QATAR UNIVERSITY

COLLEGE OF ENGINEERING

HARVESTING MARINE MICROALGAE (TETRASELMIS SP.) USING

DIELECTROPHORETIC FORCE-ASSISTED ELECTROCOAGULATION

BY

WARDAN ABDULRAZAK KHATIB

A Thesis Submitted to

the College of Engineering

in Partial Fulfillment of the Requirements for the Degree of

Master of Science in Civil Engineering

January 2023

© 2023 Wardan Abdulrazak Khatib. All Rights Reserved.

COMMITTEE PAGE

The members of the Committee approve the Thesis of Wardan Abdulrazak Khatib defended on 02/01/2023.

Dr. Mohamed Arselene Ayari Thesis/Dissertation Supervisor

> Prof. Alaa AlHawari Co-Supervisor

Prof. Mo'ayyad Shawaqfah Committee Member

Prof. Abdelbaki Benamor Committee Member

Approved:

Khalid Kamal Naji, Dean, College of Engineering

ABSTRACT

KHATIB, WARDAN A., Masters: January : [2023],

Masters of Science in Civil Engineering

Title: <u>Harvesting Marine Microalgae (Tetraselmis sp.) using Dielectrophoretic Force-</u> assisted Electrocoagulation

Supervisor of Thesis: Dr. Mohamed A, Ayari.

The attention towards algal based products has been increasing drastically in recent years. The high energy requirement for harvesting microalgae stands as the main challenge of the production stream of the biomass. The presented study examined the performance of using interdigitated electrodes for harvesting marine microalgae (Tetraselmis sp.) in electrocoagulation. It was found that the new electrode array exerted a dielectrophoretic (DEP) force that improved the performance of the process compared to using a parallel of flat sheet electrodes. This phenomenon was confirmed through numerical simulation and analysis. Through experimental validation, harvesting efficiency of 82.4% was achieved with 10 mins electrolysis time when applying 50 mA/cm² current density at an inter-electrode spacing of 1 cm. Moreover, the maximum harvesting efficiency achieved was 96,18% at an inter-electrode spacing of 0.5 cm of the new electrode array. The specific energy consumption of the new electrode shows 38% enhancement than using a pair of parallel flat sheet electrode. The aluminum analysis confirmed better coagulant attachment when using the interdigitated electrode array than using the conventional flat sheet electrode. The enhanced performance of the electrocoagulation process was confirmed in terms of dielectrophoretic induction, electrode passivation, and improved current efficiency in the proposed electrode array.

DEDICATION

To my parents, professors, friends, and colleagues who contributed to the success of

this journey.

ACKNOWLEDGMENTS

I would like to acknowledge Qatar University for providing all the support to achieve the goals of this study. I would like to extend my gratitude and sincere appreciation to my supervisors Dr. Mohamed Arselene Ayari and Prof. Alaa Al-Hawari for their continuous support and guidance. Special thanks to Dr. Probir Das from the center of sustainable development, Algal Technologies Program (ATP), for providing all necessary materials to make this research successful. I would also like to thank the Central Laboratories Unit (CLU) at Qatar University for carrying out the aluminum analysis.

DEDICATIONiv
ACKNOWLEDGMENTSv
LIST OF TABLES viii
LIST OF FIGURESix
Chapter 1: Introduction
Chapter 2: Literature Review5
2.1 Microalgae Harvesting Techniques5
2.1.1 Membrane processes and filtration
2.1.2 Centrifugation
2.1.3 Flotation process11
2.1.4 Coagulation and flocculation14
2.2 Electrocoagulation
2.2.1 Mechanism
2.2.2 Previous studies on electrocoagulation process
2.3 Dielectrophoresis
Chapter 3: Materials and Methods
3.1 Microalgal Strain
3.2 Numerical Analysis
3.3 Experimental Setup & Procedure
3.4 Aluminum Content

CHAPTER 4: Results and Discussion	43
4.1 Numerical Analysis	43
4.2 Impact of Electrolysis Time	45
4.3 Impact of Current Density	48
4.4 Impact of Inter-electrode Distance	
4.5 Energy Consumption	54
4.6 Aluminum Content in the Harvested Microalgae	56
Chapter 5: Conclusion and Future Work	60
References	63

LIST OF TABLES

Table 1. Previous Studies on Harvesting Micoalgae by Filtration
Table 2. Previous Studies on Harvesting Micoalgae by Centrifugation10
Table 3. Previous Studies on Harvesting Microalgae by Flotation
Table 4. Previous Studies on Harvesting Microalgae by Bio-flocculation and Auto-
flocculation17
Table 5. Summary of the Advantages and Disadvantages of Microalgae Harvesting
Methods
Table 6. Anodic and Cathodic Reaction for Iron Electrodes (Ghernaout, 2019) 23
Table 6. Anodic and Cathodic Reaction for Iron Electrodes (Ghernaout, 2019)23Table 7. Anodic and Cathodic Reactions for Aluminum Electrodes (Ghernaout, 2019)
Table 6. Anodic and Cathodic Reaction for Iron Electrodes (Ghernaout, 2019)23 Table 7. Anodic and Cathodic Reactions for Aluminum Electrodes (Ghernaout, 2019)
Table 6. Anodic and Cathodic Reaction for Iron Electrodes (Ghernaout, 2019)

LIST OF FIGURES

Figure 1. Schematic of filtration scenarios (a) Cross-flow Filtration (b) Dead-end
Filtration (c) Forward Osmosis (d) Vacuum Filtration7
Figure 2. Scehmatic of the electrocoagulation process
Figure 3. Presentation of the electric double layer (Park & Seo, 2011)25
Figure 4. Dielectrophoretic scheme showing the positive and negative driving direction
on particles in a non-uniform elecric field
Figure 5. Algae sample used in the experimental investigation
Figure 6. Schematics of the geometry utilized for numerical study (a) EC-DEP electrode
array, (b) EC electrode array
Figure 7. Schematic diagram of the experimental setup41
Figure 8. Square of the electric field $(\nabla E ^2)$ distribution for EC-DEP module for
current density of (a) 50 mA/cm ² , (b) 40 mA/cm ² , (c) 30 mA/ cm ² & (d) 20 mA/cm ²
and for EC module with current density of (e) 50 mA/cm ²
Figure 9. Square of the electric field $(\nabla E ^2)$ distribution for EC-DEP module with inter-
Figure 9. Square of the electric field $(\nabla \mathbf{E} ^2)$ distribution for EC-DEP module with inter- electrode spacing of (a) 0.5 cm, (b) 0.75 cm, (c) 1.00 cm & (d) 1.25 cm and for EC
Figure 9. Square of the electric field $(\nabla \mathbf{E} ^2)$ distribution for EC-DEP module with inter- electrode spacing of (a) 0.5 cm, (b) 0.75 cm, (c) 1.00 cm & (d) 1.25 cm and for EC module with inter-electrode spacing of (e) 0.5 cm
Figure 9. Square of the electric field $(\nabla \mathbf{E} ^2)$ distribution for EC-DEP module with inter- electrode spacing of (a) 0.5 cm, (b) 0.75 cm, (c) 1.00 cm & (d) 1.25 cm and for EC module with inter-electrode spacing of (e) 0.5 cm
Figure 9. Square of the electric field $(\nabla \mathbf{E} ^2)$ distribution for EC-DEP module with inter- electrode spacing of (a) 0.5 cm, (b) 0.75 cm, (c) 1.00 cm & (d) 1.25 cm and for EC module with inter-electrode spacing of (e) 0.5 cm
Figure 9. Square of the electric field $(\nabla \mathbf{E} ^2)$ distribution for EC-DEP module with inter- electrode spacing of (a) 0.5 cm, (b) 0.75 cm, (c) 1.00 cm & (d) 1.25 cm and for EC module with inter-electrode spacing of (e) 0.5 cm
Figure 9. Square of the electric field $(\nabla \mathbf{E} ^2)$ distribution for EC-DEP module with inter- electrode spacing of (a) 0.5 cm, (b) 0.75 cm, (c) 1.00 cm & (d) 1.25 cm and for EC module with inter-electrode spacing of (e) 0.5 cm
Figure 9. Square of the electric field $(\nabla \mathbf{E} ^2)$ distribution for EC-DEP module with inter- electrode spacing of (a) 0.5 cm, (b) 0.75 cm, (c) 1.00 cm & (d) 1.25 cm and for EC module with inter-electrode spacing of (e) 0.5 cm
Figure 9. Square of the electric field $(\nabla \mathbf{E} ^2)$ distribution for EC-DEP module with inter- electrode spacing of (a) 0.5 cm, (b) 0.75 cm, (c) 1.00 cm & (d) 1.25 cm and for EC module with inter-electrode spacing of (e) 0.5 cm
Figure 9. Square of the electric field $(\nabla \mathbf{E} ^2)$ distribution for EC-DEP module with inter- electrode spacing of (a) 0.5 cm, (b) 0.75 cm, (c) 1.00 cm & (d) 1.25 cm and for EC module with inter-electrode spacing of (e) 0.5 cm

mA/cm ² , arc length, distance from the top edge (for EC array) and distance from the
top electrode
Figure 14. Effect of inter-electrode spacing on algal harvesting efficiency (10 minutes,
50 mA/cm ²)
Figure 15. Specific energy consumption of the electrocoagulation process for EC and
EC-DEP array at different inter-electrode spacings
Figure 16. Effect of current density on the aluminum content in the harvested
microalgae (0.5 cm)
Figure 17. Clean EC electrode surface (a), EC array at 40 mA/cm ² (b) & EC array at 50
mA.cm ² (c) and Clean EC-DEP electrode surface (d), after EC at 40 mA/cm ² (e) & after
EC at 50 mA/cm ² (f)

CHAPTER 1: INTRODUCTION

The biome of waterbodies is rich in distinct variety of microalgae (Das, Thaher, Khan, AbdulQuadir, & Al-Jabri, 2019). These microalgae can be harvested and used for different applications including production of biodiesel, biocrude oil and bioethanol (Das, Thaher, et al., 2019). Moreover, algae can be used for carbon capture, wastewater treatment and in pharmaceuticals (Das, Thaher, et al., 2019). Recently, studies have suggested that microalgae are rich in protein and can be used as food source for animal and fish (Sathasivam, Radhakrishnan, Hashem, & Abd_Allah, 2019). Since, production of algae does not require land or freshwater, it can be deemed as a suitable source of food for animals and fish in the world. The weather in many aqueous open spaces is suitable for algal growth throughout the year and large-scale production is feasible as such in the Gulf region and the Middle East (Das, Khan, et al., 2019).

The term 'algae' represents all the organisms that perform photosynthesis resulting in the production of complex sugar consuming water and CO₂ while releasing O₂. This process requires high moisture levels for continuous growth. Algae can be classified into two main categories, namely, macroalgae and microalgae. Macroalgae (also called seaweeds) are free floating and can also be seen attached at seabed, rivers and lakes, whereas microalgae are unicellular microorganisms (Raven & Giordano, 2014). Every year, about 10 million tons of microalgae are harvested by different industries for a wide variety of utilizations (Spolaore, Joannis-Cassan, Duran, & Isambert, 2006).

The commercial utilization of algal products interest has evolved and started in the western countries in the early 20th century with the demanding increased population needs. In 2004, the produced biomass of microalgae reached up to 5 million kg/year (Pulz & Gross, 2004). This led to support the national income with 1.25 billion US\$ in USA contributing to the overall economic performance of the country (Moshood, Nawanir, & Mahmud, 2021). Similarly, the number of large-scale factories for algal cultivation has increased from 46 to 110 from 1980 to 1997 in Asia (Y.-K. Lee, 1997). This attention towards increasing algal based products can be attributed to the rich nutrient content and the diverse applications potential.

The protein content of several microalgae species is greater than the protein content of vegetables. Microalgae such as Chlorella spp., Scenedesmus spp., Dunaliella spp., Aphanizomenon flos-aquae, cyanobacteria Spirulina spp. and Arthrospira are considered to be nutrient-dense food and great source of fine chemicals (Kent, Welladsen, Mangott, & Li, 2015; Koyande et al., 2019). These microalgae are rich in lipid, protein, chlorophyll, carotenoids and pigments. Moreover, microalgae are considered as a good source of vitamins A, B1, B2, B6, B12, C and E. Furthermore, microalgae possess a considerable quantity of potassium, iron, magnesium, calcium and iodine (Koyande et al., 2019). All these nutrients have made microalgae an essential ingredient of different food products, such as candy bars, gums and beverages. In addition to that, microalgae can be used as a source of natural food coloring and nutritional supplements.

