frontiers
in Marine Science

MINI REVIEW
published: 05 February 2019
doi: 10.3389/fmars.2019.00029

Overview of Carbon Capture Technology: Microalgal Biorefinery Concept and State-of-the-Art

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The impending danger of climate change and pollution can now be seen on the world panorama. The concentration of CO_2, the most important Green House Gas (GHG), has reached to formidable levels. Although carbon capture and storage (CCS) methods have been largely worked upon, they are cumbersome in terms of economy and their long term environmental safety raises a concern. Alternatively, bio-sequestration of CO_2 using microalgal cell factories has emerged as a promising way of recycling CO_2 into biomass via photosynthesis which in turn could be used for the production of bioenergy and other value-added products. Despite enormous potential, the production of microalgae for low-value bulk products and bulk products such as biofuels, is heretofore, not feasible. To achieve economic viability and sustainability, major hurdles in both, the upstream and downstream processes have to be overcome. Recent technoeconomic analyses and life-cycle assessments of microalgae-based production systems have suggested that the only possible way for scaling up the production is to completely use the biomass in an integrated biorefinery set-up wherein every valuable component is extracted, processed and valorized. This article provides a brief yet comprehensive review of the present carbon sequestration and utilization technologies, focusing primarily on biological CO_2 capture by microalgae in the context of bio-refinery. The paper discusses various products of microalgal biorefinery and aims to assess the opportunities, challenges and current state-of-the-art of microalgae-based CO_2 bioconversion, which are essential to the sustainability of this approach in terms of the environment as well as the economy.

Keywords: microalgae, biorefinery, carbon capture, bio-sequestration, CO_2 mitigation, biofuel

OPEN ACCESS

Edited by:
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Specialty section:
This article was submitted to Marine Biotechnology, a section of the journal Frontiers in Marine Science

Received: *01 August 2018*
Accepted: *21 January 2019*
Published: *05 February 2019*

Citation:
Singh J and Dhar DW (2019) Overview of Carbon Capture Technology: Microalgal Biorefinery Concept and State-of-the-Art. Front. Mar. Sci. 6:29. doi: 10.3389/fmars.2019.00029

INTRODUCTION

The increased concentration of Green House Gases (GHGs) are causing dramatic climatic changes (rise in temperature, changes in the distribution, intensity and pattern of rainfall, rising sea levels, floods, droughts and increased occurrence of extreme climatic phenomena) as a result of well-known phenomenon "Global Warming" (Alexander et al., 2006; Church and White, 2006; Rignot and Kanagaratnam, 2006; Meinshausen et al., 2009; Rockstrom et al., 2009; Solomon et al., 2009; Dawson et al., 2011). The temperature of the planet has risen by 0.85°C from 1880 to 2012 and it has been forecasted that by the end of this century, a rise of 1.4–5.8°C would be witnessed

(De Silva et al., 2015). The concentration of CO_2, the most important GHG and the major contributor to global warming, has reached to formidable levels. Corresponding to a 32% increase, from around 280 ppm to 400 ppm, since the industrial revolution (De Silva et al., 2015). The primary causes being irrational use of fossil fuels and change in land use pattern (Goldemberg, 2007; Atsumi et al., 2009). Not merely global warming, the increased CO_2 concentration in the atmosphere has also led to a 30% increase in the ocean acidity, which in turn is affecting the biodiversity adversely (Doney et al., 2009; Hofmann and Schellnhuber, 2010; Farrelly et al., 2013). The Kyoto Protocol and the Paris Agreement (2015), have set a number of policy actions for participating countries to curb climate change impact. The major requirement being reduced CO_2 emissions by reduced fossil fuel utilization and increased carbon capture and sequestration (Cheah et al., 2016; Pires, 2017). This minireview aims to discuss briefly yet comprehensively the various CCS methodologies, focusing mainly on the potential of microalgae mediated carbon capture within the framework of a biorefinery approach: bioconversion and valorization of captured CO_2, current state of the technology, recent developments, challenges and future prospects.

