Chapter I
Introduction

1.1. Background

The biggest challenges facing society in the 21st century include food crises, environmental degradation, climate change mitigation, and energy scarcity (Cvetković *et al.*, 2022). One of the causes of environmental degradation is water pollution. Therefore, finding the right solution is crucial to minimize the impact on environmental degradation. The wastewater treatment paradigm has changed from contaminant reduction to nutrient recovery in various forms, such as bio-solids (Mainardis *et al.*, 2022). On the other hand, many parties are worried about the impact of carbon emissions resulting from fossil fuels, so many parties are trying to find reliable non-fossil energy substitutes (Khan, et al., 2022). Among today's renewable energy resources, biofuels from microalgae have gained the interest of researchers over the past few decades. Microalgae are promising biological resources for various applications, including food, feed, medicine, cosmetics, fuels, wastewater treatment, biofertilizers, and bioplastics (Figueiredo, Ferreira, Quelhas, *et al.*, 2022). As a biomass, microalgae can be beneficial in solving the energy needs issue. Furthermore, generating renewable hydrogen from waste water is a good option for producing clean energy and lowering greenhouse gas emissions (Cvetković *et al.*, 2022).

The use of microalgae is an efficient method for nutrient recovery, wastewater treatment, greenhouse gas reduction, and biomass production that can be used for the production of biofuels such as biodiesel, methane, bioethanol, and others (Rodríguez-Palacio *et al.*, 2022). Microalgae are recognized as viable feedstocks for producing large-scale biofuels (Khan, et al., 2022). Microalgae use photosynthesis during the cultivation process, converting solar energy into chemical energy (Saadaoui *et al.*, 2021). During the growth phase, energy absorbed from the sun combined with water, CO_2 gas, nitrogen, and phosphorus produces biomass (Christenson and Sims, 2011). In addition, microalgae cultivation also

serves to store abundant CO_2 from the atmosphere and reduce global warming because algae consume about 1.8 tons of CO_2 to produce 1 ton of biomass (Saadaoui *et al.*, 2021). Yen & Brune (Yen &; Brune, 2007) states that CO_2 absorption through microalgae is one or two times more effective when compared to land plants.

Due to their high content of nutrients and health-promoting compounds, microalgae have been evaluated as ingredients in bakery product formulations (Lafarga, Mayre, *et al.*, 2019), broccoli soup (Lafarga, Acién-Fernández, *et al.*, 2019), pasta (Rodríguez De Marco *et al.*, 2014), and other food ingredients. Although the number of food products containing microalgae launched into the market is increasing every year (Lafarga, 2019). Previous research stated that the strong color of microalgae (generally green) is not liked by consumers and the public does not like the pungent taste & smell when microalgae are added to food products (Chacón-Lee and González-Mariño, 2010). In addition, the low production capacity of microalgae currently causes microalgae not to be widely available in the market. People may still lack knowledge about microalgae as biofuel. Consumers rarely have access to scientific articles, and their knowledge of microalgae depends on internet searches that generally provide no scientific evidence. The public may still be uninformed about the benefits of microalgae for biodiesel production, aquaculture, and wastewater treatment.

The utilization of microalgae for various purposes is hindered by the challenge of creating reliable and economical systems for the large-scale cultivation and harvesting of microalgae. When microalgae are harvested, they will produce wastewater containing excess nitrogen and phosphorus. These compounds cause eutrophication of water bodies (rivers, lakes, and seas) and ecosystem damage if not effectively addressed (Xu *et al.*, 2020; Khan, Anjum, *et al.*, 2022). Although microalgal utilization is growing rapidly from laboratory to commercial pilot standards, the weakness lies in microalgal harvesting (Misra *et al.*, 2015). After harvesting microalgae, the resulting effluent will contain an excess amount of nitrogen and phosphorus. These substances induce eutrophication in aquatic ecosystems (Fasaei *et al.*, 2018). The main challenges of microalgae harvesting are

their small cell size, low biomass concentration, and electrostatic repulsion between cells (Barros *et al.*, 2015).