Due to the severe population growth and increased demand for food, animal nurturing for meat increased dramatically in the last few decades. As a result, the demand for animal and fish food has increased worldwide. A possible food source for animal can be achieved by microalgae due to its availability with ecofriendly production characteristics (Dineshbabu, Goswami, Kumar, Sinha, & Das, 2019). Microalgal species like Pavlova, Nannochloropsis, Arthrospira, Chlorella, and Schizochytrium are rich in nutritional components such as in proteins and vitamins. As a result, they are being widely used as a fish feed to farm zooplankton, molluscs, crustaceans, shrimps and other fish types (Dineshbabu et al., 2019). Similarly, microalgae like Schizochytrium sp., Isochrysis sp. and Pavlova sp. are rich in Polyunsaturated Fatty Acid (PUFA), pigments that improve the antioxidant properties of meat and enhance meat coloration (Fabregas & Herrero, 1990). Consequently, they have been effectively used as a food component for domesticated animals (Dineshbabu et al., 2019).

One of the common applications of algal products has been enormously involved in the cosmetic market. These products were presented for skin-care use such as anti-aging products and emollients. Moreover, they can be found in sun protection and hair care products. This was the outcome of investors believing in the ability of achieving better quality of their own brands (Stolz, 2005). The protein-rich constituent of algal products was a key factor for their utilization is developing pharmaceutical care. Other application of cosmetics can be found in products with skin-tightening features that is able to enhance cell proliferation leading to natural results of skin recovery (Berthon et al., 2017).

One of the associated challenges with the algal industry biomass production stream is the harvesting and separation technology used. Microalgae can be harvested by membrane processes, coagulation and flocculation, flotation process or centrifugation (Singh & Patidar, 2018). The selection of the used technology is dependent on many factors such as energy consumption and the required quality of the produced biomass. Generally, separation of microalgae encompasses three main stages, thickening, dewatering and drying. Firstly, thickening process involves aggregating the algal mass into a concentrated square. This process can be achieved through the abovementioned technologies. Secondly, dewatering process involve the removal of the sole algae biomass from the aqueous medium. Finally, algal drying is key step former to the downstream target production stage (Barros, Gonçalves, Simões, & Pires, 2015).

The objective of this thesis is to evaluate an enhanced harvesting technique of microalgae to be utilized in many applications as discussed earlier. In the subsequent chapter, a literature review on the different harvesting technologies will be presented and compared. The advantages and disadvantages of each harvesting technique will be addressed. A literature review was made on the electrocoagulation process and its applicability in harvesting microalgae. The methodology used to carry out this research is illustrated in chapter 3. The main findings and discussion of the numerical and experimental investigations is shown in chapter 4. The final chapter will conclude this work and present a future outlook of related research.

CHAPTER 2: LITERATURE REVIEW

2.1 Microalgae Harvesting Techniques

Algal harvesting is the separation of microalgae from the aqueous solution. The algal harvesting technique depends on the energy requirement, quality of the final product, feasibility of reusing the culture medium, density, microalgal cell size and physiognomies of the cells (Uduman, Qi, Danquah, Forde, & Hoadley, 2010). The process is made complex by small cell size ($<30 \mu$ m), high zeta potential, higher algal growth rate, growth in dilute suspension and negligible difference of density between micro algae and the growth media (Uduman et al., 2010). At present, algae are harvested by chemical, biological, electrical and mechanical methods. However, Uduman et al. (2010) argued that with current technologies, a one-step algal harvesting process is not sufficient (Uduman et al., 2010). A two-step process involves concentrating of algal slurry to 2-7% TSS. The second step pertain to dewatering the slurry concentrated to 15 to 25% TSS (Brennan & Owende, 2010). The terms used for describing the efficiency of the harvesting process are the recovery efficiency (RE) and concentration factor (CF) (Pahl et al., 2013). RE and CF can be calculated as:

$$RE = \frac{mass \ of \ cell \ removed}{mass \ of \ cell \ in \ the \ initial \ culture} \tag{1}$$

$$CF = \frac{concentration of algae in the final product}{concentration of algae in the initial culture}$$
(2)

By measuring the dry weight of the harvested biomass, the mass of the microalgae can be estimated. The concentration of algae in the culture can be evaluated by measuring chlorophyll content and optical density (Pahl et al., 2013). The main algae harvesting techniques used are membrane processes, centrifugation, flotation and coagulation and flocculation (Pahl et al., 2013).

2.1.1 Membrane processes and filtration

Filtration is a dewatering process that separate microalgae from the aqueous medium which can be achieved through membrane processes. It can be done by forcing the fluid to pass through a porous membrane after applying pressure, or under gravity, or using vacuum force. After the process, microalgal cells are collected from the retaining surface (Batista et al., 2018; T.-T. Nguyen et al., 2021). The quality of the produced biomass using this process is relatively good due to the infinitesimal cell disruption and the absence of chemical involvement. The main drawback associated with filtration is fouling caused by the clogging of the membrane pores which results in increased operational and maintenance cost. Generally, for the filtration, membranes are made of polymers like polyvinylidene fluoride (PVDF), polyacrylonitrile (PAN), polytetrafluoroethylene (PTFE), polyvinyl chloride (PVC), polyether sulfone polyvinyl-pyrrolidone (PES-PVP) or polyether sulfone (PES). Researchers studying membrane filtration aim to reduce membrane fouling and to enhance the membrane flux (Hafiz, Hawari, & Altaee, 2019). Numerous filtration schemes were used in order to harvest microalgae such as dead-end filtration, cross-flow or tangential flow filtration, submerged filtration, forward osmosis, pressure and vacuum filtration (Mkpuma, Moheimani, & Ennaceri, 2022). Figure 1 demonstrates the mechanism of some filtration schemes used for microalgal separation.

According to Das et al. (2019), tangential-flow-filtration (TFF) is more suitable for the suspended microalgae because of its minor membrane fouling problems. The retentate passes tangentially through the membrane carrying the suspended matter away from the membrane pores which will significantly reduce fouling (Das, Thaher, et al., 2019). Das et al. (2019) studied the effect of salinity on harvesting microalgal biomass using TFF. A separation of 100% efficiency was obtained where there were no observations of microalgal cells in the permeate water. Das et, al. (2019) used a pilot scale TFF to harvest marine microalgae Picochlorum sp. and Tetraselmis sp. Das et al. (2019) concluded that increasing salinity will reduce the permeate flux and will increase the energy consumption of the process (Das, Thaher, et al., 2019; Fayad, Yehya, Audonnet, & Vial, 2017).



Figure 1. Schematic of filtration scenarios (a) Cross-flow Filtration (b) Dead-end Filtration (c) Forward Osmosis (d) Vacuum Filtration

Bilad et al (2012) used submerged microfilters with improved flux step method (IFM) for concentrating freshwater green microalgae Chlorella vulgaris and marine microalgae Phaeodactylum tricornutum. Bilad et al. (2012) reported that using microfiltration to concentrate microalgae is economically feasible for large scale harvesting. Efficiency of 98% in harvesting was achieved for Chlorella vulgaris with an energy consumption of 0.64 kWh/kg biomass. For Phaeodactylum tricornutum, the harvesting efficiency of 70% with an energy demand of 0.98 kWh/kg biomass using polyvinylidene fluoride PVDF membranes was achieved (Bilad, Vandamme, Foubert, Muylaert, & Vankelecom, 2012). A summary of recent studies used filtration process for harvesting microalgae is presented in Table 1.

Species	Туре	Recovery	$EC (kWh/m^3)$	Reference	
Tatrasalmis sp	LIE EO	CE-1 23	0.36	(Hafiz, Hawari, Das,	
Terraseiniis sp	01-10	CI=1.25	0.50	Khan, & Altaee, 2020)	
Chlorella vulgaris	FO	CF=4	NA	(Munshi et al., 2018)	
Chlorella vulgaris	MF	98%	0.27	(Bilad et al., 2012)	
Phaeodactylum	ME	700/	0.25	(Dilad at al. 2012)	
tricornutum	ΜΓ	/0%	0.23	(Bilau et al., 2012)	
Coelastrum	Pressure	CE 245	0.99	$(C_{\rm ent}f_{\rm et}, 1, 2012)$	
proboscideum	Chamber	Сг=243	0.88	(Cern et al., 2012)	
Coelastrum	Pressure	CE 75	0.2	$(C_{\rm ent}f_{\rm et}, 1, 2012)$	
proboscideum	Chamber	CF=75	0.3	(Cerif et al., 2012)	
Coelastrum	Pressure	CE 190	5.0	$(C_{\rm ent}f_{\rm et}, 1, 2012)$	
proboscideum	Chamber	CF=180	5.9	(Cerii et al., 2012)	
		CE 17	NT A	(Larronde-Larretche &	
Chiorella vulgaris	FU	CF=1./	INA	Jin, 2017)	

 Table 1. Previous Studies on Harvesting Micoalgae by Filtration

UF-Ultrafiltration, CF- Concentration Factor, MF- Microfiltration, FO-Forward Osmosis, EC-Energy Consumption, NA-Not Available

2.1.2 Centrifugation

In centrifugation, the algal solution is placed in a centrifuge tube connected to rotor with high speed. This high-speed rotation promotes centrifugal force that is 4,000-14,000 times stronger than the gravitational force. Despite the high separation speed and high separation efficiency, the energy consumption of this method is drastically high, and the capital cost of the centrifuge units is higher than the apparatus of the other separation technologies (Milledge & Heaven, 2011). Milledge et al. (2011) studied the effect of using disk stack centrifugation mechanism on microalgae cell disruption. He stated that a mechanical disruption may exist from hydro-mechanical forces. This disruption is preferred over the chemical disruption where the chemical disruption is associated with product contamination. There are many centrifugal configurations that can be applied to separate microalgae available in the literature. For example, disc stack configuration, nozzle discharge centrifuge and a continuous flow centrifuge unit (Najjar & Abu-Shamleh, 2020; Rios, Clavero, Salvadó, Farriol, & Torras, 2011). Table 2 summarizes recent studies used centrifugation as a separation step for microalgae.

Rios et al., (2011) used a continuous centrifugation apparatus with a preconcentrating tank to separate Nannochloris sp. from the culture medium. Low flow rate with applied continuous centrifugation led to high harvesting efficiency (>90%), though the high energy demand. Higher applied flow rate led to lower harvesting efficiency (<90%) with relatively lower cost. Maximum microalgae recovery efficiency of 94% was achieved with a continuous flowrate of 0.93 L/min and a recovery efficiency of 17% was obtained using a flow rate of 23 L/min. Generally, lower flow rate lead to higher recovery performance and vice versa (Rios et al., 2011).

Species	Туре	Recovery	EC (kWh/m ³)	Reference
Nannochloris	Continuous-flow	0.60/	20	(Dassey &
sp.	centrifuge	90%	20	Theegala, 2013)
Nannochloris	Continuous-flow	17%	0.8	(Dassey &
sp.	centrifuge	1770	0.8	Theegala, 2013)
				(Grima, Belarbi,
Haematococcus	Disc-stack			Fernández,
nluvialis	centrifuge	CF=120	1	Medina, & Chisti,
pravians	continuge			2003; Panis &
				Carreon, 2016)
Tetraselmis	Self-cleaning	70	1.2	(Tredici et al.,
suecica	disk centrifuges	70	1.2	2015)
Scenedesmus				
sp. and C.	Centrifuge nozzle	CF=15	0.9	(Grima et al.,
proboscideum				2003)
Scenedesmus				
sp. and C.	Decanter bowl	CF=22	8	(Grima et al.,
proboscideum	Centrifuge			2003)
С.				(Grima et al
proboscideum	Hydro cyclone	CF=0.4	0.3	2003)

 Table 2. Previous Studies on Harvesting Micoalgae by Centrifugation.