CO_2 CAPTURE AND STORAGE METHODS

Currently there are many physico-chemical carbon capture and sequestration strategies that are combinedly categorized as carbon capture and storage (CCS) methodologies. CCS operate over 3 major steps: CO_2 capture, CO_2 transportation and CO_2 storage. CO_2 capture is done from large point sources such as power plants and cement manufacturing plants. The separation and capture of CO_2 from other exhaust components is usually done via following methods: (i) chemical absorption; (ii) physical adsorption; (iii) membrane separation; and (iv) cryogenic distillation (Figueroa et al., 2008; Pires et al., 2011, 2012). This highly concentrated CO_2 is then compressed and transported to storage points via pipelines or ship (Svensson et al., 2004; McCoy and Rubin, 2008). Next, the captured CO_2 is stored into reservoirs, viz. geological storage, oceanic storage wherein the CO_2 is directly injected deep into the ocean, saline formations, aquifers or depleted oil/gas wells (Lackner, 2003). Despite remarkable storage potential of the aforementioned CCS, considerable drawbacks remain, including expensive operation and transportation, environmental threat of long term CO_2 leakage and other uncertainties (Lam et al., 2012; De Silva et al., 2015). Moreover, physico-chemical CCS methods are practically successful only for capturing CO_2 from point sources producing high concentrations of CO_2 i.e., diffused, non-point emissions and low concentrations of CO_2 cannot be captured (Nouha et al., 2015). **Table 1** briefly illustrates the various CCS methodologies, their mechanisms, merits and limitations with respective references. Aside to physical and chemical CCS, the biological route can be taken for capturing CO_2 via natural sinks: (i) forestation; afforestation, reforestation, and the farming of crops and livestock, the biomass can be further valorized (Farrelly et al., 2013; Cheah et al., 2016). (ii) ocean fertilization; fertilizing oceans with iron and other nutrients prompting increased carbon dioxide uptake by the phytoplanktons (Williamson et al., 2012) (iii) microalgae cultivation (Lam et al., 2012; Cheah et al., 2016; Yadav and Sen, 2017; Zhou et al., 2017).

CO_2 Capture by Microalgae

The term "microalgae" is generally used for both prokaryotic blue green algae (cyanobacteria) and eukaryotic microalgae including green algae, red algae, and diatoms. Microalgae are being sought as alluring biofactories for the sequestration of CO_2 and simultaneous production of renewable biofuels, food, animal and aquaculture feed products and other value-added products such as cosmetics, nutraceuticals, pharmaceuticals, bio-fertilizers, bioactive substances (Ryan, 2009; Harun et al., 2010). Microalgae possess strategies, well known as CO_2 concentrating mechanism (CCM) for efficiently photosynthesizing by acquiring inorganic carbon even from very low atmospheric CO_2 concentrations (Whitton, 2012). These microorganisms surpass other feedstocks in terms of their abilities to flourish in extreme environments and simple yet versatile nutritional requirements. Microalgae do not require arable land and are capable of surviving well in places that other crop plants cannot inhabit, such as saline-alkaline water, land and wastewater (Searchinger et al., 2008; Wang et al., 2008). Furthermore, microalgae can be fed with notorious waste gasses such as CO_2 and NO_x, SO_x from flue gas, inorganic and organic carbon, N, P and other pollutants from agricultural, industrial and sewage wastewater sources so as to provide us with opportunities to transform them into bioenergy, valuable products and forms that cause least harm to the environment (Chisti, 2007; Hu et al., 2008; Pires et al., 2012; Singh and Thakur, 2015). The uncomplicated cellular structures and rapid growth of microalgae endow them with CO_2 fixation efficiency as higher as 10–50 folds than terrestrial plants (Li Y. et al., 2008; Khan et al., 2009).

