

A Biorefinery Concept for Energy Intensive Industries Focusing on Microalgae and Anaerobic Digestion

Katie E.H. Suominen, Suvi Ojanen and Pekka Ahtila

Department of Energy Technology, Aalto University, Aalto FI-00076, Finland

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Abstract: The biorefinery concept will be important to the energy industry as it allows a multi-process, multi-product biomass based industry. Continued increases in the prices of fossil fuels, the uncertainty of their availability and the environmental impacts of their extraction are favouring the implementation of sustainable energy production. This article provides a literature review of algal biomass utilisation, process utilisation, technological and economic factors when applying the biorefinery concept to energy intensive industries (whether retro-fitting or new buildings). This report focuses on opportunities in Finland for innovation, process integration and the development of supply chains whilst using flue gases as a feedstock for the microalgae. Currently, most research is on thermal combustion technologies. Microalgae provide an excellent opportunity to reduce carbon dioxide emissions by mitigation in such industries as pulp and paper. However, a beneficial driver would be feed-in tariffs or green trade certificates but are not necessary for the potential success within the industry. Reducing the overall economic costs with process integration and efficient technologies is beneficial for commercialisation of microalgae biorefineries. Microalgae biorefinery with a high efficiency could help improve the cost effectiveness of microalgae derived biofuels. The remaining algae after harvesting could be used for biogas production, which could be upgraded for vehicle fuel or the production of heat and power. An economically viable microalgae biorefinery with appropriate technologies and integrated for optimum efficiency is therefore possible.

Key words: Anaerobic digestion, biorefinery, energy efficiency, flue gas, microalgae.

1. Introduction

In recent times biorefinery technologies have been increasing in demand as there has been a growing interest in sustainable development, securing fuel and food supplies. A biorefinery integrates biomass conversion methods to produce heat, electricity, biomethane, biofuels, and high value products such as pharmaceuticals and omega 3 fatty acids [1-3]. The integration of energy intensive industries into biorefineries can bring both environmental and economic benefits such as mitigation of some GHG (greenhouse gases), a shift from fossil fuel to biofuels and even food security, if energy crops are not used.

There is an increasing demand for upgraded biogas, fuelled by an ever growing concern for the

environment, climate change and air quality issues especially in the urban environment. The European Union (EU27) is predicted to be responsible for 11% of the GHG emissions globally [4]. The EU saw a 10% drop in natural gas use during 2012 [4]. In order to replace all the transport fuels in Europe using biodiesel derived from microalgae, the area required would be 9.25 million hectares which would give a productivity of 40,000 l/ha-year [5].

Waste streams from industrial processes can be used as a feedstock to provide nutrients for the microalgae such as flue gases and waste water streams [6]. Flue gas contains NO_x (nitric oxides), SO_x (sulphur oxides) aromatics (dioxins, furans), alkenes, alkanes, particulates, fly ash and heavy metals [6]. The composition of flue gas and the species of microalgae selected are highly important as the flue gas could contain compounds that inhibit the

Corresponding author: Katie E.H. Suominen, M.Sc., LCIWM, research interests: biorefineries, biogas technologies, industrial energy efficiency. Email: kate.suominen@aalto.fi.

microalgae's production. The composition of the flue gas is dependent on the fuel combusted and can vary if fuel types are co-combusted [6].

Currently, microalgae options are being researched by both industry and academics due to having many advantages as a biomass product for energy production and other high value products [7]. It is capable of using seawater, brackish or freshwater from watercourses thus reducing the need for freshwater within industry and no arable land is necessary [7, 8].

The benefit of operating a microalgae photobioreactor on a large scale is their ability to cope with large amounts of biomass. This has fuelled the recent research into the feasibility of large scale microalgae photobioreactors integrated into biorefineries from new or existing plants [3]. The present assumption is that the barriers which are preventing the commercialisation of microalgal biorefineries will be removed by technical advances [9, 10]. Integrating a biorefinery into an energy intensive process can help the industry to open new product and profit streams. This is a very important issue currently as the pulp and paper industry is undergoing a period of economic uncertainty as part of the European recession [11].

In this review, it is assumed that pulp and paper mills use a lignocellulosic based feedstock and residues could be used in the biogas process. Historically, a lignocellulosic feedstock has been problematic as they have high retention times in the biogas reactor [12]. Using both primary and secondary sludge's from the pulp and paper industry Elliott and Mahmood [12] found that advances in preconditioning technologies may have decreased the residence time to 7 days in the biogas reactor [12] which would result in higher production times, efficiency and financial gains. Closed photobioreactor systems are also used as they are the most efficient as reducing contamination to achieve high value products [1].