2.1.3 Flotation process

Flotation of microalgae occurs when an upward force carries the suspended microalgal cells to the surface of the culture medium. The lifting force is induced by providing gas bubbles to the growth medium promoting agglomeration of microalgae that can be skimmed later. This process is often preceded by coagulation/flocculation (Rubio, Souza, & Smith, 2002). This method can be fast and efficient when density of microalgal cells is low compared to the culture medium. Thus, for low density microalgae, flotation is more suitable than gravitational sedimentation. According to Singh et al. (2018), there are four flotation approaches can be used for harvesting microalgae, dissolved-air-flotation (DAF), dispersed-air-flotation (DiAF), ozonation-dispersed flotation (ODF) and electrolytic-flotation (Singh & Patidar, 2018).

Dissolved air flotation (DAF) is carried by supplying tiny bubbles at a highpressure rate sufficient for carrying suspended microalgae to the surface. Dispersed air flotation (DiAF) is achieved by passing continuous air through porous material. Electrolytic flotation is induced by bubble formation during electrolysis. Finally, ozonation-dispersed flotation (ODF) is carried out by the generation of positively charged bubbles that adhere to the negatively charged surface of microalgal cells (Singh & Patidar, 2018). These processes depend on the size of bubbles, flux, and the adhesion ability of the microalgae. Microbubbles promote faster attachment to microalgae due to their large surface area and low-rise velocity (J. Hanotu, H. Bandulasena, & W. B. Zimmerman, 2012). This method has an advantage of short processing time and is suitable for large scale microalgae harvesting with low space requirements. Gejji et al. (2018) used ionic polyelectrolytes in an organic system to increase the hydrophobicity of microalgae that enhanced flotation to obtain higher harvesting efficiency (Gejji & Fernando, 2018). Electrocoagulation-flotation separation method is commonly used as a two-step microalgae separation technique. In this method electrocoagulation is carried out using aluminum or iron electrodes. Using harvesting period of 45 mins, controlled pH of 7, and 5 mA/cm² current density, the recovery efficiency reached 100% for Microcystis aeruginosa (Gao et al., 2010). The energy consumption was 2.28 kWh/m³. During the oxidation of the electrodes, microbubbles were released from the electrodes and adhered to the microalgal flocs which resulted in the flotation of these microalgal cells. The flotation alone in this process contributes to the process efficiency by 25-50% according to (Gao et al., 2010).

Cheng et al. (2011) used ozonation-dispersed flotation to separate freshwater microalgae Chlorella vulgaris and Scenedesmus obliquus. In these studies, the aeration of the culture medium using pure oxygen did not promote flotation. However, ozone was a very good oxidizing agent for the organic matter which promoted bubble generation. In this process, during cell lysis, biopolymers were released and acted as a coagulant that promoted effective separation. The applied ozone dose in the experimental phase to achieve flotation was 0.2-0.5 mg/mg and <0.05 mg/g for Scenedesmus obliquus and Chlorella vulgaris, respectively. The turbidity removal of Scenedesmus obliquus and Chlorella vulgaris medium reached to 94.1% and 98% by ozone flotation, respectively. Cheng et al. (2011) also noticed increase in the lipid content of the microalgae which makes it useful for biofuel production. However, the researchers indicated that applying ozone flotation to a large-scale microalga harvesting environment may lead to contamination issues (Cheng et al., 2010; Cheng et al., 2011). Table 3 Shows recent studies that used flotation assisted techniques to harvest microalgae.

Species	Туре	Recovery	pН	Temperature	Time	Reference
						(J. Hanotu,
						H. C. H.
Dunaliella	Micro-	08%	5	25	_	Bandulasena,
salina	flotation	2070	5	23	-	& W. B.
						Zimmerman,
						2012)
Scenedesmus	Heat-aided	93.6%				(Xue et al
obliquus	air-	CF=25	3	70	15 min	(1100 et al., 2019)
oonquus	flotation	CI -25				2017)
Chlorella	Heat-aided	26.5%				(Xue et al
vulgaris	air-	CF=5.5	3	90	15 min	2019)
,	flotation	01 010				_01))
Scenedesmus	BDAF	>99%	5	25	10 min	(Ometto et
obliquus					- •	al., 2014)
Chlorella	BDAF	>99%	5	25	10 min	(Ometto et
vulgaris						al., 2014)
Arthrospira	BDAF	>99%	5	25	10 min	(Ometto et
maxima			10 min	al., 2014)		
						(Coward,
Chlorella sp	Foam	CF=306.89	-	-	30 min	Lee, &
I III IIIIIII	Flotation	CI =500.07			50 1111	Caldwell,
						2014)
Mixed	Vacuum	CF=130.6	-	-	60 min	(Barrut et al.,
	DiAF					2013)
Mixed	Vacuum	CF=9.9	_	_	60 min	(Barrut et al.,
111 MCU	DiAF					2013)

Table 3. Previous Studies on Harvesting Microalgae by Flotation

2.1.4 Coagulation and flocculation

Coagulation and flocculation is a simple and efficient harvesting mechanism that is carried out by the addition of coagulants that neutralizes the negative charges on the surface of the microalgae particles and promotes flocculation. The coagulating agent will reduce the electrostatic repulsion forces between microalgal suspended cells. The neutralized particles will agglomerate resulting in relatively large particles compared to the microscopic microalgae which are then separated by sweeping or gravitational sedimentation. The main drawback of this technology is the difficulty of removing excessive coagulants attached to the algal biomass that can cause contamination to the final products leading to promoted toxicity (Barros et al., 2015). According to Vandamme et al. (2013) three ways of flocculation can take place during microalgae harvesting by chemical coagulation and flocculation (Vandamme, Foubert, & Muylaert, 2013). Sweep flocculation occurs when the microalgal cells are entrapped in crowd mineral precipitation. Bridging occurs where an intermediate colloid or particle combine two particles forming a bridge between them. Electrostatic patching occurs where a polymer reverses the charge on the microparticle surface (Vandamme et al., 2013). Depending on the type of coagulants, the process can be classified into four types: Autoflocculation, Bioflocculation, Chemical coagulation/flocculation and Electrocoagulation/flocculation.

Autoflocculation or natural flocculation is a phenomenon that occurs without the addition of any supplementary chemicals to the solution. This process occurs when the pH value of the culture medium is high and CO_2 supply in the medium is low in parallel with the presence of calcium and magnesium hydroxides. During photosynthesis, the microalgae absorb the dissolved CO_2 in the medium and increases the pH. Autoflocculation is known for being inexpensive, low energy consuming process, nontoxic to the microalgal biomass, and it is less disruptive to the cells compared to other technologies. However, there is a high possibility of cellular composition alteration (Knuckey, Brown, Robert, & Frampton, 2006). The gravity settling of the flocs is known to be rapid in this method. Autoflocculation is very efficient method in the occasions of algal blooms (Singh & Patidar, 2018).

Bioflocculation process is the process where addition of microorganisms such as bacteria, fungi, and some microalgal strains produces bio-coagulants that aggregate the suspended algae into big flocs. Most of the bio-flocculants produced by bacteria are organic extracellular polymetric substances (EPSs) (Barros et al., 2015). EPSs causes cell adhesion without cell stressing or cell decomposition over an increased period of time. This adhesion will form microalgal bacterial flocs in the aqueous medium giving the ability to settle quicker than microalgal cells. Bioflocculation is highly species dependent according to (Muradov et al., 2015). Using different microorganisms will result in different separation performance. Bioflocculation has an advantage of avoiding the usage of expensive and harmful chemical flocculants (Muradov et al., 2015). Bioflocculation Harvesting process can be based on algae-algae, algae-bacteria, algae-fungi microbial partners, and it is classified as a thickening process of microalgae before dewatering (Ummalyma et al., 2017).

Bioflocculation is a very complex process that relies on high number of variables. These variables are related to the flocculating agent and the microorganismal strain to be used (Nazari, Freitag, Cavanhi, & Colla, 2020). Whereas this process also is highly dependent on the quality of the culture medium, pH, temperature, algal content, algal species, biomass density, and reaction time. Moreover, using bacteria or fungi may result in microbial contamination which will eliminate the feasibility for using microalgal biomass as a food source (Liu et al., 2014). The biological flocculation

agents are nontoxic and allows recycling of the culture medium (Ummalyma et al., 2017).

Ummalyma et al (2015) investigated the effect of three different microorganisms for harvesting Chlorella vulgaris. Applying different experimental conditions, the three microorganisms used were Ettlia texensis, Scenedesmus Obliquus and Ankistrodesmus falcatus. The maximum recovery efficiency was achieved at room temperature is 55% by Ettlia texensis (Ummalyma et al., 2017). Whereas a recovery efficiency of 93% was achieved by adding diluted solution of Paenibacillus bacteria at the same room temperature with applying adjustment of pH 5-11. However, these similar results were not seen for Anabaenaflos-aquae and Microcystis aeruginosa (Oh et al., 2001). Table 4 present previous studies on harvesting microalgae using bio-flocculation and auto-flocculation.

Species	Type/Microorganism	pН	Temperature	Time	Recovery	Reference
Desmodesmussp.	Autoflocculation	3.5	5	15d	>95%	(Chen et al., 2020)
Chlorella vulgaris	A stivisted sludge derived					
Chlamydomonas	Activated studge derived-	7.2		120	97 240/	(Choi, Hendren, Kim, Dong,
asymmetrica	extracentular polymeric	1.5	-	min	87.24%	& Lee, 2020)
Scenedesmussp.	substance (ASD-EPS)					
			25	180	72%	(Salim, Vermuë, & Wijffels,
Neochioris oleoadunaans	Aspergillus oryzae	-				2012)
Chlorella vulgaris	Aspergillus oryzae	4.5-7	25	72 h	97%	(Zhou et al., 2013)
Chlorella vulgaris	Aspergillus oryzae	4-5	25	72 h	93%	(Zhou et al., 2013)
Chlorella vulgaris	Aspergillus oryzae	4-5	25	48 h	63%	(Zhou et al., 2013)
Chlorella vulgaris	Paenibacillussp.	5-11	-		93%	(Oh et al., 2001)
Chlorella vulgaris	Cunninghamella echinulata	4-5	25	24 h	99%	(Xie, Sun, Dai, & Yuan, 2013)
N	bacterium Streptomycessp	7	21	9 min	99.18%	(Sivasankar, Poongodi, Lobo,
Nannochloropsis sp.						& Pugazhendhi, 2020)
Scenedesmus rubescens SX	Autoflocculation	10	-	10 min	92.0±6.0%	(T. D. P. Nguyen et al., 2019)
N	Solibacillus silvestris	8.7	25		88%	(Wan, Zhao, Guo, Alam, &
Nannochioropsis oceanica				-		Bai, 2013)

Table 4. Previous Studies on Harvesting Microalgae by Bio-flocculation and Auto-flocculation

Species	Type/Microorganism	рН	Temperature	Time	Recovery	Reference
Chlorella muneroidosa	Waste eggshell	4	25	30 min	99%	(Pandey, Pathak, Kothari,
Chioretta pyrenotaosa						Black, & Tyagi, 2019)
Chlonellammanaidana	Aspergillus fumigatus	4.5	60	240	000/	(Bhattacharya, Mathur,
Chiorenapyrenoidosa				min	90%	Kumar, & Malik, 2019)
Chlouellagen ge	Disurctus estructus	3-6	25	150	61 960/	$(\mathbf{I}, \mathbf{I}, \mathbf{I}, \mathbf{O}(\mathbf{I}))$
Chiorellasp sp.	Pleurotus ostreatus			min	04.80%	(Luo et al., 2019)
Chlorellasp	A probio potivotod sludo					
Pediastrum sp	Retoric activated stude		22.20	00 min	0.00/	(Van Den Hende, Vervaeren,
Phormidium sp.		-	23-29	90 mm	90%	Desmet, & Boon, 2011)
Scenedesmus sp.	(Wastewater) s sp.					
Pleurochrysis carterae	Destarial in coulum (Ten water)	-	-	30 min	93%	(A. K. Lee, Lewis, & Ashman,
	Daciental moculum (Tap water)				CF=131	2009)
Chlorella vulgaris	Scenedesmus obliquus	-	25	180	55%	(Salim et al., 2012)
Chlorella vulgaris	Ankistrodesmus falcatus	-	25	180	34%	(Salim et al., 2012)
Chlorella vulgaris	Tetraselmis suecica	-	25	180	50%	(Salim et al., 2012)

Chemical coagulation and flocculation is universally used in water and wastewater treatment to remove suspended matter and turbidity. This method has been studied to be a promising universal method due to its applicability to a wide variety of algae species. Additionally, the energy requirements associated with this method is small compared to other separation technologies. In this method, the addition of cationic compounds will neutralize the negative charges on the surface of microalgae particles and help in the formation of flocs by coalescence of these suspended particles. Chemical coagulation is relatively cheap when applied to large-scale microalgae harvesting environment. Electro negativity and the solubility of the used coagulant are the two main important factors for the determination of the coagulation effectiveness. Ideally, less solubility with high electronegative ions will achieve higher separation efficiency (Barros et al., 2015). For chemical coagulation and flocculation ferric chloride, ferric sulphate, and aluminum sulphate are widely used as coagulants. Such coagulants may cause changes to the culture medium and increase the total dissolved solids in the solution. Chemical coagulation can be applied for microalgae thickening as a pre-treatment step for dewatering to reduce energy consumption and increase the effective particle size of microalgae. One of the advantages associated with this process is regarded to the recyclability of the culture medium (Ummalyma et al., 2017).

