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Microalgae for biofuels via thermochemical conversion processes: A review of cultivation, harvesting and drying processes, and the associated opportunities for integrated production

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ABSTRACT

Biofuels can be derived from waste biomass feedstocks, such as municipal, agricultural, forestry and industrial waste. There are several advantages in switching to microalgae for biofuel production. Microalgae has a rapid growth rate, so is more productive, so requires smaller areas for cultivation per unit of biomass produced. Microalgae can absorb "waste" CO₂, does not compete with food crops (for land and freshwater), and can be cultivated in wastewater, doubling as a wastewater treatment. This paper gives an overview of microalgae cultivation, focusing on the early energy-intensive stages: growth, harvesting and drying. The harvesting and drying steps constitute a significant economic bottleneck, due to their high energy costs. This review also covers microalgal cultivation and its integration with wastewater treatment, carbon and energy sources, and the utilization of microalgal biofuel co-products from thermochemical conversion, as this route is the most likely to mitigate the techno-economic downsides of microalgal biofuel production.

1. Introduction

Rising demand for energy, environmental concerns over the emission of greenhouse gases from fossil fuels, and the projected shortage of fossil fuels has led to the search for alternative sustainable sources of energy, including various biomass feedstocks (Singh and Cu 2010). Biomass is used worldwide as a feedstock for the production of heat, power, biofuel and value-added chemicals (Cheng, 2009). Key biomass sources include aquatic biomass, municipal waste, agricultural solid waste, forestry residue and industrial wastes (Basu, 2010).

The main source of global energy is oil (petroleum), at 31.8% of combined global energy. Currently, energy from waste and biofuel accounts for only 9.5% (IEA, 2020). Globally, bioenergy constitutes 53.2% of the total renewable energy supply, the remainder being mainly geothermal, hydro, solar, wind and tidal energy (OECD, 2020). Fossil fuels currently contribute much more than renewables, but they are finite, and the rate of fuel consumption is increasing due to increasing industrial activity and demand for liquid transportation fuels. The average prices for OPEC crude oil were 1.63 US\$ per barrel in 1960. Prices peaked at 109.5 US\$ per barrel in 2012, however, from 39.3 USD\$ per barrel in 2016, the most recent price of crude oil is \$40.1 per barrel

in 2020 (Sonichsen, 2020a). The price fluctuates due to political instability and security issues around the globe. This is a problem for us all, as the global energy economy is currently overwhelmingly dependent on these feedstocks (Schobert, 2013). However, global biofuel consumption has increased monotonically for the past eighteen years (EIA, 2020), and its supply is more evenly spread worldwide, and therefore less prone to such effects. While increasing crude oil prices and environmental concerns are the main reasons for the gradual move away from crude oil (Walker et al., 2019), biofuel production has been promoted by various governmental subsidies worldwide, which allows it to compete to some extent. Though these subsidies may be worthwhile in the long term, they can also be socially unacceptable by citizens (Reboredo et al., 2016).

A study of the impact of oil prices on bioenergy, emissions and land use showed that higher crude oil prices might lead to the production of more biofuel and lower greenhouse gas emissions because lower crude oil price with an increased endowment of resources from the oil and gas lead to more energy from biofuel (Winchester and Ledvina, 2017).

Biomass is a promising feedstock not only for the production of valuable fuels but also for value-added chemicals. The conversion can be achieved through thermochemical or biochemical conversion processes. Biomass can be broadly divided into land-based and aquatic biomass

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Received 23 January 2021; Received in revised form 21 February 2021; Accepted 21 February 2021 Available online 4 March 2021 2589-014X/© 2021 Elsevier Ltd. All rights reserved. (algae). The major constituents of land-based and aquatic biomass differ significantly. Most land-based biomass is lignocellulosic, so is comprised of cellulose, hemicellulose, and lignin. Aquatic biomass is mainly composed of lipids, carbohydrate, and protein (Yuan et al., 2015). This means that it could be the feedstock for a range of other products, beyond biofuels. Some species have high lipid or carbohydrate contents, which can be used for biofuel production by chemical (e.g. transesterification of oils to biodiesel) or biological routes (fermentation, for bioethanol).

Microalgae can grow in fresh, saline, brackish aqueous media and wastewater. Therefore, microalga could be used for the treatment of a variety of contaminated aquatic medium due to the heavy metal sequestration ability of microalgae with little nutritional input (Rai et al., 1981). Microalgae has a high growth rate: for example, microalgae can yield up to 10,000 gals per acre of biofuel per year, while corn typically yields only 60 gals per acre (Ullah et al., 2014). Therefore the transition from crop derived biofuels to microalgae derived biofuels would be worthy, as it would exert tremendously, reduction in land area requirement for cultivation. Microalgae strains can be controlled to regulate their compositions, to yield higher concentrations of compounds of interest via modulation of nutritional conditions and process variables (Miao and Wu, 2004).

The apparent prospective of microalgae as a source of renewable sustainable liquid derived fuel is a sturdy driver behind integrated conversion processes of microalgae. This provides the core rationale for substantial public support aimed towards the research and development of microalgae. This review article summarises and discusses: (i) the control of predatory contaminants in microalgae cultivation systems, and (ii) the modification of microalgae cultivation systems to enhance biomass productivity, and (iii) the lowest selling prices of liquid biofuel from chemical and thermochemical processes determined via technoeconomic analysis.

The work aims to give an overview of the first steps in microalgal biofuel production: cultivation, harvesting, drying. It also discusses integrated microalgae biomass production, as this is the envisaged route to overcoming many of the economic obstacles to microalgal biofuel production. This element is largely discussed in terms of predicted selling prices of liquid biofuels, determined via techno-economic analysis (TEA).

2. Algae

Algae are photosynthetic organisms that exist in most aquatic habitats and vary from unicellular forms (microalgae) to complex multicellular forms such as seaweeds (macroalgae).

The US algal collection, for example, contains some 300,000 samples including both natural and genetically modified organisms, although they are estimated to be more than a million (Smithsonian, 2020). One class of microalgae, the diatoms, make up the bulk of the World's phytoplankton, making it the largest portion of biomass on Earth (Gupta and Demirbas, 2010).

Algae is not characterized, and organisms falling known as "algae" are found throughout some quite distinct phyla (Fig. 1). Algae can be prokaryotes or eukaryotes. Prokaryotes are single-celled organisms with no true nucleus; instead, they have an imaginary nucleus known as a "nucleoid", while eukaryotes are organisms that have complex cell structures and possess a true nucleus. Plants, animals, fungi and some unicellular organisms also have a eukaryotic cell structure. Prokaryotic and eukaryotic algae are further classified into six kingdoms (Woese and Fox, 1977), as shown in Fig. 1. Algae are grouped into two classes; macroalgae and microalgae-based on number of cells (Fig. 2).

2.1. Macroalgae

Macroalgae are composed of multiple cells organized in structures resembling the leaves, stems and roots of higher plants, while microalgae are mostly unicellular microscopic photosynthetic organisms. Macroalgae are multicellular photosynthetic organisms with a rapid growth rate that yields a large amount of biomass (Bharathiraja et al., 2015). However, most macroalgae species are less than 5% lipid, on a dry weight basis (McDermid and Stuercke, 2003), so they are unlikely to provide a good source of biofuel via chemical route. However, macroalgae can be digested to produce bioethanol, due to its high carbohydrate content (Tan and Lee, 2014). An economic evaluation of the viability of macroalgae substrate demonstrated a payback time of 20 years (Morand et al., 2006), which is of value in terms of pollution reduction but is not currently viewed as an acceptable duration for return in investment (McKennedy and Sherlock, 2015). An alternative way of reducing the obstacles to macroalgae digestion routes is via



Fig. 1. Classification of Algae. Redrawn from Demazel (2008).



Fig. 2. (a) Macroalgae Laminaria digitate (Lambert, 2009) and (b) Microalgae Chlorella vulgaris (Lamouroux, 2018).

biorefining, by co-locating with an organic waste source. Karray et al. (2017) studied the anaerobic co-digestion of macroalga (*Ulva rigida*) with carbohydrate-containing wastewater generated by sugar industry for methane and biogas production (Karray et al., 2017). The anaerobic co-digestion of the green macroalgae with wastewater was carried out between an organic rate loading (ORL) of 0.08 to 2.6. The highest yield of methane and biogas was attained at ORL of 1.66 with a volatile solid reduction of 84.3%, and 75% maximum methane of total biogas. Hence, the co-digestion of macroalgae with carbohydrate-containing wastewater from sugar industry offers a promising alternative to the conventional anaerobic digestion, which is a more suitable way of utilizing macroalgae feedstock than thermochemical conversion processes.

