algal harvesting efficiency. From Fig. 8 it can be seen that for EC electrode array, 0.5, 0.75, 1 and 1.25 cm electrode distance resulted in harvesting efficiency of 92.9%, 87.5%, 59.9% and 53.5%, respectively. Whereas, for the EC-DEP electrode array, electrode distance of 0.5, 0.75, 1 and 1.25 cm resulted in 96.2%, 92.7%, 88.3% and 84.9% algal harvesting efficiency, respectively. In both EC and EC-DEP arrays, decreasing the electrode distance increased the harvesting efficiency due to the reduced electrical resistance in the electrocoagulation reactor (Du et al., 2013; Gao et al., 2010a). Ghosh et al. (2008) also suggested to use lower electrode distance to improve effectiveness of electrocoagulation and to reduce the energy consumption (Ghosh et al., 2008). From Fig. 8 it can also be observed that, for 0.5, 0.75, 1 and 1.25 cm electrode distance, the harvesting efficiency of the EC-DEP array was 3.28%, 5.28%, 28.40% and 31.36% higher, respectively than the harvesting efficiency obtained using the EC array. This is because of the additional DEP force exerted in the EC-DEP array as indicated by the numerical study in section 3.1. Fig. 8 also indicates that increasing the distance between the electrodes from 0.50 cm to 0.75 cm, 1.00 cm and 1.25 cm decreases the harvesting efficiency by 3.41%, 7.91% and 11.29%, respectively. Whereas the harvesting efficiency difference increased significantly by 5.4%, 33.03% and 39.37% for electrode distance of 0.50, 0.75, 1.00 and 1.25 cm, respectively in the EC electrode array. The difference in the EC-DEP electrode array is not very significant because even at higher electrode distances, the EC-DEP

electrode array exhibits electric field intensity higher that 2000 V/m. This high electric field intensity is exhibited due to higher resistance in the EC-DEP electrode array which will result in additional aluminium production during electrocoagulation. The additional aluminium is found in the harvested algae and is discussed in detail in section 3.5.

3.5. Energy consumption

The specific energy consumption of the electrocoagulation process with EC and EC-DEP electrode array was studied for current densities of 20, 30, 40 and 50 mA/cm². For this study, the electrolysis time and electrode distance were kept constant at 10 min and 1 cm, respectively. The specific energy consumption was calculated using equation (15). Fig. 9 shows the effect of current density on the specific energy consumption of EC and EC-DEP electrode array. As seen from Fig. 9 using the EC electrode array with electrode distance of 1 cm, application of 20, 30, 40 and 50 mA/cm² current density resulted in specific energy consumption of 2.24, 3.05, 4.15 and 4.38 kWh/kg, respectively. Whereas, using the EC-DEP electrode array, applying a current density of 20, 30, 40 and 50 mA/cm² resulted in specific energy consumption of 1.41, 2.22, 3.01 and 3.84 kWh/kg, respectively. The results in Fig. 9 shows that the energy consumption of both electrode arrays increased with increasing current density. The trend of increasing energy consumption with the increase in current density is expected according to equation (15). Moreover, Fig. 9 shows that the EC-DEP electrode array results in lower energy consumption than the EC electrode array for all evaluated current densities. This due to the higher algal harvesting efficiency obtained by the EC-DEP electrode array. The proposed EC-DEP electrode array achieved lower energy consumption compared to Hawari et al. (2020) and Uduman et al. (2011) who harvested the same marine microalgae (Tetraselmies sp.) with an energy consumption of 4.62 and 9.16 kWh/kg, respectively (Hawari et al., 2020; Uduman et al., 2011).