The process used for harvesting microalgae in this study falls under the concept of coagulation and flocculation and it is discussed in detail in the following section. The major advantages and disadvantages of different algal harvesting techniques are summarized in Table 5.

Harvesting Method	Advantages	Disadvantages
Membrane processes	 High recovery efficiency 	Membrane fouling and clogging is huge
	Cost effective	issue
	No use of chemicals	 Periodic maintenance and membrane
	Low energy consumption	replacement are required
	Water is recycled	
Centrifugation	➢ Fast method	Very expensive method
	High separation efficiency	High energy demand
	 Applicable to all microalgal species 	Difficult to be applied in a large scale
		Cell disruption may exist
Flotation	Suitable for large scales	May require the use of chemical
	Low space requirements	flocculants
	Short operation time	Ozone flotation is expensive
Coagulation and flocculation		
Auto-flocculation	In expensive and Easy	Existence of some minerals is required
	Allow recycling of the broth	Mineral contamination may occur
	 Low potential of toxicity and contamination 	
	Used for large scale	
	Less cell damages	
	No energy requirements	

Table 5. Summary of the Advantages and Disadvantages of Microalgae Harvesting Methods

Harvesting Method	Advantages	Disadvantages
Bio-flocculation	In expensive and Easy	 Highly pH dependant
	Allow recycling of the broth	 Highly species dependant
	Low potential of toxicity	 Potential of microbial contamination
	Low energy requirements	 Changes in cellular composition
Chemical Coagulation/flocculation	Simple and fast method	 Expensive chemical coagulants
	No energy requirements	Chemicals can cause toxicity to the
	Applicable to a wide variety of	produced biomass
	microalgal stains	 Recyclability of the aqueous solution medium is limited
Electrocoagulation/flocculation	 High recovery efficiency 	High energy demand
	 Can be applied to a broad variety of strains 	 Potential for metallic contamination
	No use of chemicals is involved	
	Long processing time	

2.2 Electrocoagulation

2.2.1 Mechanism

In electrocoagulation, sacrificial electrodes are used to destabilize the suspension of algal particles in the aqueous medium. From the sacrificial electrodes, and through electrolytic oxidation, 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, Bourniquel, Danquah, & Hoadley, 2011). Electrocoagulation encompasses three stages. 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 or through gravitational sedimentation to the base of the reactor (Matos, Santos, Nobre, & Gouveia, 2013). A schematic of the electrocoagulation process using aluminum electrodes is shown in Figure 2. During electrocoagulation using aluminum electrodes, the following electrochemical reaction takes place in the reactor (Ghernaout, 2019):

$$AI \to AI^{3+} + 3e^{-} \tag{3}$$

$$2H_2 0 \to 0_2 + 4H^+ + 4e^- \tag{4}$$

$$Al^{3+} + 3H_20 \leftrightarrow Al(0H)_3 + 3H^+$$
(5)

$$2H_20 + 2e^- \rightarrow H_2 + 20H^-$$
 (6)

Complete reaction would be:

$$2AI + 6H_2O \leftrightarrow 2AI(OH)_3 + 3H_2 \tag{7}$$

The anodic and cathodic reactions of different electrode materials, namely iron and aluminum are presented in Table 6 and Table 7.

Table 6. Anodic and Cathodic Reaction for Iron Electrodes (Ghernaout, 2019)

Reaction mechanism for Iron (Fe) electrodes			
Mechanism #1 (pH 2)	Anodic	$2Fe_{(s)} - 4e^- \rightarrow 2Fe_{(aq)}^{2+}$	
		$2H_2 O_{(l)} - 4e^- \to O_{2(g)} + 4H^{2+}_{(aq)}$	
	Cathodic	$2H^+_{(aq)} + 8e^- \rightarrow 4H_{2(g)}$	
Mechanism #2 (pH 7)	Anodic	$2Fe_{(s)} - 4e^- \rightarrow 2Fe_{\prime(aq)}^{2+}$	
		$2Fe_{(aq)}^{2+} - e^- \to Fe_{(aq)}^{3+}$	
		$Fe_{(s)} - 3e^- \rightarrow Fe_{(aq)}^{3+}$	
	Cathodic	$8H_2O_{(l)} + 8e^- \to 4H_{2(g)} + 80H_{(aq)}^-$	
Mechanism #3 (pH 12)	Anodic	$2Fe_{(s)} - 6e^- \rightarrow 2Fe_{(aq)}^{3+}$	
	Cathodic	$6H_2O_{(l)} + 6e^- \to 3H_{2(g)} + 60H_{(aq)}^-$	

Reaction mechanism for Iron (Fe) electrodes

Table 7. Anodic and Cathodic Reactions for Aluminum Electrodes (Ghernaout, 2019)

Reaction mechanism for aluminum (AI) electrodes			
Mechanism #1 (pH 7)	Anodic	$Al_{(s)} - 3e^- \rightarrow Al_{(aq)}^{3+}$	
		$2H_2 O_{(l)} - 4e^- \to O_{2(g)} + 4H^+_{(aq)}$	
	Cathodic	$4H_2O_{(l)} + 4e^- \rightarrow 2H_{2(g)} + 40H_{(aq)}^-$	
		$Al_{(s)} + 40H_{(aq)}^ 3e^- \rightarrow AL(0H)_{4(aq)}^-$	



Figure 2. Scehmatic of the electrocoagulation process.

The suspended matter in water bodies carry negative charge on its surface. The stability of these particles in the medium can be obtained by holding similar charges. As a result, suspension of particles is achieved due to the generated repulsion columbic forces. Nevertheless, the overall net charge of the medium is neutral due to the existence of positive ions. This led to the formation of an electric double layer. The electric double layer is a phenomenon that plays a key role in colloids electrostatic stabilizing mechanism. This occurs due to the adsorption of negatively charged ions to the surface of the suspended matter. This initiates a potential on the surface of the particle and decreases gradually towards the outer layer of the electric double layer (Doggaz, Attour, Mostefa, Tlili, & Lapicque, 2018). The electric double layer is illustrated in Figure 3. As shown in Figure 3, the formation of the electric double layer includes three stages (Park & Seo, 2011). The first layer is the surface charge layer where the negatively

charged ions settle on the surface of the particle. The second layer is the stern layer (or the Helmholtz layer) which holds counterions that is attracted through the electrostatic force. The last layer is called the diffuse layer. A higher concentration of counterions exist in this layer where it is affected by the charged particles in the surrounding (Doggaz et al., 2018; Park & Seo, 2011). The overall net charge of the particle will become neutral as a result. The destabilization mechanism of the suspended matter occurs due to the produced charged positive coagulants interacting with the existed negative particles. This obstructs the electrostatic repulsion of the stabilized particle. Thus, reducing the electric double layer thickness and promoting floc formation. The formed floc will eventually settle or float by the hydrogen gas forming a thick layer of matter at the surface of the broth. This will be demonstrated in the subsequent section.



Figure 3. Presentation of the electric double layer (Park & Seo, 2011)

2.2.2 Previous studies on electrocoagulation process

Electrocoagulation is one of latest methods used for microalgae separation. Electrocoagulation avoids the use of chemicals which makes it an environmentally friendly process. Additionally, avoidance of chemicals will decrease the potential of producing toxic microalgal products. Nevertheless, metallic contamination may occur (Vandamme, Cláudia Vieira Pontes, et al., 2011). Electrocoagulation/flocculation is also known for being highly efficient with efficiency ranging between 80-99% (Hawari, Alkhatib, Das, Thaher, & Benamor, 2020). The factors and parameters that affects the harvesting efficiency in electrocoagulation are electrode configuration, electrode material, applied current, electrolysis time, source of current, pH, temperature and salinity.

In electro-coagulation-flocculation, sacrificial and non-sacrificial electrodes can be used. Non-sacrificial based methods involve the movement of the negatively charged microalgae cells toward the anode in the solution without any metallic dissolution (Barros et al., 2015). The negative charges are lost from the surface of microalgae once reaching the anode, and aggregation of microalgae takes place. The use of non-sacrificial electrode does not avoid the use of coagulant and mostly used to avoid metallic contamination and reduce cost. Misra et al. (2014) investigated the validity of using non-sacrificial electrodes for producing sustainable biodiesel from microalgae. He used carbon electrodes with DC current source to achieve maximum harvesting efficiency of 94.52% with an energy consumption of 1.6 kWh/kg after 60 mins processing time. In this study, NaCl (6 g/L) was added in the solution to increase the harvesting efficiency of the freshwater microalgae Chlorella sorokiniana through increasing its electric conductivity (Misra, Guldhe, Singh, Rawat, & Bux, 2014).

Sacrificial electrodes are widely used for this process due to its high efficiency.
Unlike the non-sacrificial electrode, metallic ions, namely, the coagulants, are released to the solution from the electrodes due to electrolytic oxidation induced by the applied current. These ions will destabilize the solution due to the reduction of zeta potential charge of the microalgae surface and promotes agglomeration. Vandamme et al. (2011) used aluminum and iron electrodes in his experiment for harvesting Chlorella vulgaris (Vandamme, Cláudia Vieira Pontes, et al., 2011). The results showed that the usage of aluminum is more efficient than iron due to several reasons. Firstly, the energy requirement for iron electrodes is significantly higher than the aluminum electrode due to its low current efficiency. Moreover, Aluminum electrodes dissociates at a higher rate than iron electrodes. Furthermore, the oxidation of ferric elements from the electrode resulted in appearance of brownish color on the microalgae surface due to attachment of ferric oxides. In this study, 80% Chlorella vulgaris was obtained with 0.5 kWh/kg energy demand (Vandamme, Cláudia Vieira Pontes, et al., 2011).

Shi et al. (2017) used aluminum (Al) electrodes to extract microalgae from freshwater using electrocoagulation (Shi et al., 2017). A harvesting efficiency of 98% was achieved after 7 mins of electrolysis time by applying 6.7 mA/cm² direct current (DC). The energy consumption was 0.29 kWh/kg of algae (Shi et al., 2017). Fayad et al. (2017) used electrocoagulation process to harvest algae using aluminum (Al) electrodes. An extraction efficiency of 40% was achieved by applying 2.9 mA/cm² current density using a direct current source (DC) for 60 minutes (Fayad et al., 2017). The extraction efficiency was improved to 95.8% by Kim et al. (2012) by applying a DC current density of 8.2 mA/cm² for 15 minutes (Kim, Ryu, Kim, Han, & Yang, 2012b). By applying a current density of 8.3 mA/cm² for 10 minutes application time, Matos et al. (2013) obtained 99% microalgal recovery (Matos et al., 2013). Uduman et al. (2011) used stainless steel electrodes and achieved 99% extraction efficiency by

applying 1.5 mA/cm² current density for 15 minutes (Uduman et al., 2011). Although the applied current density was low, the energy requirement using stainless steel was 20 times higher than that of aluminum (Al) electrodes. All the previous mentioned researchers used a direct current (DC) power source. Contrary, Hawari et al. (2020) studied the effect of dielectrophoresis (DEP) on electrocoagulation by using AC power source (Hawari et al., 2020). Hawari et al. (2020) used cylindrical electrode configuration to separate microalgae from seawater and achieved 90.9% separation efficiency after 10 mins electrolysis time. The applied current density was 7.1mA/cm² and the energy demand was 4.62 kWh/kg (Hawari et al., 2020). Table 8 presents previous studies used electrocoagulation for harvesting microalgae under different conditions.