Recently, many research studies have come up showing the positive impact of growing microalgae under high concentrations of Ci in the form of pure gaseous CO_2, real or simulated flue gas, or soluble carbonate (bicarbonate), reporting increased carbon bio-fixation and biomass productivity (Ho et al., 2010; Sydney et al., 2010; Yoo et al., 2010; Tang et al., 2011; Singh et al., 2014; Aslam et al., 2017; Kuo et al., 2017). Detailed information can be found in elaborated reviews by Lam et al. (2012); Cheah et al. (2015); Thomas et al. (2016); Vuppaladadiyam et al. (2018). The fate of the supplied carbon can end up in making skeleton for lipids, proteins, sugars and pigments (Sydney et al., 2010). Despite such remarkable potential, the production of microalgae for low-value bulk products, such as proteins for food/feed applications, fatty acids for nutraceuticals or bulk products such as biofuels, is heretofore, not economically feasible (Williams and Laurens, 2010; Zhou et al., 2017). Recent technoeconomic analyses and life-cycle assessments of microalgae-based production systems have suggested that the only possible way of realizing the potential production is to completely use the biomass in an integrated biorefinery set-up wherein every valuable component is extracted, processed and valorized (Chew et al., 2017).

TABLE 1 | Comparative description of different carbon capture technologies.

Method	Mechanisms	Advantages	Shortcomings	References
CO_2 capture				
Adsorption	CO_2 capture using solid adsorbent such as activated carbon, zeolite, Na_2CO_3, CaO, etc.	• Low waste generation	• Energy inefficient • Flue gas pre-treatment necessary before channeling to adsorber due to high moisture content and presence of contaminants (e.g., SOx and NOx)	Li G. et al., 2008; Hunt et al., 2010; Pires et al., 2011; Wang et al., 2011; Lam et al., 2012
	CO_2 capture using metal–organic frameworks (MOFs)	• High porosity crystallinity and high surface area	• Powdered MOFs have low mechanical strength and difficult handling	Lin et al., 2016; Nandasiri et al., 2016; Trickett et al., 2017
Chemical absorption	Based on chemical absorption and desorption. CO_2 dissolved/captured chemical solvents, such as monoethanolamine (MEA), amine and potassium hydroxide (KOH)	• High CO_2 solubility • Thermally stable	• High solvent loss due to evaporation • React with components other than CO_2, like SO_2 resulting in irreversible degeneration of solvent • High energy consumption for solvent regeneration • Thermally unstable • Equipment corrosion	Kittel et al., 2009; Cole et al., 2011; Pires et al., 2011
	Ionic liquid for CO_2 absorption	• Environmentally safer as substitute the use of hazardous solvents	• Cost intensive • Difficult to scale-up ionic liquids	Ziobrowski et al., 2016
Membrane technology	Separation of CO_2 from the main stream by passing through a membrane that acts as a filter with selective permeability. Usually polymeric membranes are used	• High separation efficiency and packing density due to the small installation requirements	• Energy intensive as cooling of hot flue gas is essential • High moisture content in the flue gas affects membrane performance due to competitive sorption and plasticisation of the polymer • High membrane cost, fouling of membrane and high membrane surface area requirement	Scholes et al., 2009; Pires et al., 2011; Lam et al., 2012
Cryogenic separation	Consecutive refrigeration and condensation of gas mixture at different condensation temperatures to separate CO_2	• High capture efficiency (up to 99.9%)	• High energy requirement for refrigeration • Flue gas moisture removal is required before cooling to avoid plugging by ice formation • Solidified CO_2 is continuously built up on the heat-exchanger surfaces and needs to be removed.	Tuinier et al., 2010; Lam et al., 2012
CO_2 storage				
Geological sequestration	Injection of CO_2 into deep geological reservoirs, depleted oil/gas wells, and coal seams	• Huge storage capacity and use of saline formations, barren spaces • Replenish depleted oil/gas reserves	• High operational cost • Risk of CO_2 leakage and environmental damage • Specific geomorphic structure requirement	White et al., 2003; Kovscek and Cakici, 2005; De Silva et al., 2015
Oceanic injection	Injection of CO_2 into deep ocean	• Huge CO_2 storage capacity	• Cost intensive • Potential threat to marine life	Kita and Ohsumi, 2004; Zhou et al., 2017
Biological CO_2 capture				
Forestation	Afforestation, reforestation, and the farming of crops and livestock	• No hazards of chemicals	• Long time requirement • Large area requirement • Can affect biological diversity • Compete with food crops for arable land	Farrelly et al., 2013; Cheah et al., 2016
Oceanic fertilization	Fertilizing oceans with iron and other nutrients prompting increased carbon dioxide uptake by the phytoplanktons	• Significant potential for CO_2 capture	• Cost intensive • May have uncertain and unintended impacts • May affect marine biodiversity	Williamson et al., 2012
Microalgae-based carbon capture and utilization	Bioconversion CO_2 into biofuels and other valuable products via photosynthesis	• Highly efficient in a wide range of CO_2 concentration • Faster growth rate than plants • No requirement for arable land • Co-production of food, feed, biofuel and value-added products	• Economically cumbersome culture systems and downstream processing mainly harvesting • Sensitive to other flue gas components (NOx, SOx), predation, contamination and extreme culture conditions (pH, temperature, salinity etc)	Ryan, 2009; Harun et al., 2010; Kao et al., 2014; Singh et al., 2014; Varshney et al., 2014