3.6. Aluminium content

Aluminium content was analysed in the harvested algal biomass. The algae used for aluminium analysis was collected after electrocoagulation at an electrode distance of 0.5 cm after 10 min electrolysis time using EC and EC-DEP electrode arrays. Fig. 10 shows the aluminium content in the harvested microalgae at current densities of 20, 30, 40 and 50 mA/cm². As seen from Fig. 10, the EC electrode array resulted in 3.10, 11.39, 17.02 and 15.48 mg/g aluminium in the harvested microalgae for current densities of 20, 30, 40 and 50 mA/cm², respectively. Whereas the EC-DEP electrode array resulted in 6.79, 16.24, 19.07 and 19.78 mg/g aluminium in the harvested microalgae for current densities of 20, 30,

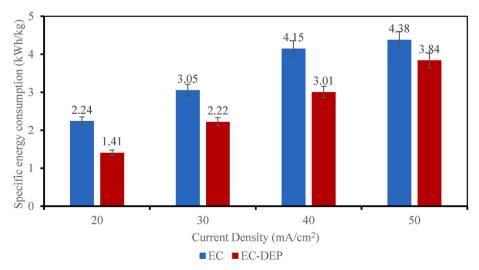


Fig. 9. Specific energy consumption of the electrocoagulation process for EC and EC-DEP array at different electrode distances.

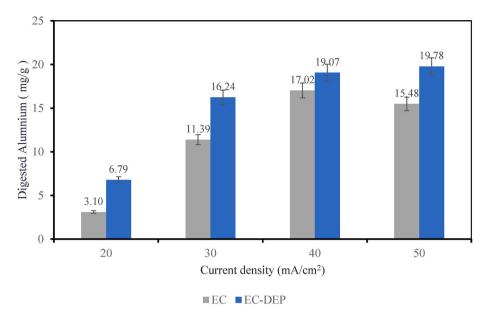


Fig. 10. Effect of current density on the aluminium content in harvested microalgae (0.5 cm).

40 and 50 mA/cm², respectively. As observed from these results, while using the EC-DEP array, increasing the current density increased the aluminium content in the harvested microalgae. This is because, at a higher current density more aluminium hydroxide Al(OH) $_3$ would be produced in the electrocoagulation reactor (Arain et al., 2015). This additional aluminium hydroxide resulted in higher algal harvesting efficiency and hence higher aluminium content in the harvested microalgae. This trend was not observed while using the EC electrode array, it was observed that as the current density increased from 40 mA/cm² to 50 mA/cm² the aluminium content in the harvested algae decreased from 17.02 to 15.48 mg/g. This could be due to electrode passivation at higher current densities which indicates low current efficiency.

Current efficiency is the ratio of the actual mass of a substance liberated from an electrolyte by the passage of current to the theoretical mass liberated according to Faraday's law (Ahmadi and Ghanbari, 2016; Izquierdo et al., 2010). The higher amount of Aluminium in the collected microalgae in the EC-DEP electrode array compared to the EC electrode array indicates that the current efficiency of the proposed EC-DEP electrode array is higher than the current efficiency of the conventional EC electrode array. Moreover, it was found that the aluminum hydroxide Al(OH)₃ formed during the electrocoagulation process deposited on the electrode surface which caused electrode passivation. Fig. 11 shows the electrocoagulation process. As seen in Fig. 11 (b) and (c)

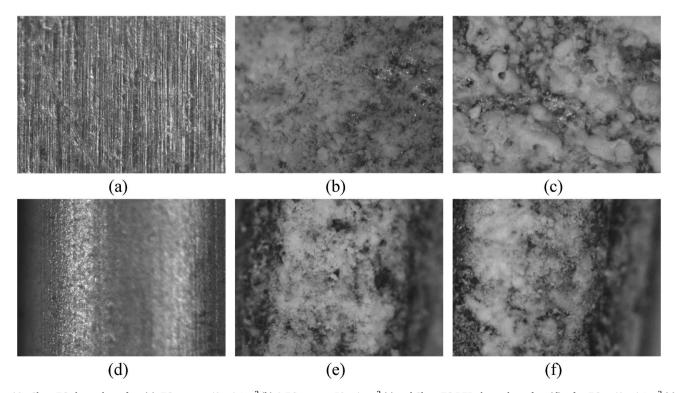


Fig. 11. Clean EC electrode surface (a), EC array at 40 mA/cm² (b) & EC array at 50 mA cm² (c) and Clean EC-DEP electrode surface (d), after EC at 40 mA/cm² (e) & after EC at 50 mA/cm² (f).

passivation of the EC electrode array intensifies when 50 mA/cm² current density was applied. On the other hand, Fig. 11 (e) and (f) shows that for the EC-DEP array the electrode passivation is relatively similar when 40 mA/cm² and 50 mA/cm² current densities are applied. For the EC array, the higher degree of passivation at 50 mA/cm² current density reduced coagulant production rate and resulted in 10% lower aluminum content in the harvested microalgae, compared to 40 mA/cm² current density. Thus, it can be concluded that the proposed electrode array can reduce the degree of electrode passivation and improve the utilization of produced coagulants through improved harvesting efficiency.