One of the main disadvantages of electrocoagulation are the high potential for metallic contamination and the high energy consumption. These problems can be reduced by inducing dielectrophoretic force through changing the shape of the electrodes. In this research, a new interdigitated electrode array will be tested accompanied by AC power source.

Microalgae strain	Electrode Material	Source of Current	Harvesting Efficiency (%)	Current Density (mA/cm ²)	Electrolysis Time (min)	Settlement Time (min)	Electrode Consumption (%)	Energy Demand (kWh/kg)	Reference
Fresh water (Chlorella vulgaris)	Carbon (Non- sacrificial)	DC	94.52	1.5	60	30	NA	1.6	(Misra et al., 2014)
Marine (Phaeodactylum tricornutum)	Al	DC	80	0.6, 3	30, 10	30	< 2.5, NA	0.3, 0.5	(Vandamme, Pontes, et al., 2011)
Freshwater (Chlorella vulgaris)	Al	DC	95	12	10	30	NA	2	(Vandamme, Pontes, et al., 2011)
Marine (Nannochloris oculate)	Al	DC (Polarity exchange)	95.8	2.7, 5.4, 8.2	15, 10, 5	NA	≤4	< 3	(Kim, Ryu, Kim, Han, & Yang, 2012a)
Marine (Dunaliella salina)	Al	DC	94.9	9	3 in a 300 ml beaker	8 min mixing and 2 min settlement	NA	0.13	(Zenouzi, Ghobadian, Hejazi, & Rahnemoon, 2013)
Marine (Tetraselmis sp.)	Stainless steel	DC	99	1.5	15	60	NA	9.16	(Uduman et al., 2011)

Table 8. Previous Studies on Harvesting Microalgae by Electrocoagulation

Microalgae strain	Electrode Material	Source of Current	Harvesting Efficiency (%)	Current Density (mA/cm ²)	Electrolysis Time (min)	Settlement Time (min)	Electrode Consumption (%)	Energy Demand (kWh/kg)	Reference
Marine (Chlorococcum sp)	Stainless steel	DC	98	1.5	15	60	NA	4.44	(Uduman et al., 2011)
Marine (Nannochloropsis sp.)	Al	DC	97	8.3	10	30	1.39	NA	(Matos et al., 2013)
Fresh water (Chlorella vulgaris)	Al	DC	40, 72, 99	2.9, 4.8, 6.7	60	45	≤2.1	1.6	(Fayad et al., 2017)
Fresh water (Chlorella vulgaris)	Al	DC	98	2.2, 4.4, 6.7	7, 6, 4	10	1.6, 4.2, 4.9 (mg/L)	0.087, 0.222, 0.294	(Shi et al., 2017)
Marine (<i>Tetraselmis</i> sp.)	Al	AC	90.9	7.1	30, 10 (@ optimum condition)	60	NA	4.62	(Hawari et al., 2020)

2.3 Dielectrophoresis

A dielectrophoretic (DEP) force is a force generates on the dielectric particles in a non-uniform electric field (Hawari, Du, Baune, & Thöming, 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. This force is referred to as the dielectrophoretic force (Cetin & Li, 2011). Two types of DEP forces can affect the suspension of particles: positive DEP (pDEP) and negative DEP (nDEP). The particles will be pulled towards the stronger electric field if the permittivity of the particle is higher than the surrounding solution. Thus, pDEP will be created. Further, weaker electric field will attract the particles if their permittivity were lower than the permittivity of the aqueous medium, inducing nDEP (Du, Hawari, Baune, & Thöming, 2009a; Çetin & Li, 2011). In general, the determination of the dielectrophoretic effect on the particle is attributed to the polarizability of the particle compared to the surrounding medium. Figure 4 illustrates dielectrophoresis affecting particles in a 2dimensional space. If the particle is attracted towards the regions closer to the electrodes with the high electric field, it can be classified as pDEP (Yu et al., 2005). The type of DEP force exerted on a particle can be classified using the Clausius-Mossotti factor (\tilde{K}) that is calculated using equation (8):

$$\tilde{K} = \frac{\tilde{\varepsilon}_{p} - \tilde{\varepsilon}_{M}}{\tilde{\varepsilon}_{p} + 2\tilde{\varepsilon}_{M}} = \frac{\varepsilon_{p} - \varepsilon_{m} - \frac{j(\sigma_{p} - \sigma_{m})}{\omega}}{\varepsilon_{p} + \varepsilon_{m} - \frac{j(\sigma_{p} - \sigma_{m})}{\omega}}$$
(8)

$$\tilde{\varepsilon} = \varepsilon - \frac{j\sigma}{\omega} \tag{9}$$

Here, $\tilde{\epsilon}_M$ 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 $\left(\frac{\text{rad}}{s}\right)$, 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 (10):

$$\mathbf{j} = \sqrt{-1} \cdot (\mathbf{E} \cdot \nabla) \mathbf{E} = \frac{1}{2} \nabla |\mathbf{E}|^2 \tag{10}$$

The complex permittivity is used to replace the absolute permittivity by using alternating current (AC). Finally, the DEP force acting on a particle with radius r can be calculated using (11) (Hawari et al., 2015):

$$F_{DEP} = 4\pi a^3 \varepsilon_0 \varepsilon_M re[\tilde{K}] (E \cdot \nabla) E \tag{11}$$



Figure 4. Dielectrophoretic scheme showing the positive and negative driving direction on particles in a non-uniform elecric field.

It was shown that DEP force can manipulate suspended particles in an aqueous medium in a nano and micro scale. Thereby, DEP effect has been applied extensively in micro cytometry. Fuhr et al., 1992 used interdigitated electrode configuration to collect yeast cells (Fuhr et al., 1992). Similarly, Huang et al., 1993 utilized DEP force in a micro channel to direct yeast cells in a linear pattern in a microfluidic channel (Huang, Wang, Tame, & Pethig, 1993). This has been further extended to control the flow of viruses in a positive or negative pattern using DEP. Moreover, it was found that biological polymers such as DNA can be concentrated in a microfluidic channels using negative DEP (Fiedler, Shirley, Schnelle, & Fuhr, 1998; Morgan, Holmes, & Green). Particle manipulation was extensively studied for medical and biological purposes in a micro level. Nevertheless, the application of DEP effect in larger scale is scarce and limited in the literature. Most recently, Hawari et al., 2015 evaluated the dielectrophoretic effect on multiple applications in wastewater treatment (Hawari et al., 2015). The authors used dielectrophoretic force to decrease the fouling and recover the filtrate flux in submerged membrane bioreactors. It was shown that the electric field can be correlated directly to the fouling suppression performance leading to enhanced flux. Moreover, Almukdad et al., 2020 generated a non-uniform electric field in order to evaluate the performance against removing heavy metals from wastewater in electrocoagulation (Almukdad, Alfahel, & AlHawari, 2020). The generated electric field was able to remove Fe and Mn ions higher that were found by generating a uniform electric field. This can open a promising potential for DEP implementation in water and wastewater treatment applications.

It was found by (Alkhatib, Hawari, Hafiz, & Benamor, 2020) that the performance of the electrocoagulation process can be further enhanced by inducing dielectrophoretic (DEP) force in the reactor. This study investigates the performance of a new cylindrical interdigitated electrodes (IDEs) array in the electrocoagulation process. The new electrode array 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, the new electrode array is expected to improve algal harvesting efficiency. The impact of current density, electrolysis time and inter-electrode spacing 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 aluminum content in the harvested microalgae and energy consumption will also be evaluated.

CHAPTER 3: MATERIALS AND METHODS

3.1 Microalgal Strain

In this study, marine microalgae (Tetraselmis sp.) sample 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). The algal broth was found to have an optical density of 0.300. Table 9 summarizes the initial characteristics of the algal broth. A sample of the algal broth is shown in Figure 5.

Table 9. Initial Characteristics of the Algal Broth

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



Figure 5. Algae sample used in the experimental investigation.

3.2 Numerical Analysis

To investigate the impact of the dielectrophoretic force in the proposed electrocoagulation setup, a numerical simulation model was built using COMSOL Multiphysics 5.5. As seen from equations (10) and (11), 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 Figure 6. The effect of current density and inter-electrode spacing on the square of the electric field was assessed in both geometries. While studying the effect of current density, the inter-electrode spacing 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 analyzing the effect of the interelectrode spacing, 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 inter-electrode spacing. For comparison, the square of the electric field of the EC array was also evaluated at an inter-electrode spacing 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. The quasi-electrostatic approximation was adopted to solve for the current densities. As such, The root mean square (rms) of the electric field is calculated using equation (12) (Du, Hawari, Baune, & Thöming, 2009b):

$$\mathbf{E} = -\nabla \boldsymbol{\varphi} \tag{12}$$

Here, φ is the rms of the electrostatic potential which can be given by the Laplace's equation (13):

$$\nabla^2 \varphi = 0 \tag{13}$$

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

$$\varphi_1 = U_o \tag{14}$$

$$\varphi_2 = 0 \tag{15}$$

Here, U_0 is the rms of the oscillating potential drop. To ensure mesh-independent results, adaptive mesh refinement (AMR) in the numerical analysis was used.



Figure 6. Schematics of the geometry utilized for numerical study (a) EC-DEP electrode array, (b) EC electrode array.

3.3 Experimental Setup & Procedure

The schematics of the experimental setup of the electrocoagulation process is shown in Figure 7. All the experiments were carried out in a graduated beaker with a volume of 1L. 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 study, this electrode array will be referred to EC-DEP. The two parallel flat sheet aluminum electrodes have an area of 40.82 cm². This electrode array will be referred as EC in this study. Using these two electrode arrays, the effect of electrolysis time, inter-electrode spacing and current density on the algal harvesting process were 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 minutes of settling time. The optical density of the collected samples was then measured. The algal harvesting efficiency (η) was calculated using equation (16):

$$\eta = \left(\frac{OD_0 - OD_t}{OD_0}\right) 100\% \tag{16}$$

39

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 with rubbing sandpaper and washed with distilled water to remove any precipitates. Furthermore, the energy consumption C_{energy} (kWh/m³) was calculated using equation (17):

$$C_{\text{energy}} = \frac{U \times I \times t}{1000 \times v \times C_{i}}$$
(17)

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 (18):

$$R = \frac{\rho L}{A} \tag{18}$$

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



Figure 7. Schematic diagram of the experimental setup.

3.4 Aluminum Content

To measure the aluminum content in the harvested algal biomass, the harvested algae samples were freeze dried for 1 day. 10mg 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°C in a furnace for 5 hours. 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 0.2µm pore size syringe filter (GD/X Whatman, UK). Finally, the samples were analyzed by inductively coupled plasma optical emission spectroscopy (ICP-OES) (Perkin OPTIMA 7300 DV, USA). The dissolved aluminum content was also evaluated after the electrocoagulation process with ICP-OES. 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.

CHAPTER 4: RESULTS AND DISCUSSION

4.1 Numerical Analysis

In the numerical study, the effect of current density and inter-electrode spacing on the square of the electric field was evaluated. The effect of current density was studied at an inter-electrode spacing of 1cm. For the EC-DEP 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. Figure 8 shows the effect of current density on the square of the electric field distribution for the studied electrode arrays. As seen from Figure 8, for the EC-DEP electrode array, the maximum squared electric field intensity of $1 \times 10^9 \text{ V}^2/\text{m}^3$ was found at the surface of the electrodes and the intensity decreased with decreasing current density. Whereas for the EC electrode array, the square delectric field intensity was found only around the top and bottom edges of the plates.