Recent advances in microalgal technology have resulted in many efficient harvesting techniques to improve microalgal harvesting. Techniques used to harvest microalgae include centrifugation, coagulation, ultrasonic, pH change, filtration, etc (Nguyen *et al.*, 2019). Currently, centrifugation and membrane filtration are the most commonly used techniques for harvesting large-scale microalgae cultures. Nevertheless, both centrifugation and membrane filtration procedures require complex maintenance. Therefore, alternative methods are required to simplify the maintenance procedure and lower costs associated with harvesting, while maintaining the harvesting speed and biomass quality (Guldhe *et al.*, 2016).

Electrocoagulation (EC) is a microalgae harvesting method that needs to be explored because of its ease of operation, fast harvesting, adaptability, environmentally friendly properties, and low footprint (Das, Sharma and Purkait, 2022). Utilizing EC can result in energy savings of up to 89% compared to using centrifugation alone (Fayad *et al.*, 2017). Electrocoagulation is a combination of water electrolysis and the coagulation process. When the electrode releases metal ions (e.g. Fe^{3+}), these ions will react with hydroxyl ions to create metal-hydroxides or polyhydroxides, such as $Fe(OH)_3$. These compounds act as coagulants throughout the coagulation process. Electric voltage, initially microalgae concentration, and electrolysis time are crucial factors in EC procedures (Lucakova *et al.*, 2021a; Meetiyagoda and Fujino, 2022a).

Most electrocoagulation studies use *Chlorella* as a harvested microalgae (Lal and Das, 2016; Rahmani *et al.*, 2017; Castellaños-Estupiñan *et al.*, 2018). In addition, there are several other freshwater and marine microalgae harvested using EC such as *Botryococcus* (Xu *et al.*, 2010), *Desmodesmus* (Montaño Saavedra *et al.*, 2019), *Microcystis* (Meetiyagoda and Fujino, 2022a), and *Nannochloropsis* (Matos et al., 2013). Even though Dunaliella salina remains a microalgae species that receives minimal attention (Liu *et al.*, 2017).

The EC system using rectangular-shaped Fe anodes has a harvesting efficiency of <85% with an operating time of 3 hours and a precipitation time of 4

hours (Mahmood, Hwan Kim and Park, 2021). The electrode used has an area of 24 cm2 and a concentration of D. salina of 0.5 g/L. Harvesting *D. salina* uses two rectangular electrodes with a surface area of 24 cm^2 and the best electrolysis time of 20 minutes(Maleki, Almassi and Nasirian, 2020). A maximum microalga harvesting efficiency of 98.06% was obtained using rectangular-shaped aluminum electrodes and a stirring speed of 222 rpm. Wastewater will produce excess nutrients like nitrogen and phosphorus shortly after harvesting microalgae. These compounds cause the eutrophication of water bodies (rivers, lakes, and seas) and ecosystem damage if not effectively addressed (Xu *et al.*, 2020; Khan, Anjum, *et al.*, 2022).

On the other hand, the electrolysis of water produces microbubbles (O2 and H2 gases). Microbubbles generated from water electrolysis are highly recommended for further investigation (Das, Sharma and Purkait, 2022). The combination of O$_2$ and H$_2$ gas is then called HHO gas, hydroxy gas, oxy-hydrogen, or Brown's gas (Subramanian and Ismail, 2018; Sudrajat *et al.*, 2020). An advantage of the EC method is its efficiency in harvesting microalgae and producing HHO gas, which can be utilized as fuel gas.

Currently, there is limited data available regarding the effectiveness of using spiral electrocoagulation (SEC) to harvest D. salina microalgae, as well as the examination of HHO gas generated during the process. Meetiyagoda and Fujino stated that it is very important to ensure that microalgae harvesting wastewater does not pollute water bodies (Meetiyagoda and Fujino, 2022a). The lack of knowledge about the basic aspects of microalgae among consumers in Indonesia, especially in young age groups, shows the need for efforts to increase understanding of the use of microalgae as raw materials for making fuel. We need to analyze consumer knowledge, awareness, acceptance, and willingness to pay regarding fuels derived from microalgae.