The paper is organized as follows: Section 2 discusses the biorefinery feedstock options; Section 3

develops the importance of microalgal selection; Section 4 explains the technologies which could be applied for a microalgal biorefinery; Section 5 is explores the numerous by products which can be obtained from a microalgal biorefinery; Section 6 is an analysis of the technical feasibility of the system as a whole; Section 7 concludes this work and recommends further research.

2. Biorefinery Feedstock Options

For a biorefinery to be successful it must have the potential for a biomass source for the lifetime of the plant which can be between 20-40 years [13]. It is also important where possible for sustainability purposes that the feedstock does not interfere with the production of food.

Different combinations of the feedstock can be obtained from a variety of places without affecting agricultural lands. For example, they can be derived from lignocellulosic residues from pulp and paper, black liquor, tall oil or agricultural residues. They can also be acquired from waste sludge, waste oils, fish waste, industrial food waste (from supermarkets or food processing plants), animal wastes from agriculture, the organic fraction of municipal waste, fats from slaughter houses as well as meat processing plants [13].

There are many factors which affect the type of feedstock which can be selected for the process such as local climatic conditions, socio-economic (is it socially acceptable to use) and competition issues. For example, when using organic food wastes are there any other industries competing for the same waste supply, is that supply available all year or just seasonally. To avoid conflicts with food production dedicated food crops should be avoided where possible unless the crop has been spoiled and is not fit for human or animal consumption.

Energy intensive industries like pulp and paper mills can use waste pulp, residues from the pulp making process, tall oil, black liquor or even if

conditions are favourable virgin pulp. Having an available feedstock reduces economic costs of obtaining the feedstock. Additional feedstock variations can be purchased to improve the biogas fermentation process such as fish waste, industrial food wastes or the organic fraction of municipal wastes.

3. Microalgae Selection

Microalgae populate almost every ecosystem on Earth. They are microbial eukaryotes which are capable of adapting to a large variety of environmental conditions [7]. There are two types of algae, autotrophic and phototrophic. Autotrophic is used on large scale applications as it consumes carbon dioxide (CO₂) by the process of photosynthesis or chemosynthesis. Basically they are capable of producing their own food source [14]. Heterotrophic uses the CO₂ indirectly as it cannot fix CO₂ itself and uses organic carbon sources so it less efficient [14]. Microalgae show a huge variety for applications in industry, they can be used directly as a source of biomass for biochemical options such as biogas production/fermentation options, thermochemical options such as gasification and pyrolysis and due to their high oil content which can be extracted from the algae [14].

Microalgae are the largest taxa of plant life in the world [2]. Microalgae can also be produced on non-arable land and in many different water types; freshwater, brackish and saltwater (marine). The production of microalgae in such ways reduces the need to arable land and has no effect on food production.

It should be considered that microalgae can adapt to the local environment, if they are similar to their native conditions [2]. Fig. 1 shows the main stages of algae harvesting and drying.



Fig. 1 Generalised algae production flow diagram for microalgae production [15].

3.1 The Role of Microalgae

The advantages of microalgae based products are that production can be all year around (additional heating and lighting maybe required). Therefore, production of oil from microalgae is higher than that of rapeseed oil [1, 16] which only grows seasonally in summer in mid latitude areas such as Finland. Microalgae can be grown in water so require much less space thus do not have any impact on agricultural land [1, 17].

Microalgae are quick growing and many species have oil content between 20%-70% (dry weight biomass) [1, 3, 18]. Varying growth conditions can drastically increase the amount of oil produced by the microalgae [1, 19] which can be altered to favour the highest yield in large scale conditions. The lipids in microalgae have two times more energy when compared to carbon atoms in carbohydrates [2] which means they contain twice the amount of energy in comparison. Microalgae have the ability to obtain the nutrients needed for cultivation and production from wastewaters and flue gases from industrial processes [1].

The lipids stored within the microalgae are generally removed before the conversion of biomass to the end product [2] (before the biogas process). This allows for a higher profit margin especially if the next stage is to produce high value products. Due to the high value products being processed from the microalgae oil where possible it would be highly beneficial to use microalgae with high lipid contents [3]. Microalgal biomass has a heating value around 24 MJ/kg [3, 20] which means it is highly compatible for the production of biofuels, biogases and other products [3]. Table 1 gives examples of both freshwater and marine algal species and their chemical composition.