The effect of the inter-electrode spacing 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 analyzed with 0.5 cm inter-electrode spacing. Figure 9 shows the square of the electric field distribution for the studied electrode arrays. As seen in Figure 9, the maximum squared electric field intensity of $4 \times 10^9 \text{ V}^2/\text{m}^3$ is observed at the surface of the electrodes in the EC-DEP array. However, as the inter-electrode spacing increases, the area of maximum squared electric field intensity around the surface of the electrode decreased (Figure 9 (a), (b), (c) and (d)). Furthermore, the numerical study on the EC array shows that the squared electric field intensity was found only around the edges of the plates (Figure 9 (e)). Hence, indicating minimal DEP force distribution is the EC array setup compared to the EC-DEP array.



Figure 8. 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.



Figure 9. Square of the electric field $(\nabla |E|^2)$ distribution for EC-DEP module with inter-electrode spacing of (a) 0.5 cm, (b) 0.75 cm, (c) 1.00 cm & (d) 1.25 cm and for EC module with inter-electrode spacing of (e) 0.5 cm.

4.2 Impact of Electrolysis Time

The impact of electrolysis time on the algal harvesting efficiency was studied using EC and EC-DEP arrays. The inter-electrode spacing and applied current density were fixed at 1 cm and 50 mA/cm², respectively. Figure 10 (a) shows the effect of electrolysis time on the algal harvesting efficiency. As seen in Figure 10, after one minute, the algal harvesting efficiency for both electrode arrays was 6.89%. After 20 minutes 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 (3)) (Hawari et al., 2020). Between a pH of 5 and 7, these Al^{3+} 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. This will allow the Van der Wall's force to dominate (Dayarathne, Angove, Aryal, Abuel-Naga, & Mainali, 2021). 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 H_2 and consumption of OH^- by 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 Aluminum (Arain et al., 2015). During this study, a pH of 5.32 and 5.57 was recorded after 20 minutes of electrocoagulation using the EC-DEP and EC electrode array, respectively. Although electrocoagulation of both electrode arrays resulted in similar harvesting efficiency after 20 minutes of operation, EC-DEP electrode array reached 82.4% algal harvesting efficiency within 10 minutes. Whereas the EC electrode array reached 59.9% algal harvesting efficiency after 10

minutes. 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., 2009b). The presence of an intense DEP force in the EC-DEP array was confirmed by the numerical study in section 4.1. The DEP force will promote collision among microalgae and assist the Van der Waal's force in promoting coagulation (Hawari et al., 2015).



(a)



(b)

Figure 10. Effect of electrolysis time on harvesting efficiency (1 cm inter-electrode spacing, 50 mA/cm²) (a) % Removal (b) Clarity

4.3 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 inter-electrode spacing was maintained at 1 cm and the electrocoagulation process was carried out for 10 minutes. The studied current densities were 20, 30, 40 and 50 mA/cm². Figure 11 shows the effect of current density on the algal harvesting efficiency. After electrolysis, layer aggregation took place and settled by gravity to the bottom of the medium. Moreover, algal layer was formed by the flotation of the generated hydrogen bubbles to the surface of the medium as presented in Figure 12. As seen in Figure 11, using the EC array, 24.9%, 39.1%, 47.2% and 59.9% algal harvesting efficiency were 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 were 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 increased the algal harvesting efficiency. Increasing the current density increases the production rate of Al^{3+} in the reactor (Gao et al., 2010). Production of more Al^{3+} 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 Figure 11 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 Waal'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 Figure 8, 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 Figure 13. Figure 13 shows the electric field intensity at the surface of the EC and the EC-DEP electrode arrays. As seen from Figure 13 (a), for the EC-DEP array, the highest electric field intensity was 2,900 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 1,700 V/m was observed. Whereas Figure 13 (b) shows that for the EC electrode array, the electric field intensity at the top and bottom edges of the electrode was 2,900 V/m. Away from the edges, the electric field intensity remains constant at 1,900 V/m. The recurrence of high electric field intensity in the proposed EC-DEP electrode array would result in more production of aluminum in the electrocoagulation process which will enhance the harvesting efficiency. The more production of aluminum using the EC-DEP array compared to the EC array was confirmed in the amount of aluminum in the harvested microalgae. The amount of aluminum in the harvested algae is explained further in section 4.6.



Figure 11. Effect of applied current on harvesting efficiency (1 cm inter-electrode spacing, 10 minutes electrolysis time)



Figure 12. Formation of algal surface layer and sludge sedimentation after electrocoagulation







Figure 13. Electric field intensity at the surface of (a) EC-DEP electrode array and (b) EC electrode array. (inter-electrode spacing 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.

4.4 Impact of Inter-electrode Distance

The impact of inter-electrode spacing on the algal harvesting efficiency was studied for both EC and EC-DEP arrays. The studied inter-electrode spacings 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 minutes, respectively. Figure 14 presents the impact of the inter-electrode spacing on the algal harvesting efficiency. From Figure 14 it can be seen that for EC electrode array, 0.5, 0.75, 1 and 1.25 cm interelectrode spacing resulted in harvesting efficiency of 92.9%, 87.5%, 59.9% and 53.5%, respectively. Whereas, for the EC-DEP electrode array, inter-electrode spacing 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 inter-electrode spacing increased the harvesting efficiency due to the reduced electrical resistance in the electrocoagulation reactor (Du et al., 2013; Gao et al., 2010). Ghosh et al. (2008) also suggested to use lower inter-electrode spacing to improve effectiveness of electrocoagulation and to reduce the energy consumption (Ghosh, Solanki, & Purkait, 2008). From Figure 14 it can also be observed that, for 0.5, 0.75, 1 and 1.25 cm interelectrode spacing, 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 4.1. Figure 14 also shows 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 inter-electrode spacing of 0.50, 0.75, 1.00 and 1.25cm, respectively in the EC electrode array. The difference in the EC-DEP electrode array is not very

significant because even at higher inter-electrode spacings, the EC-DEP electrode array exhibits electric field intensity higher that 2,000 V/m. This high electric field intensity is exhibited due to lower resistance in the EC-DEP electrode array which will result in additional aluminum production during electrocoagulation. The additional aluminum is found in the harvested algae and is discussed in detail in section 4.6.



Figure 14. Effect of inter-electrode spacing on algal harvesting efficiency (10 minutes, 50 mA/cm²).

4.5 Energy Consumption

The specific energy consumption of the electrocoagulation process with EC and EC-DEP electrode arrays was studied for current densities of 20, 30, 40 and 50 mA/cm². For this study, the electrolysis time and inter-electrode spacing were kept constant at 10 mins and 1 cm, respectively. The specific energy consumption was calculated using equation (17). Figure 15 shows the effect of current density on the specific energy consumption of EC and EC-DEP electrode array. As seen from Figure 15 using the EC electrode array with inter-electrode spacing 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 Figure 15 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 (17). Moreover, Figure 15 shows that the EC-DEP electrode array results in lower energy consumption than the EC electrode array for all 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).



Figure 15. Specific energy consumption of the electrocoagulation process for EC and EC-DEP array at different inter-electrode spacings.

4.6 Aluminum Content in the Harvested Microalgae

In this study, aluminum analysis was done on the harvested algal biomass and the algal broth. The algae and the algal broth used for aluminum analysis was collected after electrocoagulation at an inter-electrode spacing of 0.5 cm where the electrolysis time was 10 mins using both EC and EC-DEP electrode arrays. Figure 16 shows the aluminum content in the harvested microalgae and algal broth at current densities of 20, 30, 40 and 50 mA/cm². As seen from Figure 16, the EC electrode array resulted in 3.10, 11.39, 17.02 and 15.48 mg/g aluminum 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 aluminum in the harvested microalgae for current densities of 20, 30, 40 and 50 mA/cm², respectively. As observed from these results, while using the EC-DEP array, increasing the current density increased the aluminum content in the harvested microalgae. This is because, at higher current density more aluminum hydroxide Al(OH)₃ would be produced in the electrocoagulation reactor (Arain et al., 2015). This additional aluminum hydroxide resulted in higher algal harvesting efficiency and higher aluminum content in the harvested microalgae. However, this trend was not observed while using EC electrode array as increasing current density from 40 mA/cm² to 50 mA/cm² decreases the aluminum content from 17.02 and 15.48 ppm. The reasons for reduced aluminum content in the harvested microalgae while using the EC array is current efficiency and electrode passivation. Moreover, Figure 16 also shows that, increasing the current density decreases the aluminum content in the algal broth. Although at higher current density, more aluminum is being produced, the produced aluminum is being spent for coagulation of the microalgae. This is proved by the increasing aluminum content in the harvested microalgae at higher current densities.



Figure 16. Effect of current density on the aluminum content in the harvested microalgae (0.5 cm).

Current efficiency can be defined as the actual mass of a substance liberated from an electrolyte by the passage of current divided by the theoretical mass liberated according to Faraday's law (Ahmadi & Ghanbari, 2016; Izquierdo et al., 2010). The higher amount of aluminum 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, the aluminum hydroxide Al(OH)₃ formed during the electrocoagulation process will deposit on the electrode surface and cause electrode passivation. Figure 17 shows the electrode surface of the EC and EC-DEP array before and after the electrocoagulation process. As seen in Figure 17 (b) and (c) passivation of the EC electrode array intensifies when 50 mA/cm² current density was applied. On the

other hand, Figure 17 (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.



Figure 17. 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).

CHAPTER 5: CONCLUSION AND FUTURE WORK

This study investigated the performance of a novel dielectrophoretic force induced by a cylindrical interdigitated electrode array (EC-DEP) for harvesting marine microalgae (*Tetraselmis* sp.) in electrocoagulation process. The performance of the proposed electrode array was compared with a conventional flat parallel plate electrode array (EC). Through numerical study, the induction of the dielectrophoretic force was confirmed in the electrocoagulation process. During experimental analysis, the following conclusions were obtained:

- After 10 minutes of electrocoagulation, the EC-DEP electrode array showed 37.6% higher algal harvesting efficiency. This is because the DEP force induced by the EC-DEP electrode array will intensify the collision among microalgae and assist the Van der Waal's force in promoting coagulation.
- Application of 20 mA/cm² current density using the EC-DEP electrode array shows 83% higher algal harvesting efficiency compared to the EC electrode array. However, for the EC-DEP electrode array, the difference in algal harvesting efficiency is only 2% when the current density increased from 40 mA/cm² to 50 mA/cm². This is due to decrease in the pH value of the solution during electrocoagulation which reduces the formation rate of Al(OH)₃ at higher current densities.
- Reducing the distance between the electrode improves the algal harvesting efficiency due to reduced electrical resistance in the electrocoagulation reactor.
 At an inter-electrode spacing of 0.5 cm, electrocoagulation process with the EC-DEP electrode array results in 96.2% algal harvesting efficiency.

- Although the aluminum content in the harvested microalgae increases with increasing current density, for the EC electrode array, due to electrode passivation the aluminum content decreases by 28% when the current density increased from 40 mA/cm² to 50 mA/cm². Moreover, electrode surface scans showed that reduced electrode passivation happens in the proposed EC-DEP electrode array which helps to sustain higher algal harvesting efficiency at higher current densities.
- Energy analysis showed that, compared to the EC electrode array, using the EC-DEP electrode array resulted in lower specific energy consumption. This happens due to the higher algal harvesting efficiency achieved by EC-DEP electrode array.

The performance of the proposed electrode array proved the enhancement in the electrocoagulation process. Nevertheless, this was applied in harvesting marine microalgae herein, in this study.