4. Conclusion

This study investigated the performance of a novel dielectrophoretic force induced by cylindrical interdigitated electrode array (EC-DEP) for harvesting marine microalgae (Tetraselmis sp.) in electrocoagulation. The performance of the proposed electrode array was compared with a conventional flat parallel plate electrode array (EC). Through numerical study, the induction of dielectrophoretic force was confirmed in the electrocoagulation process. During experimental analysis, applying 50 mA/cm² current for 10 min using the proposed EC-DEP electrode array resulted in 88.3% algal harvesting efficiency. Whereas similar operating condition resulted in 59.9% algal harvesting efficiency using the conventional EC electrode array. The improvement in algal harvesting efficiency using the EC-DEP module can be attributed to three main factors. Firstly, the recurrence of high electric field intensity in the proposed EC-DEP electrode array resulted in additional aluminium production in the electrocoagulation process. Secondly, the EC-DEP electrode array intensified the collision among microalgae and assisted the van der Waals's force in promoting coagulation. Thirdly, reduced electrode passivation in the proposed EC-DEP electrode array helped to sustain higher algal harvesting efficiency at higher current densities.

Credit author statement

Wardan A. Khatib: Validation, Formal analysis, Investigation, Writing – original draft. Arslan Ayari: Conceptualization, Methodology, Writing-Review. Ahmed T. Yasir: Numerical Analysis, Writing-Review. Mohamed Talhami: Analysis and Validation. Probir Das: Resources, Conceptualization. MA Quadir: Aluminium digestion and Resources. Alaa H. Hawari: Conceptualization, Project administration, Supervision,

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This project was made possible by UREP award (UREP25-060-2-025) from Qatar National Research Fund (QNRF). The statements made herein are solely the responsibility of the authors. The authors also wish to thank the Central Laboratories Unit (CLU) at Qatar University for carrying out the aluminium analysis. Open Acess funding provided by Qatar National Library.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.

org/10.1016/j.jenvman.2021.112761.