The study used spiral electrocoagulation with voltage variations, electrolysis time, and initial *D. salina* concentration, as well as the angle of the reactor, to optimize the efficiency of harvesting *Chlorella sp* and *D. salina*. Additionally, the study analyzed HHO gas, wastewater quality, and the knowledge,

awareness, acceptance, and willingness to pay of Generation Z consumers for microalgae fuels. We collected the data using an experimental laboratory and questionnaires.

1.2. Problem Formulation

The background description above identifies the key issues that the research needs to address. Current literature trends on HHO gas are still unclear, particularly related to renewable energy. The effectiveness of spiral electrocoagulation (SEC) in harvesting *Chlorella sp.* and analyzing dissolved hydrogen gas levels is a question, because optimization of this method is necessary to improve efficiency. In addition, the concentration of dissolved hydrogen and the efficiency rate of harvesting *D. Salina* using SEC with non-sacrified electrodes are also not fully understood. The balance between ammonium and ammonia in wastewater during the process of collecting microalgae using SEC needs to be evaluated because it affects the quality of wastewater nutrients. Another challenge is the efficiency of harvesting *D. Salina* at low concentrations as well as maintaining the quality of nutrients in the wastewater. In addition, the volume and efficiency of HHO gas production during the harvesting of *D. Salina* with SEC requires further study to find the optimal production method. Proper operating conditions for harvesting *D. Salina* and the nutrient content in wastewater are also issues that have not been fully resolved. Finally, the understanding, awareness, acceptance, and willingness of Generation Z to pay for fuel from microalgae still needs more research to find out their attitudes towards the use of microalgae-based energy.

1.3. Originality

The research with entitled Innovation of Spiral Electrocoagulation to Optimize Microalgae Harvesting and HHO Gas Production has never been conducted and has nothing in common with previous research in title, purpose, or method. The research with entitled Innovation of Spiral Electrocoagulation to Optimize Microalgae Harvesting and HHO Gas Production has never been conducted and has nothing in common with previous research in title, purpose, or

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method. Table 1. 1 shows previous studies that this dissertation refers to over the last five years.

Table 1. 1 List research sources from the past 5 years.

No	Reference	Method	Research results
1	(Krishnamoorthy *et al.*, 2021)	Experiments were conducted using *Chlorella vulgaris, Scenedesmus acuminatu, Desmodesmus subspicatus, Mixed microalgae, Chlorella sp., Tetraselmis sp., Scenedesmus quadricauda, Dunaliella salina, Phaeodactylum tricornutum, Nannochloropsis sp., Scenedesmus obliquus, Dunaliella viridis*, Fe electrodes, Al, Mg, Steel, Boron-doped diamond-Al, Al-IrO2/TiO2, Nickel, Carbon, Stainless steel, Al-Graphite. Power used 3 to 40 V, electrolysis	Electrocoagulation-flotation is the most effective harvesting technique for recovering microalgae regardless of size, shape, surface charge, etc. - Taking into account environmental, energy, and cost aspects, it is not always necessary to obtain a recovery efficiency of > 90%, as residual cells in the medium can support flocculation in further batches/cycles. - Increasing the conductivity of the medium and reducing the electrode distance can minimize power consumption. Electrochemical techniques are exclusively suitable for pigment extraction and biofuel production, as they have a positive effect on the amount of lipids and pigments present in microalgae.