A further investigation of the proposed configuration is to study its efficiency in the separation of other targeted particles such as pollutants in water and wastewater treatment application. Moreover, application of the proposed configuration can be further extended to a pilot scale in order to validate the enhancement. Additionally, A different set of parameters can be evaluated and analyzed to obtain the optimum performance of the electrode array. The effect of temperature can be studied to examine the algal coagulation and electrode oxidation in the medium in order to be correlated to the harvesting performance. Similarly, the role of pH can be further evaluated to control the electrode passivation and determine the desired ions emission from the electrode. Economic feasibility can be set to ensure the profitable operation conditions considering all aspects and parameters. Finally, Further examination and analysis can be performed to the produced biomass for the determination of its applicability and usability in some industrial sectors. For example, increased aluminum concentration in the biomass can lead to higher toxicity for food source application. Ultimately, comparative analysis between harvesting technologies is a promising future research potential.
- 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. *Environmental Science and Pollution Research*, 23(19), 19350-19361.
- Alkhatib, A. M., Hawari, A. H., Hafiz, M. A., & Benamor, A. (2020). A novel cylindrical electrode configuration for inducing dielectrophoretic forces during electrocoagulation. *Journal of Water Process Engineering*, 35, 101195. doi:https://doi.org/10.1016/j.jwpe.2020.101195
- Almukdad, A. I., Alfahel, R. A. F., & AlHawari, A. (2020). Effects of current, electrodes spacing and operational time on the removal of heavy metals from primary treated municipal wastewater using dielectrophoresis.
- Arain, M. S., Arain, S. A., Kazi, T. G., Afridi, H. I., Ali, J., Arain, S. S., . . . Mughal, M. A. (2015). Temperature controlled ionic liquid-based dispersive micro-extraction using two ligands, for determination of aluminium in scalp hair samples of Alzheimer's patients: A multivariate study. *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy*, 137, 877-885.
- Barros, A. I., Gonçalves, A. L., Simões, M., & Pires, J. C. M. (2015). Harvesting techniques applied to microalgae: A review. *Renewable and Sustainable Energy Reviews*, 41, 1489-1500. doi:https://doi.org/10.1016/j.rser.2014.09.037
- Barrut, B., Blancheton, J.-P., Muller-Feuga, A., René, F., Narváez, C., Champagne, J.-Y., & Grasmick, A. (2013). Separation efficiency of a vacuum gas lift for microalgae harvesting. *Bioresource technology*, *128*, 235-240.
- Batista, K. B., Padilha, R. P. L., Castro, T. O., Silva, C., Araújo, M., Leite, L. F. M., . .Lins, V. F. C. (2018). High-temperature, low-temperature and weathering

aging performance of lignin modified asphalt binders. *Industrial crops and products*, 111, 107-116.

- Berthon, J.-Y., Nachat-Kappes, R., Bey, M., Cadoret, J.-P., Renimel, I., & Filaire, E. (2017). Marine algae as attractive source to skin care. *Free radical research*, 51(6), 555-567.
- Bhattacharya, A., Mathur, M., Kumar, P., & Malik, A. (2019). Potential role of N-acetyl glucosamine in Aspergillus fumigatus-assisted Chlorella pyrenoidosa harvesting. *Biotechnology for biofuels*, 12(1), 1-17.
- Bilad, M. R., Vandamme, D., Foubert, I., Muylaert, K., & Vankelecom, I. F. J. (2012).
 Harvesting microalgal biomass using submerged microfiltration membranes. *Bioresource Technology*, *111*, 343-352.
 doi:https://doi.org/10.1016/j.biortech.2012.02.009
- Brennan, L., & Owende, P. (2010). Biofuels from microalgae—A review of technologies for production, processing, and extractions of biofuels and coproducts. *Renewable and Sustainable Energy Reviews*, 14(2), 557-577. doi:https://doi.org/10.1016/j.rser.2009.10.009
- Cerff, M., Morweiser, M., Dillschneider, R., Michel, A., Menzel, K., & Posten, C. (2012). Harvesting fresh water and marine algae by magnetic separation: screening of separation parameters and high gradient magnetic filtration. *Bioresource technology*, 118, 289-295.
- Chen, Z., Shao, S., He, Y., Luo, Q., Zheng, M., Zheng, M., . . . Wang, M. (2020). Nutrients removal from piggery wastewater coupled to lipid production by a newly isolated self-flocculating microalga Desmodesmus sp. PW1. *Bioresource Technology*, 302, 122806.

Cheng, Y.-L., Juang, Y.-C., Liao, G.-Y., Ho, S.-H., Yeh, K.-L., Chen, C.-Y., ... Lee,

D.-J. (2010). Dispersed ozone flotation of Chlorella vulgaris. *Bioresource Technology*, 101(23), 9092-9096.
doi:https://doi.org/10.1016/j.biortech.2010.07.016

- Cheng, Y.-L., Juang, Y.-C., Liao, G.-Y., Tsai, P.-W., Ho, S.-H., Yeh, K.-L., . . . Lee, D.-J. (2011). Harvesting of Scenedesmus obliquus FSP-3 using dispersed ozone flotation. *Bioresource Technology*, 102(1), 82-87. doi:https://doi.org/10.1016/j.biortech.2010.04.083
- Choi, O. K., Hendren, Z., Kim, G. D., Dong, D., & Lee, J. W. (2020). Influence of activated sludge derived-extracellular polymeric substance (ASD-EPS) as bioflocculation of microalgae for biofuel recovery. *Algal Research*, 45, 101736.
- Coward, T., Lee, J. G. M., & Caldwell, G. S. (2014). Harvesting microalgae by CTABaided foam flotation increases lipid recovery and improves fatty acid methyl ester characteristics. *biomass and bioenergy*, 67, 354-362.
- Das, P., Khan, S., Thaher, M., AbdulQuadir, M., Hoekman, S. K., & Al-Jabri, H. (2019). Effect of harvesting methods on the energy requirement of Tetraselmis sp. biomass production and biocrude yield and quality. *Bioresource technology*, 284, 9-15.
- Das, P., Thaher, M., Khan, S., AbdulQuadir, M., & Al-Jabri, H. (2019). The effect of culture salinity on the harvesting of microalgae biomass using pilot-scale Tangential-Flow-Filter membrane. *Bioresource Technology*, 122057. doi:https://doi.org/10.1016/j.biortech.2019.122057
- Dassey, A. J., & Theegala, C. S. (2013). Harvesting economics and strategies using centrifugation for cost effective separation of microalgae cells for biodiesel applications. *Bioresource technology*, 128, 241-245.

Dayarathne, H. N. P., Angove, M. J., Aryal, R., Abuel-Naga, H., & Mainali, B. (2021).

Removal of natural organic matter from source water: Review on coagulants, dual coagulation, alternative coagulants, and mechanisms. *Journal of Water Process Engineering*, *40*, 101820.

- Dineshbabu, G., Goswami, G., Kumar, R., Sinha, A., & Das, D. (2019). Microalgae– nutritious, sustainable aqua-and animal feed source. *Journal of Functional Foods*, 62, 103545.
- Doggaz, A., Attour, A., Mostefa, M. L. P., Tlili, M., & Lapicque, F. (2018). Iron removal from waters by electrocoagulation: Investigations of the various physicochemical phenomena involved. *Separation and Purification Technology*, 203, 217-225.
- 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. *Journal of membrane science*, 448, 256-261.
- Du, F., Hawari, A., Baune, M., & Thöming, J. (2009a). Dielectrophoretically intensified cross-flow membrane filtration. *Journal of Membrane Science*, 336(1-2), 71-78.
- Du, F., Hawari, A., Baune, M., & Thöming, J. (2009b). Dielectrophoretically intensified cross-flow membrane filtration. *Journal of Membrane Science*, 336(1), 71-78. doi:https://doi.org/10.1016/j.memsci.2009.03.010
- Fabregas, J., & Herrero, C. (1990). Vitamin content of four marine microalgae. Potential use as source of vitamins in nutrition. *Journal of Industrial Microbiology and Biotechnology*, 5(4), 259-263.
- Fayad, N., Yehya, T., Audonnet, F., & Vial, C. (2017). Harvesting of microalgae Chlorella vulgaris using electro-coagulation-flocculation in the batch mode. *Algal Research*, 25, 1-11. doi:https://doi.org/10.1016/j.algal.2017.03.015

- Fiedler, S., Shirley, S. G., Schnelle, T., & Fuhr, G. (1998). Dielectrophoretic sorting of particles and cells in a microsystem. *Analytical chemistry*, 70(9), 1909-1915.
- Fuhr, G., Arnold, W. M., Hagedorn, R., Müller, T., Benecke, W., Wagner, B., & Zimmermann, U. (1992). Levitation, holding, and rotation of cells within traps made by high-frequency fields. *Biochimica et Biophysica Acta (BBA)-Biomembranes*, 1108(2), 215-223.
- Gao, S., Yang, J., Tian, J., Ma, F., Tu, G., & Du, M. (2010). Electro-coagulation– flotation process for algae removal. *Journal of Hazardous Materials*, 177(1), 336-343. doi:https://doi.org/10.1016/j.jhazmat.2009.12.037
- Gejji, V., & Fernando, S. D. (2018). Harvesting microalgae using ionic polyelectrolytes in an aqueous-organic two-phase system: Screening of separation parameters using model algal particles. *Process Biochemistry*, 72, 188-197. doi:https://doi.org/10.1016/j.procbio.2018.06.010
- Ghernaout, D. (2019). Electrocoagulation and electrooxidation for disinfecting water: New breakthroughs and implied mechanisms. *Applied Engineering*, *3*, 125-133.
- Ghosh, D., Solanki, H., & Purkait, M. K. (2008). Removal of Fe(II) from tap water by electrocoagulation technique. *Journal of Hazardous Materials*, 155(1), 135-143. doi:https://doi.org/10.1016/j.jhazmat.2007.11.042
- Grima, E. M., Belarbi, E. H., Fernández, F. G. A., Medina, A. R., & Chisti, Y. (2003). Recovery of microalgal biomass and metabolites: process options and economics. *Biotechnology advances*, 20(7-8), 491-515.
- Hafiz, M. A., Hawari, A. H., & Altaee, A. (2019). A hybrid forward osmosis/reverse osmosis process for the supply of fertilizing solution from treated wastewater. *Journal of Water Process Engineering*, 32, 100975.
 doi:https://doi.org/10.1016/j.jwpe.2019.100975

- Hafiz, M. A., Hawari, A. H., Das, P., Khan, S., & Altaee, A. (2020). Comparison of dual stage ultrafiltration and hybrid ultrafiltration-forward osmosis process for harvesting microalgae (Tetraselmis sp.) biomass. *Chemical Engineering and Processing-Process Intensification, 157*, 108112.
- Hanotu, J., Bandulasena, H., & Zimmerman, W. B. (2012). Microflotation performance for algal separation. *Biotechnology and bioengineering*, 109, 1663-1673. doi:10.1002/bit.24449
- Hanotu, J., Bandulasena, H. C. H., & Zimmerman, W. B. (2012). Microflotation performance for algal separation. *Biotechnology and bioengineering*, 109(7), 1663-1673.
- 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. *Journal of Environmental Management*, 260, 110106. doi:https://doi.org/10.1016/j.jenvman.2020.110106
- Hawari, A. H., Du, F., Baune, M., & Thöming, J. (2015). A fouling suppression system in submerged membrane bioreactors using dielectrophoretic forces. *Journal of Environmental sciences*, 29, 139-145.
- Huang, Y., Wang, X. B., Tame, J. A., & Pethig, R. (1993). Electrokinetic behaviour of colloidal particles in travelling electric fields: studies using yeast cells. *Journal* of Physics D: Applied Physics, 26(9), 1528.
- 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.
- Kent, M., Welladsen, H. M., Mangott, A., & Li, Y. (2015). Nutritional evaluation of Australian microalgae as potential human health supplements. *PloS one, 10*(2),

- Kim, J., Ryu, B.-G., Kim, B.-K., Han, J.-I., & Yang, J.-W. (2012a). Continuous microalgae recovery using electrolysis with polarity exchange. *Bioresource Technology*, 111, 268-275.
- Kim, J., Ryu, B.-G., Kim, B.-K., Han, J.-I., & Yang, J.-W. (2012b). Continuous microalgae recovery using electrolysis with polarity exchange. *Bioresource technology*, 111, 268-275. doi:10.1016/j.biortech.2012.01.104
- Knuckey, R. M., Brown, M. R., Robert, R., & Frampton, D. M. F. (2006). Production of microalgal concentrates by flocculation and their assessment as aquaculture feeds. *Aquacultural Engineering*, 35(3), 300-313. doi:https://doi.org/10.1016/j.aquaeng.2006.04.001
- Koyande, A. K., Chew, K. W., Rambabu, K., Tao, Y., Chu, D.-T., & Show, P.-L. (2019). Microalgae: A potential alternative to health supplementation for humans. *Food Science and Human Wellness*, 8(1), 16-24. doi:https://doi.org/10.1016/j.fshw.2019.03.001
- Larronde-Larretche, M., & Jin, X. (2017). Microalgal biomass dewatering using forward osmosis membrane: Influence of microalgae species and carbohydrates composition. *Algal research*, 23, 12-19.
- Lee, A. K., Lewis, D. M., & Ashman, P. J. (2009). Microbial flocculation, a potentially low-cost harvesting technique for marine microalgae for the production of biodiesel. *Journal of Applied Phycology*, 21(5), 559-567.
- Lee, Y.-K. (1997). Commercial production of microalgae in the Asia-Pacific rim. Journal of Applied Phycology, 9(5), 403-411.
- Liu, J., Tao, Y., Wu, J., Zhu, Y., Gao, B., Tang, Y., . . . Zhang, Y. (2014). Effective flocculation of target microalgae with self-flocculating microalgae induced by

pH decrease. Bioresource technology, 167, 367-375.