2.2. Microalgae

Microalgae are the fastest-growing plant-like organisms in the world (Falkowski et al., 2004). They exist in varying ecological habitats including seawater, brackish water and fresh water and have also adapted to survive in various severe pH and temperature conditions. These features make microalgae the most abundant organisms on earth (John et al., 2011). The proportion of carbohydrates, proteins and lipids in microalgae varies with the species of microalgae and growth conditions. Fig. 3 shows the main constituents of microalgae and their components: Microalgae can contain up to 50% protein and has very low cellulose content (Schmid-Staiger, 2009). The lipids are mainly polar



Fig. 3. Components of typical microalgae. Modified from Schmid-Staiger (2009).

and neutral (Becker, 1994). For example, a *Nannochloropsis* species was reported to contain polar lipids (25%) and neutral lipid (15%), while *Chlorella vulgaris* contained lower polar lipids (0.7%) and higher neutral lipids (57.2%). The remaining lipids were unsaponifiable matters, chlorophyllides and other unknown components (Yao et al., 2015).

3. Microalgae cultivation systems

The conventional methods of microalgae cultivation are open raceway ponds and closed photobioreactors. This section gives an overview of the designs and cultivation processes used for microalgae production.

3.1. Raceway ponds

Microalgae cultivation using open raceway ponds is well-established, having been in operation since the 1950s (Brennan and Owende, 2010). Microalgae cultivation using raceway ponds are classified into two types, (1) artificial ponds and (2) natural water ponds (Brennan and Owende, 2010). Artificial raceway ponds can be categorized into three main design: (1) Raceway system; (2) Inclined or cascading system; and (3) Circular ponds with a central pivoted rotating system (Mata et al., 2010). Open raceway pond systems for microalgae cultivation are more capital-efficient systems than closed photobioreactors due to their lower capital costs. They are also more suitable for the removal of nutrients from domestic wastewater (Rawat et al., 2011). The most commonly used artificial ponds are raceway systems. The open raceway ponds are usually constructed with a paddle wheel to maintain the circulation of the algae broth and nutrients (Fig. 4a). A carbon source usually CO₂ is sparged from beneath the raceway (Chisti, 2007). The depth of water in open raceway ponds should not be more than 30 cm to allow adequate sunlight penetration (Abreu et al., 2012; Rawat et al., 2011). The cultivation medium is permanently fed into the raceway pond during the daytime before circulation, and the paddlewheel is operated throughout the cultivation period to prevent sedimentation. After the completion of the circulation loop, the microalgae broth is harvested behind the paddle wheel (Chisti, 2007). One of the disadvantages of the open raceway pond system is susceptibility to contamination.

Organisms such as amoebas, ciliates, protozoans, and rotifers prey on microalgae, particularly in the open pond system. Therefore, chemical, biological, physical and environmental methods are employed to control these predatory organisms (Zhu et al., 2020). Table 1 exhibits some examples of these methods of controlling predatory contamination in microalgae *cultivation* systems.

The open raceway system is also open to excessive loss of water via evaporation, which reduces the efficient utilization of CO_2 by microalgae than closed photobioreactor. Furthermore, contamination with other organisms in open raceway ponds also affects the yield of microalgae biomass. Therefore, some designs of the raceway ponds are covered in greenhouses to prevent pollution, rainfall and water loss (Chisti, 2007).

There is concern that the drive for the production of cost-competitive biofuels with conventional fossil fuels in the future may result in the conversion of pasturelands to lands for the cultivation of algae and other



Fig. 4. (a) Raceway pond, (b) Flat-plate photobioreactor, (c) Tubular photobioreactor, and (d) Standard bubble column PBR, (e) Airlift PBR with internal draft tube, and (f) Airlift PBR with external draft tube.

Methods of controlling predatory contamination in microalgae cultivation systems.

Control	Microalgae	Treatment	Microlagae System	Predatory contaminant	Key findings	Reference
Chemical	Chlorella pyrenoidosa	Dodecylbenzene sulfonate (SDBS)	1000 L open raceway pond.	Brachionus calyciflorus	Attained 0.74 g L^{-1} of microalgae biomass using 10 mg L^{-1} SDBS eliminating rotifiers in microalgae culture, without negative impact on microalgae biomass.	(Zhang et al., 2021)
	Chlorella kessleri	Sodium hypochlorite bleach	1 L glass tubes with a conical bottom.	Brachionus calyciflorus	Dosage between 0.45 and 0.6 mg Cl/L at two hour interval inhibited the predation while allowing the algae growth	(Park et al., 2016)
	Chlorella kessleri		1 L column reactor	Brachionus rotifer	The inhibition of the predation was achieved at copper concentration of 1.5 ppm allowed the algal growth	(Pradeep et al., 2015)
Biological	Nannochloropsis oculata	celangulin and toosendanin	Laboratory continuously cultured in series of flasks.	Brachionus plicatilis	Mixture of Celangulin: toosendanin (1:9) had no effect on microalgal growth but eliminated rotifiers.	(Zhang et al., 2020)
	Isochrysis, Nannochloropsis and Chlorella sp.	Celangulin, matrine and toosendanin	100-mL glass flasks with aeration tubes and covered with perforated polyethylene film.	Brachionus plicatilis	Mixture of Celangulin: toosendanin (1:9) reduces biocides levels and cost of rotifier extermination in microalgae cultivations system	(Huang et al., 2014b)
Physical	Nannochloropsis salina	Hydrodynamic cavitation (HC).	Microalgae grown in a10-L flat panel photobioreactor (PBR) for 7 days, underwent four passes of the HC, and then re- turned to the PBR.	Brachionus rotundiformis	87% removal of rotifiers after a single pass of HC (at1000 individuals/mL), and up to 99% after four passes, irrespective of the initial concentration.	(Kim et al., 2017)
	Chorella specie	Pulsed Electric Fields	Industrial 2.7 m ³ tubular photobioreactor.	Non-specified predators. Mainly rotifers, but also fungi, ciliates and bacteria	Exhibited 87% decrease in active protozoan population after 6 h treatment and 100% in few days of microalgae cultivation regime.	(Rego et al., 2015)
Environmental	Synechocystis sp.	рН	800 mL PBR continuous culture	Poterioochromonas sp.	Eliminate the presence of <i>Poter</i> . Sp at pH >11, with 33% increase in carbohydrate content and 33% decrease in protein content.	(Touloupakis et al., 2016)

terrestrial feedstock dedicated for biofuel production. This concern is largely due to the requirement for a large area of land to cultivate microalgae in a raceway pond on an industrial scale for biofuel purpose. A potential land competition between open-pond microalgae cultivation and terrestrial land dedicated to feedstock supply systems in the United States was studied (Langholtz et al., 2016). A scenario for the production of second-generation biofuel yielding $41.5\times10^9\,L\,yr^{-1}$ showed the most likely land base where both types of the feedstock may be deployed. The result of a spacial meta-analysis of that scenario showed that the potential competition for pastureland would be concentrated in 110 countries. Potentially, 40% of pasturelands could be converted to algal $(1.0 \times 106 \text{ ha})$ and terrestrial $(1.7 \times 106 \text{ ha})$ feedstock supply systems. However, it was concluded that the competition between algal and terrestrial biomass is not a constraint. Instead, from a policy point of view represents a synergy for the production of bioenergy. The synergy may develop both industries in parallel to boost the production of domestic renewable energy.

3.2. Photobioreactor (PBR)

Photobioreactors have higher biomass productivity than open raceway ponds due to their lack of dependence on climate, and low susceptibility to contamination by other predatory organisms (Singh and Sharma, 2012). Table 1 shows examples of successful elimination of predatory organisms in photobioreactors. Unlike the open raceway pond, photobioreactors are closed and can have higher biomass productivity because of the improved ability to control the culture variables (Chisti, 2007). Moreover, photobioreactors lose less CO₂ and water to the atmosphere and require less land (Brennan and Owende, 2010). However, as in the case of wastewater treatment using microalgae cultivation on a commercial scale, photobioreactors do not function well at large scale (Tan et al., 2018). In particular, it is no longer feasible at an operational volume of 50–100 L or higher to pass light efficiently and evenly into the photobioreactor (Chen, 1996). Another key obstacle to the take-up of photobioreactors is that they are more expensive than raceway ponds (Slade and Bauen, 2013).

The main design of photobioreactors is flat plate, tubular, and bubble column (vertical column or airlift):

(i) Flat plate photobioreactor

The flat plate photobioreactor is a transparent culture vessel made of glass, polycarbonate, plexiglass or polyethylene film, of thickness 5–6 mm to allow optimum light penetration and to enable the culture to achieve high cell density (Gupta et al., 2015; Show et al., 2017; Tan et al., 2018).