No	Reference	Method	Research results
		time 1.32 min to 60 min, stirring speed 100 rpm.	
2	(Lucakova *et al.*, 2021a)	Experiments were conducted using *Chlorella vulgaris*, Fe electrode, EC process 28 minutes, stirring speed 400 rpm.	Reduced energy use by 80% compared to the centrifugation process, which is 3.6 kWh / kg in the centrifugation process to 0.745 kWh / kg using EC.
3	(Shamsaie and Hosseini, 2022)	The experiment was conducted using *Nannochloropsis oculata* electrodes Al and Fe, a stirring time of 15 minutes, and a stirring speed of 200 rpm.	The use of sacrificial and non-sacrificial electrodes from *Nannochloropsis oculata* causes significant changes in the lipid and its proteins
4	(Phiri *et al.*, 2021)	Experiments were carried out using *Chlorella vulgaris,* the electrodes used were Pb, Mg, Al, and Zn. Voltage 10 V, 20 V, 30 V, electrolysis time 20 minutes, and stirring speed 20 rpm.	From highest to lowest flocculation efficiency, the results are as follows: Cu, Zn, Mg, Al, and Pb at 10 V; Mg, Zn, Cu, Al, and Pb at 20 V; and Mg, Zn, Al, Cu, and Pb at 30 V. The proposed temperature increase has been influenced by decreases in electrical resistance and anodic corrosion,

No	Reference	Method	Research results
			between 1.7 C and 3.3 C, 5 C, and 8.9 C, and 10.5 C and 18.4 C respectively recorded at 10 V, 20 V, and 30 V. The waste recorded metal concentrations between 0.513 mg/L and 43.8 mg/L for Pb. 7.02 mg/L and 20.5 mg/L for Mg, 1.34 mg/L and 9.09 mg/L for Al, 0.079 mg/L and 0.089 mg/L for Zn, and 0.252 mg/L and 0.434 mg/L for Cu electrodes.
4	(Krishnamoorthy *et al.*, 2021)	Experiments were conducted using *Chlorella vulgaris, Scenedesmus acuminatu, Desmodesmus subspicatus, Mixed microalgae, Chlorella sp., Tetraselmis sp., Scenedesmus quadricauda, Dunaliella salina, Phaeodactylum tricornutum, Nannochloropsis sp., Scenedesmus*	Electrocoagulation-flotation is the most effective harvesting technique for recovering microalgae regardless of size, shape, surface charge, etc. - Taking into account environmental, energy, and cost aspects, it is not always necessary to obtain a recovery efficiency of > 90%, as residual cells in the medium can support flocculation in further batches/cycles. - Increasing the conductivity of the medium and reducing the electrode distance can minimize power consumption. Electrochemical

No	Reference	Method	Research results
		obliquus, Dunaliella viridis, Fe electrodes, Al, Mg, Steel, Boron-doped diamond-Al, Al-IrO2/TiO2, Nickel, Carbon, Stainless steel, Al-Graphite. Power used 3 to 40 V, electrolysis time 1.32 min to 60 min, stirring speed 100 rpm.	techniques are exclusively suitable for pigment extraction and biofuel production, as they have a positive effect on the amount of lipids and pigments present in microalgae.
5	(Visigalli *et al.*, 2021a)	The experiment was conducted using *Tetraselmis sp,* aluminum electrode, current density 20 - 50 mA/cm2, electrocoagulation time for 20 min, and stirring speed 200 rpm	The increase in algae harvesting efficiency using the EC-DEP module can be attributed to three main factors. First, the recurrence of high electric field intensities in the proposed EC-DEP electrode array results in additional aluminum production in the electrocoagulation process. Second, the EC-DEP electrode array intensifies collisions among microalgae and aids van der Waals forces in promoting coagulation. Third, reducing electrode passivation in the proposed EC-DEP electrode

No	Reference	Method	Research results
			array helps maintain higher algal harvesting efficiency at higher current densities.
6	(Mahmood, Hwan Kim and Park, 2021)	Experiments are conducted using *Nannochloropsis salina* uses Fe, Al, and Mg electrodes with dimensions of 25 cm2 (5 x 5 cm). Electric power ranges from 0.6 to 6.5 V. The electrolysis time is varied between 30, 60, 120, 180 minutes.	The application of EC Mg-air fuel cells has significant potential for harvesting marine microalgae