References

- Ahmadi, M., Ghanbari, F., 2016. Optimizing COD removal from greywater by photoelectro-persulfate process using Box-Behnken design: assessment of effluent quality and electrical energy consumption. Environ. Sci. Pollut. Res. Int. 23 (19), 19350–19361. https://doi.org/10.1007/s11356-016-7139-6.
- Alkhatib, A.M., Hawari, A.H., Hafiz, M.A., Benamor, A., 2020. A novel cylindrical electrode configuration for inducing dielectrophoretic forces during electrocoagulation. J. Water Process Eng. 35, 101195 https://doi.org/https://doi.org/10.1016/j.jwpe.2020.101195.
- Arain, M.S., Arain, S.A., Kazi, T.G., Afridi, H.I., Ali, J., Arain, S.S., Brahman, K.D., Mughal, M.A., 2015. Temperature controlled ionic liquid-based dispersive microextraction using two ligands, for determination of aluminium in scalp hair samples of Alzheimer's patients: a multivariate study. Spectroschim. Acta Mol. Biomol. Spectrosc. 137, 877–885.
- Çetin, B., Li, D., 2011. Dielectrophoresis in microfluidics technology. Electrophoresis 32 (18), 2410–2427.
- Du, F., Hawari, A., Baune, M., Thöming, J., 2009a. Dielectrophoretically intensified cross-flow membrane filtration. J. Membr. Sci. 336 (1), 71–78 https://doi.org/ https://doi.org/10.1016/j.memsci.2009.03.010.
- Du, F., Hawari, A., Baune, M., Thöming, J., 2009b. Dielectrophoretically intensified cross-flow membrane filtration. J. Membr. Sci. 336 (1–2), 71–78.
- Du, F., Ciaciuch, P., Bohlen, S., Wang, Y., Baune, M., Thöming, J., 2013. Intensification of cross-flow membrane filtration using dielectrophoresis with a novel electrode configuration. J. Membr. Sci. 448, 256–261.
- Gao, S., Yang, J., Tian, J., Ma, F., Tu, G., Du, M., 2010a. Electro-coagulation–flotation process for algae removal. J. Hazard Mater. 177 (1), 336–343 https://doi.org/ https://doi.org/10.1016/j.jhazmat.2009.12.037.
- Gao, S., Yang, J., Tian, J., Ma, F., Tu, G., Du, M., 2010b. Electro-coagulation-flotation process for algae removal. J. Hazard Mater. 177 (1-3), 336-343.
- Ghosh, D., Solanki, H., Purkait, M.K., 2008. Removal of Fe(II) from tap water by electrocoagulation technique. J. Hazard Mater. 155 (1), 135–143 https://doi.org/ https://doi.org/10.1016/j.jhazmat.2007.11.042.
- Gouveia, L., 2011. Microalgae as a feedstock for biofuels. In: Microalgae as a Feedstock for Biofuels. Springer, pp. 1–69.
- Hawari, A.H., Du, F., Baune, M., Thöming, J., 2015. A fouling suppression system in submerged membrane bioreactors using dielectrophoretic forces. J. Environ. Sci. 29, 139–145.
- Hawari, A.H., Alkhatib, A.M., Das, P., Thaher, M., Benamor, A., 2020. Effect of the induced dielectrophoretic force on harvesting of marine microalgae (Tetraselmis sp.) in electrocoagulation. J. Environ. Manag. 260, 110106 https://doi.org/https://doi. org/10.1016/j.jenvman.2020.110106.
- Izquierdo, C.J., Canizares, P., Rodrigo, M.A., Leclerc, J.P., Valentin, G., Lapicque, F., 2010. Effect of the nature of the supporting electrolyte on the treatment of soluble oils by electrocoagulation. Desalination 255 (1–3), 15–20.
- Jankowska, E., Sahu, A.K., Oleskowicz-Popiel, P., 2017. Biogas from microalgae: review on microalgae's cultivation, harvesting and pretreatment for anaerobic digestion. Renew. Sustain. Energy Rev. 75, 692–709 https://doi.org/https://doi.org/10.1016/ i.rser.2016.11.045.
- Matos, C.T., Santos, M., Nobre, B.P., Gouveia, L., 2013. Nannochloropsis sp. biomass recovery by Electro-Coagulation for biodiesel and pigment production. Bioresour. Technol. 134, 219–226.
- Richmond, A., 2008. Handbook of Microalgal Culture: Biotechnology and Applied Phycology. John Wiley & Sons.
- Uduman, N., Bourniquel, V., Danquah, M.K., Hoadley, A.F.A., 2011. A parametric study of electrocoagulation as a recovery process of marine microalgae for biodiesel production. Chem. Eng. J. 174 (1), 249–257 https://doi.org/https://doi.org/10.1016/j.cej.2011.09.012.
- Vandamme, D., Cláudia Vieira Pontes, S., Goiris, K., Foubert, I., Jozef Jan Pinoy, L., Muylaert, K., 2011. Evaluation of electro-coagulation-flocculation for harvesting marine and freshwater microalgae. Biotechnol. Bioeng. 108 https://doi.org/ 10.1002/bit.23199.
- Vasudevan, S., Lakshmi, J., Sozhan, G., 2011. Effects of alternating and direct current in electrocoagulation process on the removal of cadmium from water. J. Hazard Mater. 192 (1), 26–34 https://doi.org/https://doi.org/10.1016/j.jhazmat.2011.04.081.
- Zenouzi, A., Ghobadian, B., Hejazi, M.A., Rahnemoon, P., 2013. Harvesting of Microalgae Dunaliella Salina Using Electroflocculation.
- Zhao, Z., Liu, B., Ilyas, A., Vanierschot, M., Muylaert, K., Vankelecom, I.F.J., 2021. Harvesting microalgae using vibrating, negatively charged, patterned polysulfone membranes. J. Membr. Sci. 618, 118617 https://doi.org/https://doi.org/10.1016/j. memsci.2020.118617.