Table 1 Chemical composition of the dry percentage (%) matter for selected algae species [21].

| Algal species | Protein | Carbohydrate | Lipids |
|--------------------------------|---------|--------------|--------|
| Freshwater species | | | |
| <i>Scenedesmusobliquus</i> | 50-56 | 10-17 | 12-14 |
| <i>Scenedesmusquadricauda</i> | 47 | - | 1.9 |
| <i>Scenedesmusdimorphus</i> | 8-18 | 21-52 | 16-40 |
| <i>Chlamydomonsrheinhardii</i> | 48 | 17 | 21 |
| <i>Chlorella vulgaris</i> | 51-58 | 12-17 | 14-22 |
| <i>Chlorella pyrenoidosa</i> | 57 | 26 | 2 |
| <i>Spirogyra sp.</i> | 6-20 | 33-64 | 11-21 |
| <i>Euglena gracilis</i> | 39-61 | 14-18 | 14-20 |
| <i>Spirulinaplatensis</i> | 46-63 | 8-14 | 4-9 |
| <i>Spirulina maxima</i> | 60-71 | 13-16 | 6-7 |
| <i>Anabaena cylindrica</i> | 43-56 | 23-30 | 4-7 |
| Marine species | | | |
| <i>Dunalliellabioculata</i> | 49 | 4 | 8 |
| <i>Dunalliellasalina</i> | 57 | 32 | 6 |
| <i>Prymnesiumparvum</i> | 28-45 | 25-33 | 22-38 |
| <i>Tetraselmismaculata</i> | 52 | 15 | 3 |
| <i>Porphyridiumcruentum</i> | 28-39 | 40-57 | 9-14 |
| <i>Synechococcus sp.</i> | 63 | 15 | 11 |

Hulatt and Thomas' [22] study on microalgal photobioreactors in mid-temperate latitudes such as Southern Finland concluded that artificial heating was not required and that photobioreactors should be operated within the limits of the strain of microalgae selected for optimum energy efficiency.

4. Technologies for Microalgal Biomass Production

This review assumes that a closed photobioreactor is used for large scale application with an energy intensive industry converting into a biorefinery. After the oil has been extracted from the microalgae to produce high value products such as omega 3 fatty acids, the biomass is then added to an anaerobic digester. The energy intensive plant can then add another feedstock from other sources such as the organic fraction of municipal solid waste, fish wastes or other organic industrial wastes. Fig. 2 gives a simplified overview of the microalgae biorefinery suggested in this paper.

4.1 Closed Photobioreactor

Closed photobioreactor systems are the most commonly selected as the designs overcome many of

the technological issues with the open systems [1]. Closed systems are ideal for high value products such as pharmaceuticals as they remove contamination issues [1-3] and improve the productivity of the system [2]. It was also recommended that in mid-temperate ranges the gas-sparged photobioreactor should be used. A benefit of a gas-sparged reactor is that the flat plate reactors have a lower energy demand [6].

Table 2 contains a brief overview on the advantages and limitations of closed photobioreactors. As with all technologies there are some disadvantages which may be overcome or their efficiencies improved with innovations in the technologies themselves. For example, algal build up/growth on the walls maybe improved with better mixing techniques. A lot of research has been undertaken on tubular and flat plate reactors in the past as they are relatively cheap to construct. However, from this table closed photobioreactors look like a promising solution to some of the problems for large scale commercialisation as they are compact, good mixing capabilities with low energy requirements and are ideal for large scale industries. The issues with O₂ inhibition maybe overcome with some more research into bubbling or the amount of gas pumped into the system.

4.2 Anaerobic Digestion (Biochemical Conversion)

Biogas is produced when organic substrates are broken down by micro-organisms under anaerobic (no oxygen) conditions to produce a mixture of gases; mainly CO₂ and biomethane (CH₄). Organic substrates which are suitable as a feedstock are energy crops, manures, sewage sludge, industrial waste and the organic fraction of municipal solid waste. This paper covers the fractions from energy intensive waste such as the pulp and paper industry and the potential for fish waste or municipal solid waste. Fig. 3 shows the basic flow diagram for anaerobic digestion.

The process of anaerobic digestion is most suitable



Fig. 2 An overview of the suggested microalgae biorefinery for energy intensive industries.