A diagram of a flat plate photobioreactor is shown in Fig. 4 (b). This type of flat plate is usually placed vertically or horizontally on the ground and achieves high energy density due to a high surface area for illumination (Lee, 2001; Tan et al., 2018). The flat plate photobioreactor system has higher photosynthetic efficiency and lower build-up of dissolved oxygen than tubular photobioreactors (Brennan and Owende, 2010). Water spray or internal heat exchangers are used to maintain the flat-plate PBR systems temperature, while agitation and aeration are achieved by rotation using a motor pump or by bubbling air from the base of each panel (Show et al., 2017). Rising bubbles mix the nutrients in the microalgae culture and prevent the accumulation of oxygen. Flat plate photobioreactors are employed for mass production of microalgae

both in indoors and outdoors cultivation techniques due to low accumulation of dissolved oxygen, and high illumination surface area in comparison to horizontal tubular photobioreactors. These reactors also have a modular design that is convenient to scale-up (Xu et al., 2009). The features of flat panel photobioreactors are important advantage for the mass production of photoautotrophic microorganisms and could be developed for the mass production of numerous algal species (Sierra et al., 2008). However, some of the setbacks of this types of system are the requirement for large landmass and the need for an elaborate setup of many units, damages caused by aeration, sterilization problems, fluctuation of temperature, and self-shading of microalgae cells (Gupta et al., 2015).

(ii) Tubular photobioreactor

Tubular photobioreactors are arrays of soft polyethylene, acrylic, plastic, glass, or highly transparent silicon rubber tubes that are vertical, horizontal, inclined or helix, and are directed towards the sunlight to achieve high solar conversion efficiency (Brennan and Owende, 2010; Elrayies, 2018).

To achieve high biomass productivity, their diameter should be below 0.1 m, to allow adequate penetration of light into the culture medium (Tan et al., 2018). The degassing column contains the microalgae medium culture, which is circulated into the solar arrays and back to the reservoir as depicted in Fig. 4 (c). Airlift or mechanical pumps are used to recycle gas to enhance the exchange of carbon dioxide and oxygen in the cultivated medium while mixing is in progress. The temperature in the degassing column is controlled by pumping cooled water. Tubular photobioreactor systems have reduced photosynthetic efficiency due to oxygen build-up, the build-up of pathogenic microorganisms in the inner walls, and high energy consumption, compared to a flat plate and bubble photobioreactors (Moreno-Garcia et al., 2017).

Tubular photobioreactor can be in vertical or horizontal positions, both of which are only suitable for photoautotrophic and photoheterotrophic (Chew et al., 2018). The vertical photobioreactors exhibit lower incident photon flux densities on the surface of the reactor, as result have higher areal production of biomass due to its higher light interception, in comparison to the horizontal tubular photobioreactors (de Vree et al., 2015).

(iii) Column photobioreactor

Column photobioreactors are usually cylindrical and have a height of up to 4 m and radii of up to 0.2 m (Show et al., 2017). These types of reactors are the most common, because of the absence of growth in the inner wall of the reactor, and their highly efficient use of CO_2 , sunlight, and land compared to open ponds. The column reactor is typically held in a vertical position, which allows the CO_2 or gas bubbles to rise rapidly and disperse at the surface of the reactor. This allows liquid to flow upwards at the central core of the reactors and downwards near the wall, approximating convection currents (Merchuk et al., 2007). Photobioreactors in vertical position also improve the dispersion of light over higher surface areas, enabling higher light intensity to reach the microalgae cell (Tan et al., 2018). This design improves gas-liquid exchange, and the mass transfer rate regulates the gas bubbles' residence time (Soman and Shastri, 2015).

Depending on the mode of aeration, column photobioreactors can be divided into a bubble (Fig. 4d) or airlift (Fig. 4e–f) column reactors. In the column photobioreactor systems, the CO_2 mass transfer and mixing are carried out by bubbling gas mixture into the reactor from a sparger (Moreno-Garcia et al., 2017). The standard bubble column photobioreactor does not have a circulation flow pattern at gas flow rates lower than 60 m/s due to lack of back mixing (Singh and Sharma, 2012), while the airlift photobioreactors have circulation draft tubes. Typically, the airlift column reactor operates base on the flow of gas via a sparger between two interconnecting regions. The gas is sparged beneath the reactor rises to the top of the liquid. The heavy bubbles at the top reach a disengagement zone and then undergo a downward movement, thereby leading to liquid motion in the reactor (Duan and Shi, 2014). The gas hold-up between the two zones as a result of the rise and downward movement of the sparged gas in the reactor considerably influence the fluid dynamics (Mohan et al., 2019).

3.3. Modified raceway and photobioreactors systems

Conventional raceway system and photobioreactor systems have been studied extensively in recent years. An overview of the raceway and photobioreactors systems is given in Sections 3.1 and 3.2 respectively. Table 2 highlights the influence of modification in the enhancement of biomass production in various types of raceway ponds and photobioreactors (PBR's) systems. In general, these modifications, however, improved microalgae biomass productivity via the following:

- i. Increasing bubble residence time
- ii. Improving fluid mixing
- iii. Enhancement of light intensity
- iv. Increasing dissolved CO2 concentrations
- v. Increase in intercepted solar radiation
- vi. Lowering volumetric power consumption
- vii. Design of compact-scale and low-cost laboratory-scale microalgae photobioreactor for rapid experiments.

The performance of conventional microalgae photobioreactor systems could be enhanced by the geometrical improvements of microalgae cultivations systems, addressing the impending challenges of processing parameters such a gas exchange, light, dissolved CO₂ concentration, power consumption, and high cost of production (Assunção and Malcata, 2020). Generally, the open raceway microalgae systems are associated with challenges such as (i) restricted choice of location due to climate conditions (ii) contamination issues, and (iii) requirements for large space. All of these associated challenges with conventional microalgae cultivation systems, largely, threaten the biotechnological process.

4. Microalgae modes of growth

The mode of growth of microalgae cultures significantly influences the growth dynamics and composition of microalgae (Cerón-García et al., 2013). The four major modes of microalgae growth based on energy and carbon supply are photoautotrophic, heterotrophic, photoheterotrophic and mixotrophic. Table 3 presents a summary of microalgae growth modes in terms of energy source, carbon source, light availability and metabolism variability.

Photoautotrophic growth of microalgae is the most common form of microalgae cultivation because most microalgae are photosynthetic (Perez-Garcia et al., 2011). In photoautotrophic metabolism, chemical energy is produced by carbon assimilation as CO_2 using sunlight.

$$nCO_2 + nH_2O = (CH_2O)_n + nO_2$$
(1)

The overall reaction for microalgae photosynthetic growth is shown in Eq. (1) (Klein et al., 2018). Photoautotrophic cultivation has an advantage of mitigating global CO_2 emission, as it only consumes CO_2 , and does not generate CO_2 like in heterotrophic and photoheterotrophic growth conditions. Mixotrophic growth nonetheless has possibilities of global CO_2 mitigation, as it has the characteristics of simultaneous utilization of carbon from both inorganic and organic sources.

Heterotrophic growth of microalgae typically uses organic carbon sources, such as glucose, acetate, or wastewater. This mode of microalgae growth is independent of solar energy and facilitates scale-up, as the design constraints around ensuring uniform illumination of the culture are removed, meaning that the reactor designs do not require a high surface to volume ratios (Brennan and Owende, 2010; Perez-Garcia

Influence of modification in the enhancement of biomass production in raceway ponds and photobioreactors (PBR's).