Previous research shows that electrocoagulation-flotation is effective for harvesting microalgae and can be used for pigment extraction and biofuel production (Krishnamoorthy *et al.*, 2021). Optimization of operating parameters for different types of microalgae carried out by Lucakova et al. (2021) showed that the use of electrocoagulation reduces energy consumption compared to centrifugation. Comparison of the effectiveness of different electrodes was carried out by Shamsaie & Hosseini (2022). The results showed that significant changes in microalgae lipids and proteins used victim and non-victim electrodes. The influence of temperature and electrical voltage shows that the increase in temperature affects anodic corrosion Phiri et al. (2021). The development of new technologies and integration with other systems, namely the EC-DEP module, can improve the efficiency of algae harvesting Visigalli et al. (2021). Mahmood et al. (2021) stated that electrocoagulation applications need to be tested on different types of microalgae.

However, more research is needed to understand the quality of the waste produced after the microalgae are harvested. Optimal operating parameters for different types of microalgae, including specific parameters such as electrolysis time and voltage, still need to be further researched to improve efficiency. The electrolysis of water produces microbubbles (O_2 and H_2 gases). Microbubbles generated from water electrolysis are highly recommended for further investigation (Das, Sharma and Purkait, 2022).

1.4. Purpose

The main purpose of this study is to evaluate and improve the efficiency of microalgae harvesting using spiral electrocoagulation (SEC) and analyzed HHO gas. The study used spiral electrocoagulation with voltage variations, electrolysis time, and initial *D. salina* concentration, as well as the angle of the reactor, to optimize the efficiency of harvesting *Chlorella sp* and *D. salina*. Additionally, I analize wastewater quality, and the knowledge, awareness, acceptance, and willingness to pay of Generation Z consumers for microalgae fuels. The specific objectives of this study are:

1. Provides reviews, assessments, and evaluations of the published literature on HHO gas, including the production and challenges of HHO gas.
2. Analyze the effectiveness of harvesting *Chlorella sp* microalgae by electrocoagulation using stainless stell &iron electrodes.
3. Analyze concentration of dissolved hydrogen gas produced from the harvesting process of *Dunaliella salina* using SEC and ORP concentrations including pH, harvesting efficiency due to voltage variations and harvesting time using EC with non-sacrificial electrodes.
4. Analyze the equilibrium of ammonium (NH_4^+) and ammonia (NH_3) during *Dunaliella salina* harvesting.
5. Analyze the harvesting efficiency of *D salina* at low concentrations and evaluation the nutrients in the wastewater produced.
6. Analyze the harvesting efficiency of *Dunaliella salina* using electrocoagulation with a helix electrode and having HHO gas as hydrogen fuel.

7. Optimize operating parameters, including initial *D. Salina* concentration, voltage, angle of reactor, and electrocoagulation time. The study is carried out using response surface methodology (RSM) to maximize the *D. Salina* harvesting process analysis of wastewater quality produced shortly after the harvesting process.

8. Analyze on consumer knowledge, awareness, acceptance, and willingness to pay regarding fuels derived from microalgae to inform marketing strategies and support the promotion of microalgae production as a potential fuel source.

1.5. Benefit

a. Scientific Benefits

Providing readers and students with new information and understanding that can serve as a reference for further study.

b. Benefits to Institutions

The results of this research can be an input for the microalgae industry

c. Practitioner Benefits

The community has valuable experience and provides additional knowledge in applying microalgae harvesting using spiral electrocoagulation.

1.6. Synthesis

Synthesis explains the relationships between chapters. The first chapter of this study explains the background of the research. Microalgae, microalgae cultivation, microalgae harvesting, characteristics and production of HHO gas, and consumer knowledge, awareness, acceptability, and willingness to pay regarding fuels generated from microalgae are reviewed in Chapter II. Chapter III analysis of the literature that has been published in scientific articles related to the production and challenges of using HHO gas. This study serves as the basis for understanding the production potential of HHO gas in the electrocoagulation process, which was tested in Chapter IV through the harvesting of *Chlorella sp* using stainless steel and iron electrodes, including optical density measurements and dissolved hydrogen gas.