Table 2 Advantages and limitations of closed photobioreactors [1, 2, 23].

| Type of photobioreactor | Advantages | Limitations |
|----------------------------|--|--|
| Tubular photobioreactor | Large surface area for illumination Relatively cheap Suitable for outdoor cultivation Good biomass production | Large area required Fouling Variations of pH, CO ₂ , oxygen (O ₂) in tubes Build up/growth on walls Heat generation sometimes |
| Flat plate photobioreactor | Easy sterilisation High biomass production Relatively cheap Easy to clean Good immobilisation for algae Suitable for outdoor cultivation Low O ₂ build up | Difficult temperature control Scale-up may require extra adaptations Build up/growth on walls Small hydrodynamic stress |
| Column photobioreactor | Easy sterilisation Compact Low energy consumption Good mixing Ideal for scale-up Good immobilisation for algae Reduced photo-inhibition and photo-oxidation | Column length may increase O ₂ inhibition |

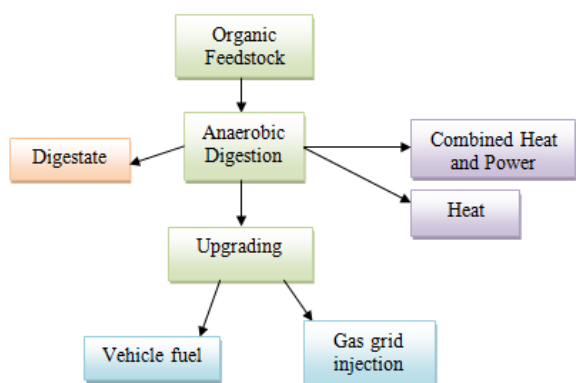


Fig. 3 Process diagram of anaerobic digestion.

for wet processes with a moisture content between 80%-90% [1]. There are two temperature ranges which are used on an industrial scale which are mesophilic (30 °C-40 °C) and thermophilic (45 °C-60 °C). Mesophilic is the most common process condition in industry as it is the most environmentally stable.

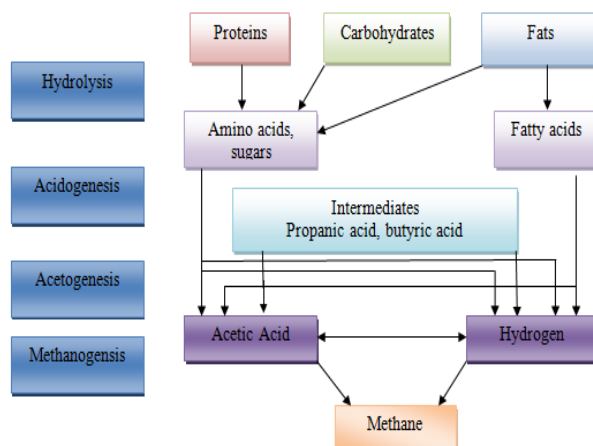


Fig. 4 Overview of the four main stages of biogas production including inputs and conversions.

There are four main stages in the biogas process: hydrolysis, acetogenesis, acidogenesis and methanogenesis which are shown in Fig. 4, along with the inputs to anaerobic digestions and how they are converted.

5. Biorefinery Products

The products from the biorefinery process are dependent on the feedstock and the processing methods selected. This paper will discuss based on using algal biomass and some other industrial or municipal waste and then the biogas process.

Table 3 shows a list of the products which can be obtained from a biorefinery. It can produce fuels for the transport sector such as biodiesel and biomethane. This will be an important source of energy with fossil fuels becoming more expensive and being finite. Biomethane after upgrading could be utilised by industry for an alternate combustion fuel to oil/coal and can be injected into the national gas grid or used for domestic purposes. The power can be sold back to the power grid resulting in more economic gains. The excess heat can be recycled and used elsewhere in the integrated biorefinery or even used to drive steam turbines to further produce power for use or sale [3, 9].

Table 3 The various products available from the biorefinery process for energy intensive industries [3, 9, 13].