Cultivation system	Modification	Microalgae specie	Key findings	References
Raceway	Addition of flow deflectors and sloping baffles built in structures	Chlorella pyrenoidosa	The productivity of microalgae increased by 65% using the combination of the flow deflectors and sloping haffles, as compared to using flow deflectors only	(Huang et al., 2015)
	Bubble breakage in raceway ponds with up-down chute baffles	<i>Chlorella</i> mutant PY-ZU1	Microalgae biomass yield increased by 29% Decreased bubble generation time (27%) and increased residence time (27%)	(Cheng et al., 2016)
	Permutated conic baffles generate vortex flow field in a raceway pond	Spirulina	Increase in bubble residence time by 84.3% Microalgae biomass increased by 39.6%	(Cheng et al., 2018)
	CO ₂ bio-utilization with a liquid–liquid membrane contactor using hollow fibre membrane (HFM) and Fat sheet liner (FSL) polysolfone membrane	Nannochloropsis sp.	90% and 47% efficiencies of CO_2 bio-utilization Achieved using FSL and HFM	(Xu et al., 2019)
Flat plate	Comparing the effect of 3 types of special mixers in Flat plate PBR	C. pyrenoidosa	Mixers increased microalgal concentration between 20.9 and 42.9%.	(Huang et al., 2014a)
	Embedded bellow polymethyl methoavilete (DMMA)	Chloralla undaaria	The use of mixers significantly improved fluid mixing along light gradient.	(Sup at al
	tubes into a flat-plate PBR as light guides.	(FACHB-31)	Enhanced light intensity inside the reactor (2–6 times)	(Sull et al., 2016)
	Perforated inverted arc trough internals in a flat-plate photobioreactor	Chlorella vulgaris (FACHB-31)	Microalgae biomass production increased by 20.9% An increase in the dissolved CO_2 concentration in microalgae culture increased by 26.0%.	(Xia et al., 2018)
	Generation of microbubbles using jet-aerated tangential	Chlorella PY-ZU1	A significant decrease in gas bubble of 80.2%	(Cheng et al.,
Tubular	Internally illuminated lightening at an average irradiance of 1.15 W m ⁻²	D. tertiolecta	Blue lighting produced 1.7 times the microalgae content compared to the red lighting.	(Rebolledo- Oyarce et al., 2019)
	Fibonacci-type tubular photobioreactor	Spirulina	Enables 1.4-times increase in intercepted solar radiation in comparison to horizontal surface. Permits the achievement solar efficiency at outdoor conditions of up to 8.6%	(Díaz et al., 2019)
	Scale-up (1250 L) of a Fibonacci-Type Photobioreactor	Dunaliella salina	More solar radiation (60%) intercepted than the horizontal surface.	(Díaz et al., 2020)
D 111 C 1			Higher microalgae concentration of 0.96 gL ⁻¹ (3 times) than that in a raceway reactor under the same environmental conditions.	
Buddle Column	Flat plate splitting a bubble column PBR acting as an internal light guide and a heat exchanger.	(P12)	The maximum biomass productivity (0.75 g L ⁻ d ⁻) was attained at with a superficial gas velocity of 0.0044 m/s.	(Fernandes et al., 2014)
			column.	
	Real time light based CO_2 feeding strategy under diurnal simulated sunlight (LED)	Chlorella sp.	High microalgae biomass were achieved in 10 L Diurnal sunlight based for two stage (6.8 g L^{-1}) and single stage (9.0 g L^{-1})	(Naira et al., 2019)
	Self-rotating bubble-driven internal mixer in a bubble- column photobioreactor under natural sunlight and simulated sunlight	<i>Chlorella</i> sp. (FC2 IITG)	Microalgae biomass production of 7.5 g L^{-1} was achieved under natural sunlight. The productivity of microalcae under simulated	(Naira et al., 2020)
			sunlight was enhanced by 13%.	(0.1.1
	Split airlift photobioreactor to investigate gas holdup and bubble dynamics.	Scenedesmus sp.	from 1 to 3 cms ⁻¹ resulted in an increased in the optical density values, and dry biomass weight behaviour.	(Sabri et al., 2020)
Air lift (with internal draft tube)	Serial lantern-shaped draft tube in a gas-lift circumflux column PBR	Chlorella (PY-ZU1)	Decreased mixing time (21%), decreased bubble retention time (74%), and increased biomass yield (74%).	
~ 4	Influence of types of sparger and regime of fluid on microalgae production (i) glass with a porous glass surface, and stainless steel in the shape of (ii) a cross (four cylindrical elements), (iii) and a star (six cylindrical elements).	Chlorella vulgaris	The highest productivity of 58.7 mg $L^{-1} d^{-1}$ was established in 8 days culture at the lowest aeration rate using the star diffuser.	(Lopez- Hernandez et al., 2019)
Air lift (with external draft tube)	Hybrid of an external loop and a vertical isosceles triangle configuration with countercurrent gas flow mixing	Non-specified microalgae specie	Reduces volumetric power consumption, and increases liquid contact time and gas mass transfer.	(Pirouzi et al., 2014)
	Milliliter scale (170 mL working volume) external-loop- airlift bioreactors for cell growth studies.	Escherichia coli	Milliliter-scale bioreactors enable multiple laboratory scale testing of microbial growth conditions. Provides a low-cost approaches, expedite accessibility and reproducibility of the experiment.	

et al., 2011).

Heterotrophic microalgae cultivation as compared with photoautotrophic cultivation have the following advantages: (i) higher biomass productivity (growth rate) and higher lipid yield; (ii) low-cost, simple bio-reactor design and scalability; (iii) ability to control certain metabolic pathways to influence biomass composition by altering the culture medium's organic substrate; and (v) potential use in wastewater treatment (Chen, 1996; Miao and Wu, 2006; Perez-Garcia et al., 2011; Perez-Garcia and Bashan, 2015).

The disadvantages of heterotrophic microalgae are:

 (i) limited amount of capable heterotrophic species. For example, Chlorella vulgaris, Chlorella protothecoides, Crypthecodinium cohnii, and Schizochytrium limacinum grown under heterotrophic

Growth modes of microalgae cultivation.

	-			
Growth mode	Energy source	Carbon source	Light required	Metabolism variability
Photoautotrophic	Light	Inorganic	Yes	No switches between sources
Heterotrophic	Organic	Organic	No	Switch between sources
Photoheterotrophic	Light	Organic	Yes	Switch between sources
Mixotrophic	Light and organic	Inorganic and organic	No, but can be used	Simultaneous utilization

Modified Perez-Garcia and Bashan (2015).

conditions are the limited microalgae species identified with high lipid contents (Medipally et al., 2015), of which *Chlorella Species* contain lipid contents between 4.8 and 60% on a dry weight basis (Carone et al., 2019; Liu et al., 2014).

- (ii) utilization of plant-based derived glucose as a substrate that could facilitate to food versus fuel crisis (Perez-Garcia et al., 2011).
- (iii) introduction of contaminants in microalgae culture due to the presence of organic substrate (Chen, 1996).

Microalgae produced via the heterotrophic mode of cultivation are mostly in biorefining due to high lipid contents. Heterotrophic microalgae are also used on an industrial scale as a supplement for food and health promotion due to presence of omega-3-fatty acid (Carone et al., 2019; Chisti, 2007; Mata et al., 2010). For example, *Crypthecodinium cohnii* is used as a feedstock for the production of docosahexaenoic acid (Omega-3-fatty acid) in the industry (Ratledge et al., 2001). *Schizochytrium limacinum* is reported to have enriched the omega-fatty acid contents of eggs in laying-hens when used as an alternative to fish oil in a feed-mixtures (Kralik et al., 2020). Commercially, *marine thraustochytrids* are grown exclusively under heterotrophic conditions, but this is for the production of polyunsaturated fatty acids (Hu et al., 2018), which are of much higher value than biofuels.

Due to the need for an organic carbon source, the heterotrophic growth of microalgae may use more energy than photoautotrophic growth (Tan et al., 2018). The input energy to adenosine triphosphate (ATP) ratio for heterotrophic microalgae is 18%, which is higher in comparison with the 10% for photoautotrophic microalgae. Therefore, the cultivation of heterotrophic microalgae could be more advantageous than photoautotrophic from an economic outlook (Yang et al., 2000).

Heterotrophic microalgal growth seems like a viable alternative for microalgal cultivation in photobioreactors, especially when the organic carbon sources in culture media replace the photoautotrophic growth fixation of atmospheric CO₂ (Perez-Garcia et al., 2011). However, heterotrophic microalgal growth generates CO₂, and therefore does not mitigate CO₂ global emission (Lowrey et al., 2015).

Certain algae can grow in the dark by using organic carbon in the form of sugar or starch. Some microalgae combine both modes of growth (autotrophic and heterotrophic): this is "mixotrophic" growth.

Mixotrophic cultivation of microalgae uses a mixture of both carbon and energy sources to reproduce their cells. Hence, mixotrophic microalgae cultivation mainly utilizes a combination of both photoautotrophic and heterotrophic condition (uses both organic and inorganic carbon sources) for their growth. Examples of microalgae species that are capable of growing under mixotrophic conditions are *Tetraselmis*, *Neochloris* and some *Chlorella* species such as *Chlorella* vulgaris, *Chlorella protothecoides*, *Chlorella* sorokiniana, and *Chlorella* volgaris, *Chlorella* et al., 2021). The disadvantages of mixotrophic cultivation are: (i) the requirement of O₂, light, CO₂, and organic carbon, (ii) lower energy conversion efficiency than heterotrophic and autotrophic growth, and (iii) the net release of carbon dioxide than heterotrophic cultivation. However, CO_2 released in anaerobic respiration in mixotrophic cultivation could be trapped and utilized for photosynthesis process under mixotrophic cultivation (Mohan et al., 2014).

4.1. Growth dynamics of microalgae

Axenic culture of microalgae is a culture that grows in the absence of other species. It is characterized by five dynamic growth phases (Coutteau, 1996) as shown in Fig. 5. The lag phase, also called the induction phase, is the phase during which there is little increase in the microalgae cell density. The lag phase in microalgae culture growth is associated with the physiological adaptation of the microalgae cell metabolism, often including an increase in the level of metabolites and enzymes in microalgae cell division and carbon fixation.