Chapter V expands this study by evaluating the production of dissolved hydrogen gas from *Dunaliella salina* harvesting, including analysis of pH, ORP concentration, harvesting efficiency, as well as variations in voltage and harvest time. In Chapter VI, the quality of wastewater produced from the microalgae harvesting was discussed, namely the analysis of ammonium (NH_4^+) and ammonia (NH_3) equilibrium during the *Dunaliella salina* harvesting. Chapter VII discusses the evaluation of the efficiency of harvesting *Dunaliella salina* at low concentrations and the study of nutrients contained in the harvested wastewater. Chapter VIII analyzes the use of electrocoagulation with helical electrodes and

HHO gas as hydrogen fuels. To optimize the harvesting process using electrocoagulation spirals, Chapter IX uses a response surface methodology (RSM) to determine the best operating parameters, such as initial concentration, voltage, reactor angle, and electrocoagulation time, and analyze the quality of the wastewater produced. Finally, Chapter X completes this research by examining the knowledge, awareness, acceptance, and willingness to pay for fuels from microalgae, in order to formulate marketing strategies that support the development of alternative fuels based on microalgae.

1.7. Research diagram

This research focuses on microalgae harvesting and HHO gas production using spiral electrocoagulation (SEC). The causal relationship is shown on the fishbone diagram (Figure 1. 1) while the details of the variations of the study are shown in the Figure 1. 2.
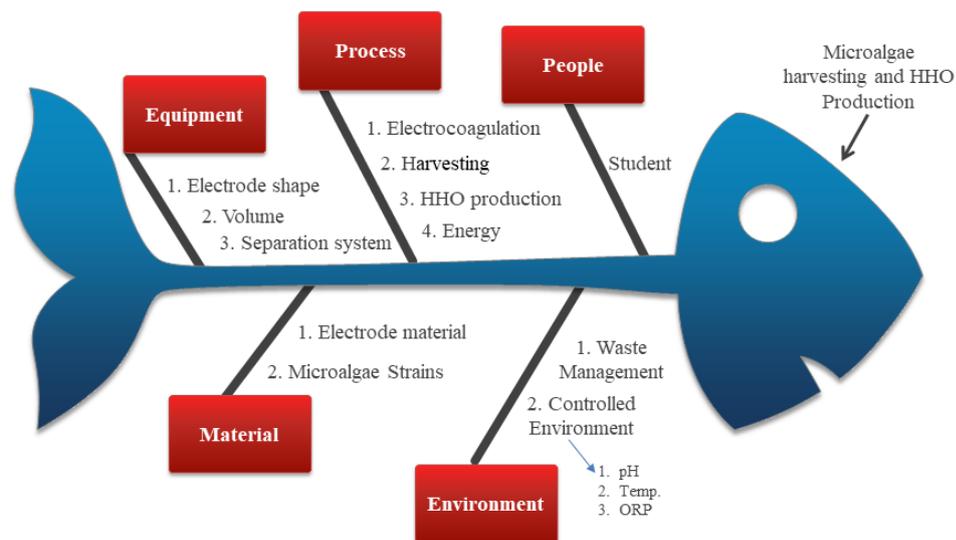

Figure 1. 1 Fishbone diagram showing the causal relationship in the process of microalgae harvesting and HHO production using spiral electrocoagulation

The aspects identified are people, process, equipment, materials, and environment of harvesting. The people aspect consists of the researcher himself who conducts the research. The process aspects include the electrocoagulation