Products from biorefineries

| |
|-------------------------------|
| Methanol |
| Ethanol |
| Other alcohols |
| Biogas |
| Bio-methane (after upgrading) |
| Biodiesel Biofuels |
| Omega 3 |
| Proteins for animal feed |
| Bulk sugars for fermentation |
| Antioxidant |
| Anti-tumoral |
| Antimicrobial |
| Anti-inflammatories |
| Adsorbents |
| Adhesives |
| Emulsifiers |
| Lignin based vanillin |
| Soil conditioner |
| Animal feed |
| Heat |
| Power |
| Electricity |

Biorefineries can produce biomaterials such as adhesives, adsorbents and emulsifiers which can all be sold to other industries as chemicals or if needed used by the biorefinery itself to reduce economic outgoings. Large scale biorefineries would probably be most interested in high value products like pharmaceuticals to maximise the amount of profit from the process, the microalgae oils could then be sold and the remaining biomass placed into the biogas process further producing more products for economic gains [3, 13].

6. Technical Feasibility

The technical feasibility of microalgal with regards to producing biofuels and other high value products has been proven economically viable [7]. It is important to note that Rawat et al. [2] stated that large scale cultivation of microalgae is already occurring in other areas than biofuels.

6.1 Application for Microalgae Technologies

In order to achieve economic feasibility the process should have a positive impact on energy supply and should not use more electricity for the process than the energy that is produced. It has been previously stated that a major benefit for microalgal biorefineries is that the sun is a free source of energy [1]. Though there are potentially some technical limitations for plant installations in Finland, which are dependent on the latitude of the plants. These factors could affect seasonal issues such as sunlight availability, day length and climatic conditions (temperatures from -30 °C to +25 °C) [1]. These factors can be overcome by the placement of photobioreactors inside temperature controlled glasshouses, also additional fluorescent lighting can be added if necessary to optimise algal production.

Selecting the species of microalgae and the type of photobioreactor is important, as the amount of microalgae that adhere to the walls of the reactor causing bio-fouling this decreases the efficiency of the systems and the amount of microalgae production [2].

This can be overcome by correct levels of mixing for the microalgae to prevent them to adhere but not too much as to inhibit them.

There are still some concerns with the extraction of the lipids from microalgae as many of the techniques used to extract the lipids are not viable with a large scale application [2]. However, so long as the end product is not for animal or human consumption the most effective method of lipid extraction for large scale purposes is solvent use [2] which can be applied to large scale installations.

Current research is suggesting that microalgal biorefineries are only economically viable if the by-products are high value such as omega 3 fatty acids [3], nutrients for pharmaceuticals [7], sugars for further fermentation for bioethanol and/or biomethane production [7]. Another highly promising factor for large scale implementation of biogas from microalgae after the extraction of high value products such as omega 3 fatty acids which would recycle nutrients into the biogas production to produce biomethane which can be sold as a by-product or used on site to reduce current fuel costs [7, 24].

6.2 Process Integration

The largest issues currently are the comparatively high costs of production of microalgae which may be overcome by integrating microalgal biorefineries into existing energy intensive industries where there could be some excess heat, electricity and other nutrients required for microalgae growth such as flue gases and wastewater streams. One of the most important issues to consider is the difficulty in scaling up the process and keeping the same efficiency as small scale production [2].

Process integration is very important to ensure that water is recycled and used efficiently where possible [11] as biorefineries use large amounts of water for the purification and separation processes. If the fresh water consumption can be reduced without the addition of extra technologies such as heat exchangers,

the economics are improved [11]. This can be achieved by using microalgae which does not require freshwater such as brackish or marine species.

According to Moshkelani et al. [11] when using the intensive energy optimisation approach the pulp and paper mills saves between 21%-34% of the overall steam consumption (savings for other technologies such as compressors) and 27% of the fresh water consumption of the plant. This provides information on the importance of process integration, how it can improve energy efficiency and reduce consumption from other sources (energy, water supplies).

6.3 CO₂ Mitigation and the Use of Flue Gases

Microalgal biorefineries with added biogas digestion are capable of CO₂ abatement [2]. The growth rate of microalgae is approximated to be around 50 times higher than that of a terrestrial plant [3, 25]. The photosynthetic efficiency of microalgae is higher when compared to terrestrial plants [3, 26]. This strengthens the use of microalgae as a biomass source as it can produce more biomass quicker than its terrestrial plant counterparts. This also suggests that microalgae absorb CO₂ quicker than terrestrial plants thus are more efficient to use for CO₂ mitigation.

Combining microalgae and flue gas is an innovative novel process to help the commercialisation of this technology [6]. Flue gas together with microalgae could intensify the removal of sulphur and carbon compounds from the waste streams by using calcium rich streams is just another way to improve CO₂ mitigation strategies [6]. Flue gases have not yet been used to their advantage not only to mitigate CO₂ but also to provide nutrients to microalgae, improve biogas production, remove sulphur and carbon compounds [6].