The economics of downstream processing of microalgae are much more favourable if high-density cultures can be used, as the size, and therefore capital costs, of equipment are reduced. Environmental conditions like carbon dioxide, light, temperature salinity, screenings of microalgal strain, mixing affect the biomass productivity of microalgae species.

Microalgae can be cultivated in batch, semi-continuous or continuous reactors. In batch cultures, microalgae cells and nutrients are inoculated into the reactor at the beginning, followed by the microalgae growth and finally the harvest when the microalga population has attained a maximum density or near-maximum density. In the second phase, there is an exponential increase in cell density, which is described by Eq. (2) (Coutteau, 1996). The specific growth rate of microalgae depends largely on the species, temperature and light intensity.

$$C_t = C_0 e^{mt} \tag{2}$$

where, C_t - the cell concentration at time t, C_o - the cell concentration at time 0 and, m- specific growth rate.

In the phase of declining growth rate (third phase), there is a reduction in the rate of cell division due to the limitation of growth rate by physical and chemical factors such as light, carbon dioxide, nutrients and pH. The 4th phase, the "stationary phase", is characterized by the maintenance of constant cell density; i.e., there is a balance between the growth rate and the limiting factor(s). In the last phase of microalgae culture growth (decline phase), sustainable growth of microalgae culture is not favourable due to depletion of nutrient, deterioration of water quality, pH disturbance and overheating. Therefore, the cell density decreases drastically and subsequently the microalgae cell culture collapses (Coutteau, 1996).

4.2. Cultivation methods of microalgae

One of the key factors in microalgae productivity is the choice of the cultivation method (Novoveská et al., 2016a). Microalgae cultivation can be carried out via batch, semi-continuous or continuous methods. In the batch culture of microalgae, the microalgae culture is fed continuously with nutrient, the culture then grows for several days and is then harvested when the microalgae attain maximum density, or close to it.

However, in the semi-continuous method, the culture medium is regularly discharged, and the remaining culture is used as the initial charge for the next cultivation. This method of cultivation can be carried out indoors or outdoors and yields more algae for a given tank size than in batch. However, the duration of microalgae growth using this method is unpredictable. Eventually, the microalgae culture for semi-continuous method becomes unsuitable for further use due to the build-up of competitors and predators (Coutteau, 1996).

In continuous culture, a near-maximum growth rate of the microalgae culture is maintained by a continuous supply of nutrients into the growth chamber, and at the same time, the excess culture is removed. The drawbacks of continuous cultivation of microalgae are (i) relatively high cost, (ii) complexity requirement for steady, uniform illumination



Age of culture

Fig. 5. Dynamic phases of microalgae culture growth. Modified from Coutteau (1996).

and temperature, which has so far restricted continuous cultivation of microalgae to indoor, and small-scale (Coutteau, 1996).

Microalgae productivity at high cell densities reduces the use of energy in the upstream harvesting stage, and also in the downstream stage, especially when high lipid contents are achieved and desired. High cell densities and biomass productivities have been reported in both fed-batch, semi-continuous and continuous microalgae cultivation (Remmers et al., 2017; Renato Coelho et al., 2014). A comparison of an axenic culture of Chlorella sp. in fed-batch and continuous cultivation methods yielded biomass productivities of 6.9 and 9.1 gL⁻¹d⁻¹ respectively. The lipid productivities of the 1.5 $gL^{-l}d^{-1}$ were attained for the batch against 1.6 gL^{-l}d⁻¹ for the continuous method (Renato Coelho et al., 2014). A quantitative comparison of microalgae (A. obliquus) lipid production in batch against continuous cultivation under nitrogen starvation yielded about double lipid yields with the batch in comparison with the continuous cultivation (Remmers et al., 2017). While microalgae batch cultivation may seem advantageous, continuous cultivation decreases stoppage for cleaning, sterilization and setup.

4.3. Harvesting microalgae

Harvesting microalgae is the process of separating microalgae from an aqueous suspension of microalgae culture media. Open pond and closed PBR systems both deliver dilute algal solution. The open pond which contains dry matter varying from 0.05–0.75%, while the closed PBR contains dry matter ranging between 0.3 and 0.4% (Fasaei et al., 2018). Hydrothermal process, a water-tolerant thermochemical conversion process requires around 95% moisture content, as higher moisture content would lead to an energy-negative mode due to heat losses and pumping for water recirculation (López Barreiro et al., 2013). Thermochemical conversion processes of microalgae via torrefaction, pyrolysis, gasification and combustion can contain up to 10% of moisture content (Chang et al., 2015; Ho et al., 2020; Sanchez-Silva et al., 2013). Microalgae culture media contain a high amount of water, particularly for the non-tolerant thermochemical conversion process, which must be removed. The recovery and processing of microalgae culture media to microalgae biomass is an essential component in microalgae production. The cost and energy required for harvesting is a significant concern (Tan et al., 2018). At the industrial scale, the harvest constitutes up to 30% of the overall cost of production (Brennan and Owende, 2010; Rawat et al., 2011). However, to improve the economic viability of the microalgae production, the cultivation of microalgae in wastewater as a culture medium and strategy of biorefinery based

production may reduce the overall cost of production (Barros et al., 2015).

The fundamental properties that influence the recovery of microalgae biomass during harvest are particle size (2–20 µm), morphology (spheres, chains, rods or filamentous), specific weight and charge (usually negative) (Pahl et al., 2013). However, other factors such as medium composition, salinity, and hydrophobicity also influence the recovery of microalgae biomass (Barrut et al., 2013). These properties vary as a function of microalgae species, growth conditions and duration of the culture. The nature of the end-product determines the selection of the appropriate harvesting method. The harvesting of microalgae is carried out via two-step or single-step concentration. The methods could be mechanical, chemical or biological, or a combination of two or more of these methods. The most reliable among these methods is the mechanical based centrifugation and are therefore commonly used for microalgae harvest, often followed by biological or chemical flocculation/coagulation thickening to reduce maintenance and operation cost. Mechanical based centrifugation utilizes centrifugal force for separation base on density differences (Christenson and Sims, 2011). In particular, the nozzle type disc centrifuges are simple to clean and sterilize and can be used for all types of microalgae. However, operational cost and high investment must be taken into consideration (Shelef et al., 1984). Microalgae harvesting methods include coagulation/flocculation, sedimentation, flotation, electrical-based processes, filtration and centrifugation. A more detailed analysis of microalgae harvest methods can be found in the literature (Singh and Patidar, 2018).

4.4. Drying

The perishable nature of microalgae makes it necessary to dry microalgae after harvest to prevent spoilage (Klein et al., 2018). After microalgae are harvested, the dewatered microalgae slurry is dried for end-use, stability, extraction and further processing (Show et al., 2013). The drying parameters affect the chemical properties of the biomass (Sahoo et al., 2017). For example, the higher water content in microalgae biomass reduces the efficiency of biodiesel processing, reducing the yield (Tan et al., 2018). Hence, refined raw materials need to be used (Atadashi et al., 2012).

However, the production of biofuel such as biodiesel from dried microalgae (via a dry route) requires a high amount of energy (Klein et al., 2018). As the lipid has to be extracted and then trans-esterified, thereby altering the sustainability of the biofuel production process (Xu et al., 2011). The presence of water as small as 0.1% in lipid might

lower the yield of methyl esters when transesterified (Atadashi et al., 2012). An alternative is biodiesel production from wet biomass (wet route) via acid-catalyzed in-situ transesterification of highly saturated microalgae (80% moisture) using a small amount of HCL, effectively resulted in the conversion of over 90% wt% of lipid (Kim et al., 2015).

The moisture content of bio-oil derived from pyrolysis of biomass depends on the moisture content of its original biomass and the formation of water during pyrolysis, which depends on the process conditions. Pyrolysis of microalgae with high moisture contents could yield bio-oil with high water contents, high concentrations of oxygenated compounds, and low viscosity, all of which can contribute to instability during storage (Lehto et al., 2013). These bio-oil applications (Xiu and Shahbazi, 2012).

For example, the water content of petroleum fuel is regulated to avoid phase separation, which can lead to corrosion, the problem in burners, or emulsification (Oasmaa and Peacocke, 2010).

An alternative route to biofuels is hydrothermal liquefaction, which has the advantage that it can tolerate water. In recent years, biofuel production via hydrothermal liquefaction of wet biomass has been considered one of the promising technologies for biofuel production (Biller et al., 2015). Nonetheless, hydrothermal liquefaction has a limitation of high energy consumption, high capital investments, and the high cost of biogas production (López Barreiro et al., 2013).