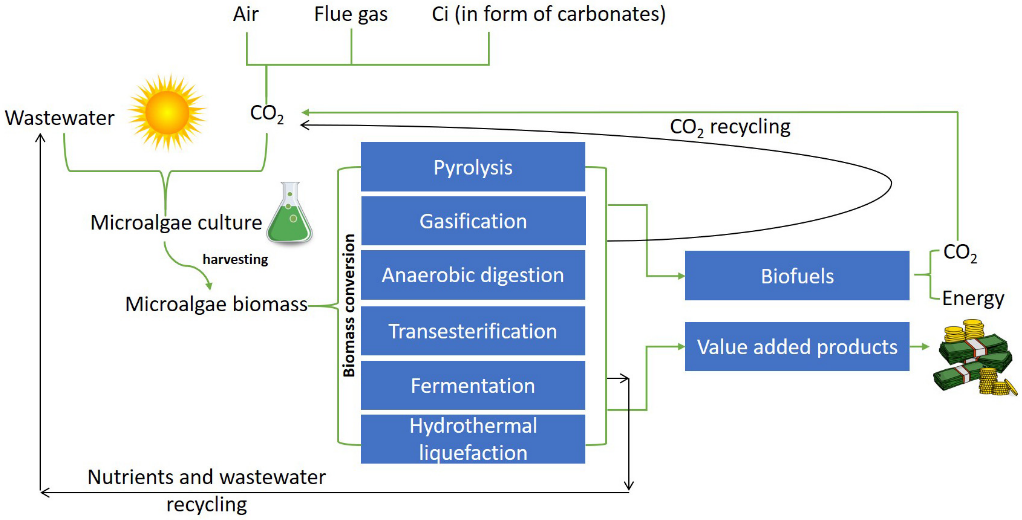

FIGURE 1 | A simplistic representation of microalgal based biorefinery system.

BIOREFINERY CONCEPT OF MICROALGAL BIOMASS

The concept of valorization of a raw material into marketable products is well known in fossil fuel refinery, similarly biorefinery concept refers to the conversion of biomass into multiple commercially valuable products and fuels (Pérez et al., 2017). **Figure 1** depicts a simplistic microalgal based biorefinery system. The various high value and low value marketable products that can be produced in an integrated biorefinery system are discussed in the following sections.

Biofuels

Rising CO_2, resultant global warming and depleting oil reserves are fueling the search for more eco-friendly forms of alternative energy. The microalgal biomass majorly constituted of lipids (7–23%), proteins (6–71%) and carbohydrates (5–64%), depending upon the microalgal specie and culture conditions (Brown, 1991; Becker, 2007; Mata et al., 2010). Microalgae have received great attention as feedstocks for production of biodiesel, biogas, biohydrogen, bioethanol, biobutanol. Biofuels from microalgae, production system, conversion technologies, life cycle analyses have been extensively reviewed, hence detailed description is not presented in this review.