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process, harvester efficiency, HHO production, and the energy required to harvest microalgae using SEC. The equipment aspect consists of the electrode shape, separation system, and reactor volume. The material aspect consists of electrode material and microalgae strains. Environmental aspects include waste management and control environment (pH, temperature, and ORP) during the microalgae harvesting process using SEC.
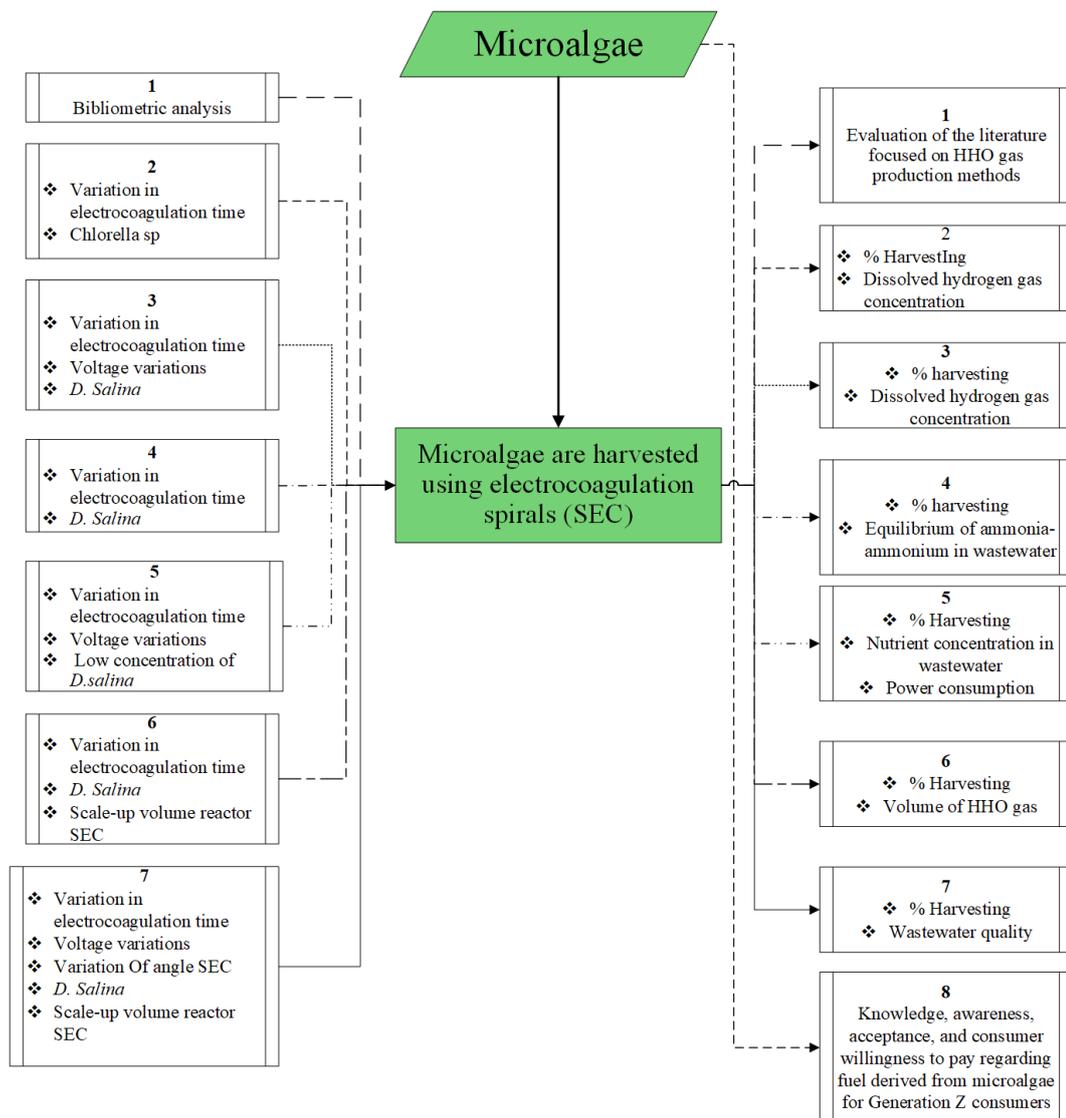

Figure 1. 2 Research variations and target parameters of the study