Drying of microalgae constitutes a significant economic holdback, as it can incur as much as 75% of the total cost of microalgae harvesting. The drying methods employed for microalgae drying include solar drying, spray drying, freeze-drying, rotary drying, incinerator drying, vacuum-shelf drying and cross-flow air drying (Show et al., 2013). All these methods are either too expensive, thereby making them economically unsuitable, or are seasonal (Sahoo et al., 2017). See the comparison in Table 4.

5. Integrated thermochemical conversion of microalgae

A schematic process of an integrated thermochemical conversion process of microalgae is presented in Fig. 6. The microalgae production (upstream), conversion process (downstream), materials waste and excess heat are integrated into this process. Depending on the type of conversion route, the constituent of the gaseous fraction of thermochemical conversion processes contain gases such as CO, CO_2 , H_2 , light hydrocarbon gases (C₁-C₂), NOx, steam and process heat. CO₂ is one of the major product in the gaseous fraction is recycled as a reactant in the microalgae photosynthesis process (Eq. (1)).

The cultivation of microalgae in wastewater provides a source of nutrient and also doubles as a wastewater treatment (Hoffman et al., 2017; Ranganathan and Savithri, 2019).

The water separated from the microalgae harvest is recycled into the microalgae cultivation system (Batan et al., 2016). The recycled water from microalgae after harvest contains nutrient supplement which is crucial in reducing cost (Farooq et al., 2015).

The cultivation of microalgae can be performed either in an open or closed reactor or a combination of both. The choice of a reactor for microalgae cultivation is influenced by factors such as the microalgae strain, metabolic regime, temperature, harvesting method, drying method and final product of interest (Brennan and Owende, 2010). Commercial microalgae production has usually been focused on small consumer markets such as whole dried microalgae for animal and human feed, and production of pigments (Borowitzka, 2015). Integrated microalgae biomass production can boost the sustainability of microalgae-based production systems on a commercial scale. Table 5 the highlight commercial-scale demonstration of algae biofuel production (ETIP, 2020).

It is only recently that the integrated microalgae biomass production concept has been considered. The concept is carried out by the employment of raw materials close to industrial facilities, such as the colocation of upstream (microalgae wastewater treatment) and downstream (thermochemical conversion processes of microalgae). It is also carried out by the recovery of several compounds from the microalgae biomass (Klein et al., 2018). Fig. 6 depicts such recoveries, as biogas cofeeding with small amount of natural gas, and recirculation of dewatered microalgae into cultivation.

For example, microalgae co-firing in coal power plants for the production of electricity has been claimed to be a promising technology for achieving the low-carbon energy emissions needed to address global warming (Giostri et al., 2016). However, the selected plant was assumed by the authors to be an advanced supercritical (ASC) coal-fired power plant without carbon capture. Being that the economics of natural gas combined cycle test without carbon capture for the production of electricity is superior, exhibiting 58.3% net electric efficiency and 829.9 MW net power output. Whereas, carbon capture has a net efficiency of 49.9% and net electric efficiency of 709.9 MW (EBTF, 2011). Secondly, the effect of the change in the composition of flue gases was neglected, as coal is co-fired with microalgae (1%). Furthermore, CO₂ (14%) in the flue gas was used as a carbon source for algal cultivation, while the biologically available NO_x and SO_x along with CO₂ in the flue gas algal

Table 4

Comparison of advantages and disadvantages of various microalgae biomass drying methods.

Drying method	Process	Advantages	Disadvantages	References
Direct Sun drying	Direct sun	Cheap and easily accessible when available	Could cause change of texture and color of algae due to disintegration of algal chlorophyll. Not available in all climates. Weather-dependent. Low reliability due to dependence of weather - uncontrollable and could cause over heating	(Show et al., 2015, 2013)
Solar	Solar water heating system (glass	More controllable than direct sun	Higher capital cost	(Chen et al., 2011;
drying	panels or tubes)	drying	Low reliability due to dependence of weather	Show et al., 2015)
		High drying rates possible	Not recommended for microalgae product intended for	
		Good solution in certain remote	human consumption due to risk of spillage and	
		locations with access to energy supply	fermentation under prolonged drying	
Spray	High pressure atomization process	Very fast (completed within few	High cost of operation	(Tan et al., 2018)
drying		seconds)	Low digestibility of spray dried algae	
Freeze	Refrigeration	Effective for the disruption of	Too expensive for large scale commercial use	(Chen et al., 2011;
drying		microalgae cells leading to high lipid		Show et al., 2015,
		extraction efficiency		2013)
Rotary	Slope rotating cylinder (movement	Advantage of sterilization and	High cost of energy to operate dryer	(Delrue et al., 2012;
drying	of content by gravity)	disruption		Show et al., 2013)
Flash	Spraying or injecting mixture of	Rapid	Quality of final dried microalgae product are influenced by	(Chen et al., 2011;
drying	wet and dried biomass into hot gas		hot gas source	Show et al., 2015,
	stream			2013)



Fig. 6. Microalgae cultivation systems and thermochemical conversion process integration with: ¹Recirculation of dewatered microalgae into cultivation; ²CO₂ emission from a nearby plant; ³wastewater treatment; ⁴Co-pyrolysis of microalgae with other feedstock; and ⁵Biogas co-fed with small amount of natural gas.

productivity were neglected. Otherwise, the co-firing of coal with significant proportions of torrefied microalgae or microalgae would cause a major environmental concern due to the emission of greenhouse gases.

5.1. Advantage and critical aspect of integrated microalgae system

The potential advantages of integrated microalgae systems for biofuel production are:

- i. the ability to sustainably convert different microalgae biomass into numerous high-value products
- ii. waste reduction by decreasing water input via the cultivation of microalgae in wastewater, also doubling as a wastewater treatment.
- iii. maximize the utilization of biomass
- iv. reduce effluent for treatment
- v. reduced charges of contaminants disposed of in the environment
- vi. reduced cost of microalgae liquid fuels (Das, 2015; Klein et al., 2018).

Large-scale integrated microalgae biorefinery requires a network of units that requires a vast amount of raw materials. This network of units is critical for the production and processing of microalgae biomass. The fundamental factors for the establishment of microalgae refinery are (i) carbon sources for microalgae metabolism, (ii) energy to power industrial equipment, and (iii) land to construct the industrial facilities. (Klein et al., 2018; Mohd-Udaiyappan et al., 2017; Novoveská et al., 2016b).

5.2. Wastewater treatment, carbon and energy sources for microalgae cultivation

Wastewater contains organic carbon, nitrogen, phosphorus and other micro/macronutrients. These nutrients are suitable for microalgae growth and at the same time can be used to treat the wastewater (Xin et al., 2016). Some of the integrated systems obtain carbon from

wastewater systems (Hoffman et al., 2017; Orfield et al., 2014; Xin et al., 2016), some as CO_2 from nearby power plants flue sources (Hoffman et al., 2017; Orfield et al., 2014), or from anaerobic digestion sources (Lundquist et al., 2010) as part of the integrated bioprocessing system. A study has also considered combining microalgae biomass production with sugarcane mills to produce biodiesel from microalgae in colocation with sugar (Lohrey and Kochergin, 2012). A conceptual process was developed in which a portion of the CO_2 produced during the sugar production at the mill was used for algae cultivation, and the excess bagasse was used for the generation of CO_2 and energy for the production of biodiesel from algae. Another example of an integrated biorefinery concept is the production of microalgae biomass integrated with an ethanol biorefinery to maximize the use of CO_2 in microalgae cultivation and utilize waste heat from a nearby ethanol biorefinery (Rosenberg et al., 2011).

5.3. Techno-economic analysis (TEA) and lowest selling prices of liquid biofuels

Techno-economic analysis (TEA) has been used to determine the lowest selling price (LSP) of biofuels and other by-products derived from biomass. It is often a valuable research tool, used to forecast the technical and economic viability of process design concepts.

Table 6 shows the predicted prices of algal oil (\$3.46), biodiesel (\$0.96–3.69), biocrude (\$0.95–\$23.96), drop-in-fuel (gasoline and diesel) (\$1.49–1.8), bio-oils produced from microalgae (\$0.41–0.61) and lignocellulosic biomass (\$1.32–1.58).

5.3.1. Algal oil and biodiesel

The TEA studies and Monte Carlo probabilistic analysis of algal oil (solvent extraction) and biodiesel production systems (via hydroprocessing of extracted solvent lipid (HESL)) integrated with recirculation of dewatered algae into algae cultivation in PBR have shown that LSP of algal oil is \$3.46. While the biodiesel produced showed an LSP of \$3.69 (Batan et al., 2016). However, another study compared the TEA

Commercial- scale demonstration of algae biofuel production.