Biodiesel

Microalgae are known to accumulate remarkable amount of lipid. As reviewed by Mata et al. (2010), the lipid content of common microalgae such as *Chlorella, Dunaliella, Isochrysis, Nannochloris, Nannochloropsis, Neochloris, Phaeodactylum, Porphyridium,* and *Schizochytrium,* varies between 20 and 50% of cell dry weight, that can be augmented to higher levels by manipulating environmental and other growth factors, process optimization and genetic modifications of the production strain. Nitrogen starvation and salinity stress are known to induce an increase in TAG (triacylglycerol) accumulation and relative content of oleic acid in most of the microalgal species (Choi et al., 2011). The fatty acid composition of most of the microalgae is dominated by C14:0, C16:0, C18:1, C18:2, and C18:3 fatty acids, yet the relative composition varies from species to species (Gouveia and Oliveira, 2009). Also, the role of HCO_3^- in inducing TAG accumulation has been widely illustrated recently (Gardner et al., 2012, 2013; Lam et al., 2012; White et al., 2013). The lipids can be converted into FAMEs (fatty acid methyl esters) via transesterification for biodiesel production. The major by-product- glycerol also finds enormous industrial application opportunities. Furthermore, the residual de-oiled microalgal biomass can be used for animal feed.

Biogas

Microalgal biomass can be efficiently used for the production of biogas, including methane, hydrogen, and biohythane (combination of methane and 5–25% hydrogen gas) (Ghimire et al., 2017). The resistance of cell wall to enzyme hydrolysis is one of the prime bottleneck in the Anaerobic digestion (AD) process. The overall economic feasibility of the process depends on the factors affecting AD, microalgal strain, biomass pretreatment, and culture methods (Jankowska et al., 2017). Lately, to make the system economically viable and environmentally sustainable, a closed-loop production scheme is being adopted wherein AD effluents are recycled and used as an input in the first step of AD. Jankowska et al. (2017) have presented a detailed review microalgae's cultivation, harvesting and pretreatment for AD for biogas production.

Bioethanol

The carbohydrate part (mainly glucose, starch, cellulose, and hemicellulose) of the microalgal dry biomass can be used for transforming into bioethanol via fermentation. Although, microalgae accumulate relatively low quantities of sugars, the absence of lignin from microalgal structure makes them advantageous over other feedstock such as corn, sugarcane, and lignocellulosic biomass (Odjadjare et al., 2015; Jambo et al., 2016). *Isochrysis galbana, Porphyridium cruentum, Spirogyra sp., Nannochloropsis oculate, Chlorella sp.,* are mainly exploited microalgae for the production of carbohydrates (Markou and Nerantzis, 2013).

Biobutanol

The green residual after microalgae oil extraction can be utilized for the production of biobutanol. The higher energy density of biobutanol and its molecular similarity to gasoline makes it more suitable than biomethanol or bioethanol as biofuel. Aside to being a biofuel, it can also be used as a solvent for industrial purposes (Yeong et al., 2018). Despite having notable significance, limited number of studies have reported laboratory stage work on the fermentation of microalgae biomass to butanol (Cheng et al., 2015; Gao et al., 2016; Wang et al., 2016). Microalgal strains with high starch and convertible sugars concentrations would be ideal for biobutanol production research. *Tetraselmis subcordiformis, Chlorella vulgaris, Chlorella reinhardtii,* and *Scenedesmus obliquus* could be among the potential candidates (Yeong et al., 2018).