Using flue gas and closed photobioreactors have higher energy efficiencies when compared to using open pond systems [18]. In closed photobioreactor such as the airlift bioreactor a CO₂ removal efficiency of 82.3% can be obtained [3].

There are many sources of CO₂ within industry which if utilised can reduce economic outgoings and increase technical innovations for the improvement of microalgal biorefineries. Sources which can be used by industry to feed the microalgae are flue gas, exhausts from combustion processes or chemical options such as using carbonates [27, 28]. The integration of these processes increase the sustainability, the energy efficiency of the process, allows CO₂ mitigation, reduces costs for CO₂ sourcing and provides nutrients for the microalgae's cultivation. It has been previously researched that the use of flue gases does not impinge on the growth of the microalgae [3, 29] which means that the microalgae must utilise compounds within the flue gas without inhabitation at certain levels.

In the future, flue gas used in this way can help expand energy pathways such as production of hydrogen from microalgae, expansion of biogas from microalgae via anaerobic digestion and many other novel innovative ideas.

Wastewater could supply needed nutrients to the microalgae [15] which are rich in nitrogen, phosphorus and other micro-nutrients necessary for microalgal growth, this is another large advantage for large scale application of microalgal biorefinery technologies. It can also decrease the cost of cleaning/removing certain contaminants from the waste stream.

At large scale applications the use of artificial media is not profitable or viable, therefore the use of wastewater and flue gases (or other waste streams) would be highly recommended for use in the process where microalgae cultivation is occurring [2, 3]. The combination of wastewater treatment and other waste streams combined with anaerobic digestion of the microalgae after valuable lipids have been removed, improves the profitability of the plant considerably [2].

6.4 Biogas with Cellulosic Materials

Feedstock selection is very important as it is around 40-60% of the operational costs for microalgae

production between 2003 and 2005 [13]. According to Rawat et al. [2] harvesting and transportation costs are low in comparison to oil crop production.

With regards to the pulp and paper industry and biogas production, cellulosic materials are not easily fermented as their structure makes them difficult to break down [30]. Pretreatment is normally used to remove or breakdown lignin based materials [30]. Pretreatments can include physical pretreatments such as mechanical treatment which grinds or crushes the lignin [30]. Physio-chemical options like hydrothermal shock treatments, liquid hot water and steam explosions being the mostly popular to breakdown the lignin. There are also chemical pretreatments where acids/alkalises are added. Finally, there are biological pretreatments where fungi can be added to cause the lignin to rot [30]. The type of pretreatment selected is based on many things including the area the technology requires, cost, energy efficiency, process efficiency and maintenance required by that system.

Mshandete et al. [31] found that sisal pulp and fish waste could be co-digested to improved methane rates in the biogas process with 33% fish waste and 67% sisal pulp waste which gave an increased methane yield of between 59-54%. Yen and Brune [32] study discovered the ideal C/N ratio for the co-digestion of algal sludge and waste paper was around 20-25/1.

7. Conclusions

Technical improvements are still necessary and further research is still required to remove all issues. However, integrated systems for energy intensive industries either new or existing could be economically viable, if they are producing high value products and then recycling the microalgae into another process such as the biogas production which after upgrading the biomethane can be used for further economic gains. Overall, these conditions contribute positively to the economic feasibility of microalgal biorefineries when used together with energy intensive industries.

The use of microalgae is beneficial as it can use flue gas or other industrial processes as a source for growth mediums and also it can filter wastewaters for other nutrients reducing the costs of contaminant removal. The use of microalgae for CO₂ mitigation is beneficial in becoming carbon neutral and reducing emissions of CO₂. For large scale production it is with the utmost importance that the correct species of microalgae is selected as this will affect the economics of scale and the efficiency of the plant.

Small scale testing must first be undergone at the potential site to make sure that the most suitable microalgae has been selected for the environmental conditions and that the most efficient process integration systems have been selected to reduce energy losses and improve the economics of the system.

These processes have a positive effect on the economics of the system which suggest that Green Trading Certificates and Feed-in Tariffs would not be required at least at large scale where profit could be made without extra support. With that in mind some financial incentive for the commercialisation of microalgal biorefineries to encourage the uptake of these technologies by industries could be useful at least in the early stages.

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