Company	Product	Product capacity and commercialization level
Sapphire energy (2012)	Green crude (cultivation, to harvest, to extraction of ready-to-	Land size: 100 acres of cultivation ponds and all other processing equipment (300 acre at full capacity)
	refine Green Crude)	Production capacity: 5000–10.000 barrels per day.
Muradel (2014)	Algal crude oil	Production capacity: 30000 l/annum at the cost of \$10.7 m, towards achieving a commercial
. ,	(functionally equivalent to fossil crude oil) via	plant of 80 million liter capacity
	subcritical water reaction	· · · ·
Solazyme and Chevron (2012)	Biodiesel and other value added co-product via:	Production capacity: Contract to supply 450,000 gal for US navy trial
	(i) Production of esters and linear fatty acids from	 Successfully deployed at commercial manufacturing scale
	microalgae, and	 Exceeded military specification
	(ii) their subsequent conversion to produce biodiesel and other value added co-product	• Exceeded requirements for jet fuel ASTM D6751 and EN 14214, and D-975
Algae.Tec Ltd.	Biofuel	• 2015: Conversion of carbon dioxide from energy plants to biofuels in collaboration with
(2012–2015)		Macquarie Generation (Australia), and Reliance (India)
		 2012: Utilization of carbon dioxide from a neighbouring ethanol plant for algal photobioreactors
		2012: industrial-scale algae to aviation biofuels production facility in Europe in collaboration with Lufthansa
Bioprocess Algae	Algae feedstock	 2009: Constructed four commercial scale "Grower Harvester" with the re-use CO₂ and excess heat from an ethanol plant
		• 2013: \$6.4 m funding from United States Department of Energy for the development military
		biofuels, focusing on quicker lipid production and lipids conversion to various hydrocarbons.
Cellena and Nestle	Algae feedstock	Land size: Operated Kona demonstration facility (since 2009) production and research facility
(2013)	Production of algae based feedstock for biofuels, animal feed, and Omega-3 nutritional oils	Production capacity: Produced over 20 metric tons of whole algae (dry weight)
		 Processed highly diverse strains of algae
Algenol Direct to	Ethanol	Land size: 2000 acres of photobioreactors, with provision for additional acreage for future scale-
Ethanol process		up
Joule Demonstration Plant	Ethanol or Hydrocarbon for diesel, jet fuel and gasoline	Production capacity: Targeting 15,000 gal diesel/acre/year and 25,000 gal of ethanol/acre/year
	Engineered catalysis to continuously convert waste	
	CO ₂ to renewable fuel	

studies of biodiesel production using UA and NREL pathways co-fed with biogas and a small amount of natural gas for heating, pretreatment to anaerobic digestion and combined heat and power (CHP), and CO₂ captured used for algal cultivation (Dutta et al., 2016). The LPS of \$0.96 was observed for the NREL pathway that involved lipid extraction, fermentation, distillation and hydrodeoxygenation, while the LSP of \$2.32 was reported for UA pathway that involved solvent extraction, trans-esterification and product purification for biodiesel production. There is a significant difference between the LSP of HESL and NREL pathways compared to that of the UA pathway. The UA pathway is a conventional production method of cultivation, harvesting, lipid production and product upgrading steps and lacks the addition of credits from the co-product. However, the economics of the NREL pathway is better owing to the integration of the production of various co-product adding revenue in return to the entire system (Dutta et al., 2016). Though more TEA studies on microalgae focused on the open pond, and there are fewer TEA studies on closed PBR especially on a commercial scale systems operation data and models. However, TEA and Monte Carlo probabilistic analysis of microalgae cultivation in a PBR for biodiesel production demonstrated the economic impact of the utilization of co-product in biodiesel production.

5.3.2. Biocrude

A study of biocrude produced via hydrothermal liquefaction from microalgae cultivated in an open pond and closed PBR without nutrient integration indicate that the LSP of biocrude produced in a closed PBR cost \$16.92 while that of the open pond with LSP is \$23.96 (Richardson et al., 2014). Biocrude produced via hydrothermal liquefaction of microalgae in an open pond integrated with carbon source from contaminated water system exhibited an LSP of \$1.83 (Hoffman et al., 2017). Furthermore, biocrude produced via hydrothermal liquefaction of microalgae produced in an algal scrubber with integration with CO₂

by power plant flue sources showed a promising LSP of \$1.38 (Hoffman et al., 2017). While an integration in microalgae production with wastewater treatment and anaerobic digestion of liquid digestate to produce microalgae for biocrude production via hydrothermal lique-faction showed a significantly lower LSP of \$ 0.95 (Ranganathan and Savithri, 2019). A significantly high LSP of biocrude is observed for microalgae cultivated both the open pond and closed PBR without nutrient integration (\$16.92 -\$23.96) (Richardson et al., 2014) despite the water-tolerant nature of hydrothermal liquefaction processes. These results, however, indicate the advantages of nutrient recycling, deployment of CO_2 flue sources from power plants and the utilization of microalgae biofuel co-product in the addition of revenue in the development and implementation of biocrude production processes.

5.3.3. Gasoline and diesel

The TEA of partial mechanical dewatering (MDCP) and thermal drying (TDCP) technologies of microalgae remnant (from microalgae cultivated in wastewater), followed by the conversion of dried microalgae into drop-in transportation fuel via catalytic pyrolysis and upgrading showed that the LSPs of the biofuels produced via the partial mechanical and thermal drying pathways were \$1.5 and \$1.8 respectively (Thilakaratne et al., 2014). These results demonstrate microalgae remnant, a co-product of microalgae lipid extraction as a promising feedstock for the aforementioned processes. The most significant challenge of these processes however is the high content of moisture of harvested microalgae. Consequently, had an impact on the energy flow analysis of lower energy conversion fuels and power for TDCP than MDCP. The lack of available data for important factors of microalgae growth and harvest in the sensitivity analysis of the microalgae remnant biofuel production is a constraint reported to limit microalgal fuel price in the studies.

Comparison of lowest selling price (LSP) of biofuels derived from microalgae using Technoeconomic analysis (TEA).

Biofuel	Summary of process	description	LSP ^h	Ref
		-	(US	
			\$/L)	
Algae oil	Feedstock	Microalgae (PBR)	3.46	(Batan et al., 2016)
(lipid)	Processing method	Algal lipid solvent extraction		
	Level of	Recirculation of dewatered algae into Algae cultivation		
	integration			
Biodiesel	Feedstock	Microalgae (PBR)	3.69	(Batan et al., 2016)
	Processing method	Hydro-processing of extracted solvent lipid		
	Level of	Recirculation of dewatered algae into Algae cultivation		
D: 1: 1	integration		0.069	
Biodiesei	Feedstock Drogossing mothod	Microalgae (Open pond) Lipid autroation formentation distillation and hydrodecovyconation (NBEL nethway) ⁱ	0.96°	(Dutta et al., 2016)
	Level of	Biogas co-fed with small amount of natural gas for heating		
	integration	Pretreatment to anaerobic digestion and CHP		
	megrution	CO ₂ captured and used for algal cultivation		
Biodiesel	Feedstock	Microalgae (Open ponds)	2.32^{g}	(Dutta et al., 2016)
	Processing method	Solvent extraction, trans-esterification and product purification for biodiesel (UA		
		pathway) ^k		
	Level of	Biogas co-fed with small amount of natural gas for heating		
	integration	Pretreatment to anaerobic digestion and CHP		
		CO ₂ captured and used for algal cultivation	9	
Biocrude	Feedstock	Microalgae (Open pond)	1.835	(Hoffman et al., 2017)
	Processing method	Hydrothermal liquefaction		
	integration	Containinated water system		
Biocrude	Feedstock	Microalgae (Algal turf scrubber)	1 38 ^g	(Hoffman et al. 2017)
Dioci ude	Processing method	Hydrothermal liquefaction	1.00	
	Level of	CO ₂ by power plant flue sources		
	integration	- • • •		
Biocrude	Feedstock	Microalgae (Open pond)	23.96 ^g	(Richardson et al., 2014)
	Processing method	Hydrothermal treatment		
	Level of	NIL		
	integration		4 6 9 9 9	
Biocrude	Feedstock	Microalgae (Closed PBR)	16.92°	(Richardson et al., 2014)
	Processing method	Nui		
	integration	NIL		
Biocrude	Feedstock	Microalgae (Raceway pond)	0.95 ^g	(Ranganathan and Savithri,
	Processing method	Hydrothermal liquefaction/Hydrotreatment/Hydrocracking		2019)
	Level of	Wastewater treatment and anaerobic digestion of liquid digestate		
	integration			
Bio-oil	Feedstock	Pine wood	$1.32^{g,i}$	(Shemfe et al., 2017)
	Processing method	Single stage regeneration with a catalyst cooler (P-1RGC)		
	Level of	NIL		
D: 1	integration		1 FORI	
B10-011	Feedstock	Pine wood	1.58%	(Shemfe et al., 2017)
	Level of	NII		
	integration	NIL .		
Bio-oil	Feedstock	Microalgae (Pond)	0.61	(Orfield et al., 2014)
	Processing method	Oil extraction		
	Level of	Flue gas and waste water co-utilization		
	integration			
(Gasoline &	Feedstock	Microalgae remnant	1.49 ^c	(Thilakaratne et al., 2014)
diesel)	Processing method	Mechanical drying/Catalytic pyrolysis		
	Level of	Waste water treatment		
(Casalina &	Integration	Microalcoa rompont	1 00d	(Thildkerstree et al. 2014)
(Gasolille &	Processing method	Thermal drving (Catalytic pyrolysis	1.00	(Illiakaratile et al., 2014)
unout	Level of	Waste water treatment		
	integration			
Bio-oil	Feedstock	Defatted microalgae	0.49 ^g	(Xin et al., 2016)
	Processing method	Microwave-assisted pyrolysis		
	Level of	Combined wastewater and microalgae cultivation for biofuel production		
	integration			
Bio-oil	Feedstock	Microalgae (PBR)	0.41 ^g	(Xin et al., 2018)
	Processing method	Microwave-assisted pyrolysis of both algae and sludge;		
	Level of	Combined wastewater and microalgae cultivation for biofuel production		
	integration			