Value-Added Products

In the context of biorefinery approach, intracellular compounds and metabolites have gained immense importance owing to their high monetary value. Microalgal pigments: chlorophyll a and b, lutein, astaxanthin, β-carotene, phycobilins, C- phycocyanin have found wide application in dyes, cosmetics, food and feed additives, nutraceuticals and pharmaceuticals, as natural colors, bioactive components, anti-oxidants, nutritive and neuro-protective agents (Koller et al., 2014; Begum et al., 2016). Microalgae are also exploited as rich source of amino acids (leucine, asparagine, glutamine, cysteine, arginine, aspartate, alanine, glycine, lysine, and valine), Carbohydrates (β1–3-glucan, amylose, starch, cellulose, and alginates), Vitamins and minerals (vitamin B1, B2, B6, B12, C, and E; biotin, folic acid, magnesium, calcium, phosphate, iodine) that are widely used in Food additives, health supplements and medicine. Microalgae, such as *Nannochloropsis, Tetraselmis,* and *Isochrysis* are used for extraction of long chain fatty acids popularly known as the omega fatty acids such as DHA (Docosahexaenoic Acid) and EPA (Eicosapentaenoic Acid), have lately gained prime attention as essential for human brain development and health. Other than these, microalgae are also used for production of Extracellular Polymeric Substances (EPSs) which have many industrial applications and Polyhydroxyalkanoates (PHAs). PHAs can be used for manufacturing bioplastics that are very sought after because of their biodegradability (Markou and Nerantzis, 2013; Koller et al., 2014).

State-of-the-Art

Although many have reported successful utilization of microalgal biomass for the production of bioproducts within a biorefinery framework, the economic feasibility is unrealized and the microalgae biorefinery is way much expensive ('t Lam et al., 2017; Zhou et al., 2017). To attain feasibility and sustainability, both upstream processing (USP) and downstream processing (DSP) need to be efficiently simplified and integrated. The efficiency of the USP is determined by microalgal strain selection, nutrient supply (CO_2, N, and P) and culture conditions (temperature, light intensity) (Vanthoor-Koopmans et al., 2013). Whereas, the constraints at the DSP level are mainly characterized by harvesting, cell disruption, and extraction methods. DSP, specifically harvesting accounts for 20–40% of the total production costs and for a multi-product biorefinery, the cost increases to 50–60% ('t Lam et al., 2017).

Bioprospecting suitable microalgae is a crucial but time intensive step, high throughput screening techniques like 96-well microplate swivel system (M96SS) have made processing upto 768 microalgal samples at the same time, possible (Han et al., 2012; Zhou et al., 2017). Microalgal production strains can be improved by induced acclimation through manipulation of various environmental stresses (Chen et al., 2017; Schüler et al., 2017). Aslam et al. (2017) showed that mixed diverse community of microalgae, dominated by *Desmodesmus spp*., could be adapted over a time of many months to survive in 100% flue gas from an unfiltered coal-fired power plant containing 11% CO_2. Carbohydrate and starch accumulation in *Chlorella* sp. AE10 was improved by a two staged process wherein the CO_2 concentration, light intensity, nitrogen concentration was changed drastically and cells were diluted at onset of 2nd stage resulting in a 42% increase in carbohydrate accumulation (Cheng et al., 2017). Besides stress manipulation and acclimatization, desirable traits of the microalgal strains can be effectively improved by genetic and metabolic engineering/synthetic biology. Lately, genome editing tools such as Clustered Regularly Interspaced Short Palindromic Repeats – CRISPR associated protein 9 (CRISPR-Cas9) and Transcription Activator-Like (TAL) Effector Nucleases (TALEN) are being used in microalgal gene alterations. Moreover, gene-interfering tools, such as CRISPR-dCas9, micro RNA (miRNA), and silence RNA (siRNA) are being explored to alter the gene expression unlike gene modification. Synthetic biology engages the use of "biobricks" to create artificial regulatory pathways that can control a desired cellular trait by modifying the metabolism. Interchangeable units such as promoters, ribosome-binding sites (RBS), terminators, *trans*-elements and regulatory molecules serve as the biobricks. Recent developments in microalgal genetic and metabolic engineering can be found in detailed reviews by Ng et al. (2017) and Jagadevan et al. (2018). Recently, Yang et al. (2017), genetically engineered the calvin cycle of *Chlorella vulgaris* enhancing its photosynthetic capacity by ∼1.2-fold, by introducing the cyanobacterial fructose 1,6-bisphosphate aldolase, guided by a plastid transit peptide. Kuo et al. (2017), screened an alkali-tolerant, *Chlorella* sp. AT1 mutant strain by NTG (N-methyl-N′-nitro-N-nitrosoguanidine) mutagenesis that survived well 10% CO_2 for prospective CO_2 sequestration.