- Luo, S., Wu, X., Jiang, H., Yu, M., Liu, Y., Min, A., ... Ruan, R. (2019). Edible fungiassisted harvesting system for efficient microalgae bio-flocculation. *Bioresource technology*, 282, 325-330.
- Matos, C. T., Santos, M., Nobre, B. P., & Gouveia, L. (2013). Nannochloropsis sp. biomass recovery by Electro-Coagulation for biodiesel and pigment production. *Bioresource Technology*, 134, 219-226. doi:https://doi.org/10.1016/j.biortech.2013.02.034
- Milledge, J., & Heaven, S. (2011). Disc Stack Centrifugation Separation and Cell Disruption of Microalgae: A Technical Note. *Environment and Natural Resources Research*, 1, 17-24. doi:10.5539/enrr.v1n1p17
- Misra, R., Guldhe, A., Singh, P., Rawat, I., & Bux, F. (2014). Electrochemical harvesting process for microalgae by using nonsacrificial carbon electrode: A sustainable approach for biodiesel production. *Chemical Engineering Journal*, 255, 327-333. doi:https://doi.org/10.1016/j.cej.2014.06.010
- Mkpuma, V. O., Moheimani, N. R., & Ennaceri, H. (2022). Microalgal dewatering with focus on filtration and antifouling strategies: A review. *Algal Research*, 61, 102588.
- Morgan, H., Holmes, D., & Green, N. G. (2003). 3D focusing of nanoparticles in microfluidic channels.
- Moshood, T. D., Nawanir, G., & Mahmud, F. (2021). Microalgae biofuels production:A systematic review on socioeconomic prospects of microalgae biofuels and policy implications. *Environmental Challenges*, *5*, 100207.
- Munshi, F. M., Church, J., McLean, R., Maier, N., Sadmani, A. H. M. A., Duranceau, S. J., & Lee, W. H. (2018). Dewatering algae using an aquaporin-based

polyethersulfone forward osmosis membrane. *Separation and Purification Technology*, 204, 154-161.

- Muradov, N., Taha, M., Miranda, A. F., Wrede, D., Kadali, K., Gujar, A., . . .
 Mouradov, A. (2015). Fungal-assisted algal flocculation: Application in wastewater treatment and biofuel production. *Biotechnology for biofuels*, *8*, 24. doi:10.1186/s13068-015-0210-6
- Najjar, Y. S. H., & Abu-Shamleh, A. (2020). Harvesting of microalgae by centrifugation for biodiesel production: A review. *Algal Research*, *51*, 102046.
- Nazari, M. T., Freitag, J. F., Cavanhi, V. A. F., & Colla, L. M. (2020). Microalgae harvesting by fungal-assisted bioflocculation. *Reviews in Environmental Science and Bio/Technology*, 19(2), 369-388.
- Nguyen, T.-T., Bui, X.-T., Ngo, H. H., Nguyen, K.-Q., Nguyen, H.-H., Némery, J., ... Varjani, S. (2021). Nutrient recovery and microalgae biomass production from urine by membrane photobioreactor at low biomass retention times. *Science of The Total Environment*, 785, 147423.
- Nguyen, T. D. P., Le, T. V. A., Show, P. L., Nguyen, T. T., Tran, M. H., Tran, T. N. T., & Lee, S. Y. (2019). Bioflocculation formation of microalgae-bacteria in enhancing microalgae harvesting and nutrient removal from wastewater effluent. *Bioresource technology*, 272, 34-39.
- Oh, H.-M., Lee, S. J., Park, M.-H., Kim, H.-S., Kim, H.-C., Yoon, J.-H., ... Yoon, B.-D. (2001). Harvesting of Chlorella vulgaris using a bioflocculant from Paenibacillus sp. AM49. *Biotechnology Letters*, 23(15), 1229-1234.
- Ometto, F., Pozza, C., Whitton, R., Smyth, B., Torres, A. G., Henderson, R. K., . . . Villa, R. (2014). The impacts of replacing air bubbles with microspheres for the clarification of algae from low cell-density culture. *Water research*, *53*, 168-

- Pahl, S. L., Lee, A. K., Kalaitzidis, T., Ashman, P. J., Sathe, S., & Lewis, D. M. (2013).
 Harvesting, Thickening and Dewatering Microalgae Biomass. In M. A.
 Borowitzka & N. R. Moheimani (Eds.), *Algae for Biofuels and Energy* (pp. 165-185). Dordrecht: Springer Netherlands.
- Pandey, A., Pathak, V. V., Kothari, R., Black, P. N., & Tyagi, V. V. (2019). Experimental studies on zeta potential of flocculants for harvesting of algae. *Journal of environmental management*, 231, 562-569.
- Panis, G., & Carreon, J. R. (2016). Commercial astaxanthin production derived by green alga Haematococcus pluvialis: A microalgae process model and a technoeconomic assessment all through production line. *Algal research*, 18, 175-190.
- Park, S.-J., & Seo, M.-K. (2011). Chapter 1 Intermolecular Force. In S.-J. Park & M.-K. Seo (Eds.), *Interface Science and Technology* (Vol. 18, pp. 1-57): Elsevier.
- Pulz, O., & Gross, W. (2004). Valuable products from biotechnology of microalgae. Applied microbiology and biotechnology, 65(6), 635-648.
- Raven, J. A., & Giordano, M. (2014). Algae. *Current Biology*, 24(13), R590-R595. doi:https://doi.org/10.1016/j.cub.2014.05.039
- Rios, S. D., Clavero, E., Salvadó, J., Farriol, X., & Torras, C. (2011). Dynamic Microfiltration in Microalgae Harvesting for Biodiesel Production. *Industrial & Engineering Chemistry Research*, 50(4), 2455-2460. doi:10.1021/ie101070q
- Rubio, J., Souza, M. L., & Smith, R. W. (2002). Overview of flotation as a wastewater treatment technique. *Minerals Engineering*, 15(3), 139-155. doi:https://doi.org/10.1016/S0892-6875(01)00216-3
- Salim, S., Vermuë, M. H., & Wijffels, R. H. (2012). Ratio between autoflocculating and target microalgae affects the energy-efficient harvesting by bio-

flocculation. Bioresource technology, 118, 49-55.

- Sathasivam, R., Radhakrishnan, R., Hashem, A., & Abd_Allah, E. F. (2019).
 Microalgae metabolites: A rich source for food and medicine. *Saudi Journal of Biological Sciences*, 26(4), 709-722.
 doi:https://doi.org/10.1016/j.sjbs.2017.11.003
- Shi, W., Zhu, L., Chen, Q., Lu, J., Pan, G., Hu, L., & Yi, Q. (2017). Synergy of flocculation and flotation for microalgae harvesting using aluminium electrolysis. *Bioresource Technology*, 233, 127-133. doi:https://doi.org/10.1016/j.biortech.2017.02.084
- Singh, G., & Patidar, S. K. (2018). Microalgae harvesting techniques: A review. Journal of Environmental Management, 217, 499-508. doi:https://doi.org/10.1016/j.jenvman.2018.04.010
- Sivasankar, P., Poongodi, S., Lobo, A. O., & Pugazhendhi, A. (2020). Characterization of a novel polymeric bioflocculant from marine actinobacterium Streptomyces sp. and its application in recovery of microalgae. *International Biodeterioration & Biodegradation*, 148, 104883.
- Spolaore, P., Joannis-Cassan, C., Duran, E., & Isambert, A. (2006). Commercial applications of microalgae. *Journal of Bioscience and Bioengineering*, 101(2), 87-96. doi:https://doi.org/10.1263/jbb.101.87
- Stolz, P. (2005). Manufacturing microalgae for skin care. Cosmetics Toiletries, 120, 99-106.
- Tredici, M. R., Bassi, N., Prussi, M., Biondi, N., Rodolfi, L., Zittelli, G. C., & Sampietro, G. (2015). Energy balance of algal biomass production in a 1-ha "Green Wall Panel" plant: How to produce algal biomass in a closed reactor achieving a high Net Energy Ratio. *Applied Energy*, 154, 1103-1111.

- 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. *Chemical Engineering Journal*, 174(1), 249-257.
- Uduman, N., Qi, Y., Danquah, M. K., Forde, G. M., & Hoadley, A. (2010). Dewatering of microalgal cultures: A major bottleneck to algae-based fuels. *Journal of Renewable and Sustainable Energy*, 2(1), 012701. doi:10.1063/1.3294480
- Ummalyma, S. B., Gnansounou, E., Sukumaran, R. K., Sindhu, R., Pandey, A., & Sahoo, D. (2017). Bioflocculation: An alternative strategy for harvesting of microalgae – An overview. *Bioresource Technology*, 242, 227-235. doi:https://doi.org/10.1016/j.biortech.2017.02.097
- Van Den Hende, S., Vervaeren, H., Desmet, S., & Boon, N. (2011). Bioflocculation of microalgae and bacteria combined with flue gas to improve sewage treatment. *New biotechnology*, 29(1), 23-31.
- 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. *Biotechnology and bioengineering*, 108. doi:10.1002/bit.23199
- Vandamme, D., Foubert, I., & Muylaert, K. (2013). Flocculation as a low-cost method for harvesting microalgae for bulk biomass production. *Trends in Biotechnology*, 31(4), 233-239. doi:https://doi.org/10.1016/j.tibtech.2012.12.005
- Vandamme, D., Pontes, S. C. V., Goiris, K., Foubert, I., Pinoy, L. J. J., & Muylaert, K. (2011). Evaluation of electro-coagulation–flocculation for harvesting marine and freshwater microalgae. *Biotechnology and bioengineering*, 108(10), 2320-

- Wan, C., Zhao, X.-Q., Guo, S.-L., Alam, M. A., & Bai, F.-W. (2013). Bioflocculant production from Solibacillus silvestris W01 and its application in cost-effective harvest of marine microalga Nannochloropsis oceanica by flocculation. *Bioresource technology*, 135, 207-212.
- Xie, S., Sun, S., Dai, S. Y., & Yuan, J. S. (2013). Efficient coagulation of microalgae in cultures with filamentous fungi. *Algal Research*, 2(1), 28-33.
- Xue, Y., Li, Y., Zou, X., Xu, K., Wen, H., Zhang, B., ... Gong, Y. (2019). Optimization of thermal pre-flocculation treatment for effective air flotation harvesting of microalgae. *Journal of Chemical Technology & Biotechnology*, 94(6), 1760-1769.
- Yu, C., Vykoukal, J., Vykoukal, D. M., Schwartz, J. A., Shi, L., & Gascoyne, P. R. C. (2005). A three-dimensional dielectrophoretic particle focusing channel for microcytometry applications. *Journal of Microelectromechanical Systems*, 14(3), 480-487.
- Zenouzi, A., Ghobadian, B., Hejazi, M. A., & Rahnemoon, P. (2013). Harvesting of microalgae Dunaliella salina using electroflocculation.
- Zhou, W., Min, M., Hu, B., Ma, X., Liu, Y., Wang, Q., ... Ruan, R. (2013). Filamentous fungi assisted bio-flocculation: a novel alternative technique for harvesting heterotrophic and autotrophic microalgal cells. *Separation and Purification Technology*, 107, 158-165.
- Çetin, B., & Li, D. (2011). Dielectrophoresis in microfluidics technology. *Electrophoresis*, 32(18), 2410-2427.