a - Euro to US\$ conversion 2011:0.78; b - Production cost; c - Mechanical dewatering; d - Thermal dewatering; e - no credit for wastewater treatment considered; f - wastewater revenue from BOD removal included; (Lundquist et al., 2010); HTL - Hydrothermal liquefaction; FWC - Flue gas and waste water co-utilization, N/A - not available, g - imperial gallon (4.55 L) was used for conversion of gallon to liter; h - lowest selling price (LSP) of biofuel product, i - £GB to US\$ conversion 1:1.25, j - National Renewable Energy Laboratory pathway, k - University of Aveiro pathway.

5.3.4. Bio-oil

This has included studies of zeolites as catalysts both for the in-situ cracking and bio-oil upgrading process. However, zeolite regeneration was often overlooked. Shemfe et al. (2017) conducted TEAs of fast pyrolysis of pine and bio-oil upgrading via zeolite cracking with (1) two generators operating in sequence (P-2RG) and (2) a single generator fitted with a cooler (P-1RGC). The P-2RG scheme exhibited a 2% higher energy efficiency than the P-1RGC scheme. However, the P-1RGC exhibited a slightly lower LSP of \$1.32 than the P-2RG, at \$1.58. However, the TEA studies of bio-oil produced from microalgae feedstock coupled with flue gas and wastewater co-utilization exhibited an LSPs of (\$0.61) (Orfield et al., 2014). TEA of combined wastewater and microalgae cultivation for biofuel production via microwave-assisted pyrolysis have shown that the LSPs of bio-oils derived from the microwaveassisted pyrolysis of defatted microalgae was \$0.49 (Xin et al., 2016) and for microalgae was \$0.41 (Xin et al., 2018). The exploitation of coproduct and enhancement of important technologies in microwaveassisted pyrolysis considerably impacted the whole economic process of integrated microalgal biomass system (Xin et al., 2016). Meanwhile, the price of crude oil, the energy consumption of pyrolysis, LHV of sludge oil and sludge oil ratio were the key factors affecting the integrated system incorporating sludge, scum treatment and biofuel development from microalgae via microwave-assisted pyrolysis (Xin et al., 2018).

The data in Table 6 share a common aim to identify economic prospect to help aid the development of microalgae biofuel technology. So far, the economic viability of biofuels is somewhat influenced by the type of feedstock, processing technique and level of integration. However, there is still need for more research to address the economic constraints that lead to the high cost of biofuel production (Xin et al., 2016) for biofuels to be able to compete with fuels derived from conventional fossil fuels: 0.25 L^{-1} for an average annual cost of organization of petroleum exporting countries (OPEC) price (Sonichsen, 2020b) and 0.31 L^{-1} for Brent crude (Economics, 2020).

6. Challenges and prospects

Microalgae is potentially a key future source of biofuels and other valuable by-products due to its rapid growth rate, high productivity per unit area, its ability to grow in a wide range of aquatic habitats, and the huge variety of natural and genetically modified microalgae species. However, before the conversion of microalgae biomass to fuels, the process stages, cultivation, harvesting, and drying, incur significant running and capital costs, substantially increasing the cost of production. Hence, future research and development should focus on:

- (i) Alternative glucose production technologies for heterotrophic and mixotrophic microalgae cultivation. Glucose is typically derived from the plant-based feedstock. It is, therefore, essential to prevent the potential food versus fuel crisis, especially if glucose would be used in microalgae cultivation on an industrial. Glycerol and lignocellulosic derived glucose could be developed as alternative technologies.
- (ii) Developing integrated systems that co-produce high-value products. This will have environmental benefits, in that it will minimize waste and reduce the emission of greenhouse gases (GHG), but there are obvious economic benefits, too, that should enable the price of the biofuels to be reduced.
- (iii) Reduction of GHG emission and energy cost. This can be achieved via the utilization of flue gases from conventional power plants, nutrient recycling (perhaps from wastewaters), and utilization of other microalgae biofuel co-product.
- (iv) Increased mechanical dewatering. This will increase efficiency and lower the selling price of biofuel, by reducing the degree of thermal drying, which is extremely energy-intensive.

TEA and sensitivity analysis studies are difficult due to the dearth of available data for microalgae growth and harvest for the production of microalgae-derived fuels.

A cost-effective microalgae-derived biofuel is yet to be achieved. Hence, there is still a substantial requirement for underpinning research on microalgae processing, to facilitate commercialization of microalgae as a feedstock for biofuel production. This is essential in addressing the current economic constraints of microalgae biofuel production. Current estimates of its costs are:

- (i) Biodiesel via transesterification between 0.96 US\$/L (Dutta et al., 2016) to 3.69 US\$/L (Batan et al., 2016).
- (ii) Bio-oil via pyrolysis between 0.41 (Xin et al., 2018) to 0.61 US\$/L (Orfield et al., 2014).
- (iii) Biocrude via hydrothermal liquefaction between 0.95 US\$/L (Ranganathan and Savithri, 2019) to 23.96 US\$/L (Richardson et al., 2014).

The upgrading of crude microalgae oil (lipid) estimated to costs about 3.5 US\$/L to biofuel cannot compete with fuel derived from conventional petroleum oil at 0.43-0.44 US\$/L (Economics, 2018; Sonichsen, 2020b). Biodiesel via transesterification exhibits an efficiency of 92% (MJ_{fuel}/MJ_{feedstock.drv}), essentially due to the high quality of microalgae lipid. Meanwhile, transesterification exhibit low potential for development, as it is close to attaining technical optimum (IRENA, 2016). The most promising thermochemical conversion technology for the production of liquid-derived biofuel from microalgae is pyrolysis, as it is estimated to have the lowest LSPs ranging between 0.41 and 0.61 US \$/L. The maximum theoretical conversion efficiency (MJ_{fuel}/MJ_{feedstock}, dry) for pyrolysis technology is 50%, higher than that of hydrothermal liquefaction (aqueous phase reforming) by about 32%. Meanwhile, the projected conversion efficiencies in the subsequent three decades are 65% for the pyrolysis, and 49% for aqueous phase reforming (IRENA, 2016).

The co-processing of microalgae with other feedstocks mitigates CO_2 , as microalgae consume CO_2 for photosynthesis process. Integrated microalgae biorefinery concepts utilize waste, excess heat and reduce or eliminate the cost of transportation. The development of novel cheap and environmentally-friendly catalysts for microalgae conversion processes could enhance the yield of biofuels. All of these would facilitate cost-effective renewable biofuel.

A variety of studies on the life cycle assessments (LCA) of microalgae biofuels were performed, based on laboratory data. However, to be accurate these models require data from industrial-scale conversion processes of microalgae, and the streamlining of allocation methods (Vienescu et al., 2018) would enable a more accurate assessment of the environmental impact of microalgal biofuels.

7. Conclusion

The early stages of microalgae cultivation systems (growth, harvesting and drying) have been reviewed as a precursor to thermochemical conversion processes, with particular regard to the process economics, as harvesting and drying are the most energy-intensive steps in the overall process. The following measures should be implemented to overcome various commercial hurdles to microalgal biofuel production:

- (i) predatory control: a simple means of increasing biomass concentration.
- (ii) increased mechanical dewatering: to reduce overall energy usage.
- (iii) modification of conventional cultivations systems: geometrical improvements to increase biomass productivity.
- (iv) integrated production: based around microalgae-to-bio-oil, as this is the lowest cost product.

Declaration of competing interest

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

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