Large scale microalgal cultivation and nutrient supply pose huge economic burden. In this context emphasis is being laid on biofilm based attached cultivation rather than aqua-suspend methods that have massive water requirement, low biomass productivity, energy intensive and cannot be easily scaled up (Kesaano and Sims, 2014; Wang et al., 2017). Microalgal production using wastewater from industrial, agricultural and sewage sources is a promising way to reduce the ecological footprints substantially (Pires et al., 2012; Singh and Thakur, 2015). Digestates, effluents from biogas production units and AD (containing concentrated nutrients including nitrogen in the form of ammonia, potassium, phosphorous, sulfur, and recalcitrant organic substances), are also being used in microalgal cultivation systems. A recent elaborated review has been done by Koutra et al. (2018).

The main DSP unit operations are harvesting, cell disruption and extraction. Centrifugation is the most efficient (>95% efficiency) method for harvesting microalgae. However, being very cost intensive, it is not suitable for large scale systems. Flocculation is a low-cost alternative. Cationic chemical flocculants and polymeric flocculants are generally used (Brennan and Owende, 2010), but can negatively affect the toxicity of the biomass and output water (Ryan, 2009). Zhou et al. (2012) reported a novel fungi assisted bioflocculation technique, in which a filamentous fungal spores were added to the algal culture under optimized conditions and the pellets were formed after 2 days that can be harvested by simple filtration. Attached culture can also make harvesting simple (Wang et al., 2017). Conventional disruption methods like bead beating, homogenizers, heating, applying high pressure and chemicals or enzymes for lysis is costly and pose risk of loss of desired multi products in biorefinery concept. Physical disruption by pulsed electric field (PEF) is a promising alternative technology as it is a low-shear technology that operates on low temperature and can aid the extraction of hydrophobic constituents of the biomass (Goettel et al., 2013; 't Lam et al., 2017). In the case of extraction technologies, ionic liquids (ILs) appear to be promising as they are advantageous over conventional solvents. ILs are organic salts that are non-volatile at room temperature. Also, they can be used for extraction of hydrophilic proteins. Imidazolium-based ILs have been successfully used for cell disruption for lipid extraction from microalgal biomass (Orr and Rehmann, 2016).

CONCLUDING REMARKS

Microalgae based carbon capture technologies are certainly promising but their successful implementation is still to be realized. Recent advances and breakthroughs in bioprospecting new strains, innovation in culture strategies and process optimization are certainly making us optimistic about the future of microalgal biorefinery. But, the prospects of successful commercial deployment lie in unsophisticated innovations in DSP, particularly harvesting, cell disruption and extraction, which can actually cut down the costs at a biorefinery level, along with process integration. Lastly, the vast data gathered through omics and labeling analysis needs to critically and holistically studied to gain in depth knowledge of the microalgal CCM, biosynthetic pathways and stress mediated responses ensuing the creation avant-garde strains and metabolic circuits via genetic/metabolic engineering approaches, that can revolutionize the whole microalgal biorefinery concept.

AUTHOR CONTRIBUTIONS

Both authors contributed equally toward the preparation of the manuscript. DD was involved in detailing and overall preparation of the manuscript. JS collected the available literature and drafted the manuscript.

FUNDING

This work was supported by grants from Department of Science and Technology (PDF/2015/001067), Government of India (GoI), New Delhi, India.

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Conflict of Interest Statement: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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