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RESOURCE RECOVERY FROM WASTEWATER, SOLID WASTE AND WASTE GAS: ENGINEERING AND MANAGEMENT ASPECTS

Enhancement of biofuel production by microalgae using cement flue gas as substrate

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Abstract

The cement industry generates a substantial amount of gaseous pollutants that cannot be treated efficiently and economically using standard techniques. Microalgae, a promising bioremediation and biodegradation agent used as feedstock for biofuel production, can be used for the biotreatment of cement flue gas. In specific, components of cement flue gas such as carbon dioxide, nitrogen, and sulfur oxides are shown to serve as nutrients for microalgae. Microalgae also have the capacity to sequestrate heavy metals present in cement kiln dust, adding further benefits. This work provides an extensive overview of multiple approaches taken in the inclusion of microalgae biofuel production in the cement sector. In addition, factors influencing the production of microalgal biomass are also described in such an integrated plant. In addition, process limitations such as the adverse impact of flue gas on medium pH, exhaust gas toxicity, and efficient delivery of carbon dioxide to media are also discussed. Finally, the article concludes by proposing the future potential for incorporating the microalgae biofuel plant into the cement sector.

Keywords Microalgae · Biodegradation · Carbon dioxide mitigation · Carbon capture · Nitrogen oxides · Sulfur Oxides · Cement industry

Introduction

The global infrastructure is mainly dependent on cement. As a result, cement is the most consumed resource in the world, second only to water (Pereira 2012). The global market size of cement was estimated to be 355.6 billion in 2016, and the expected compound annual growth rate from 2017 to 2025 is 7.8% (Miller et al. 2018). Although the cement industry has benefited humans, it has adversely affected the environment (Lara-Gil et al. 2016). The primary pollutant in the cement industry is flue gas, which is mainly generated by raw material calcination and fuel combustion process (Mahasenan et al. 2003). Carbon dioxide, one of the elements of flue gas, contributes to global warming. The cement sector contributes to nearly 6% of the overall carbon dioxide emission in the world (Van Oss and Padovani 2003).

To comprehend flue gas mitigation, the manufacturing method engaged in a typical cement plant is outlined in the following section. Depending on the nature of feed material used in a rotary kiln, the cement production is classified into wet, dry, or semi-dry processes. The newly installed plants mainly use dry feed material, while older plants use both wet and semi-dry feed materials (Schneider et al. 2011). The most widely produced cement variety is Portland cement, in which limestone and aluminosilicates are used as the raw materials. In addition, sand, bauxite, lime waste, aluminate, silicate, and iron are used as raw material substitutes as well as corrective materials in the feed. First, the raw materials are pulverized in a raw mill and dried using hot gas. The

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electrostatic precipitator or fabric filter collects the raw meal and is then fed into a kiln for calcination. The typical condition of the kiln is described below: temperature is maintained around 1450–2000 °C, the oxygen concentration is 2–4% of air, and the heating time is 30–60 min (Huntzinger and Eatmon 2009). The clinker is the end product obtained from rotary kiln. Finally, size reduction is achieved by grinding clinker with gypsum in order to obtain Portland cement. Apart from Portland cement, cement can also be blended with materials such as fly ash, blast furnace slag, and pozzolans (Chen et al. 2010).

A typical cement kiln emits flue gas composed mainly of NOx, SOx, CO_2, and dust (Table 1). The main compound of nitrogen and sulfur found in the flue gas is nitric monoxide and sulfur dioxide respectively (Van Oss and Padovani 2003). Compared to the coal-fired power plant and waste incinerator, cement flue gas includes higher concentrations of water and carbon dioxide but reduced levels of oxygen and HCl (Yen et al. 2015). The carbon dioxide content in flue gas depends on the type of raw material, fuel, combustion process, and cleaning technique used in a particular industry. The use of natural gas as fuel generates 5–6% carbon dioxide while coal produces nearly 10–15% carbon dioxide during the combustion process (Thomas et al. 2016). In the oxy-combustion method, where significant amounts of oxygen are used, flue gas with a concentration of carbon dioxide varying from 75 to 90% can be generated (Kanniche et al. 2010).

The cement industry has made numerous attempts to tackle the carbon dioxide mitigation issue. A broad range of carbon capture and storage techniques have been used to date (Leung et al. 2014). In addition, another potential route is algae-dependent biological mitigation (Lam et al. 2012). The absorption of industrial flue gas by microalgae was first proposed in 1978 (Wodzinski and Alexander 1978). Microalgae are carbon fixing photosynthetic organisms which are considered a potential biodegradation agent and a source of next-generation biofuels (Li et al. 2008; Nagappan et al. 2019a; Sumprasit et al. 2017). This is mainly attributed to microalgal rapid growth rates, higher lipid content, and the capability to grow in wastewater (Khataee et al. 2013; Maza-Márquez et al. 2014). In particular, the photosynthetic machinery in microalgae is very efficient, resulting in 10–50 times higher growth rate than conventional fuel crops (Brennan and Owende 2010; Wu et al. 2013). Apart from biofuel, microalgae produces a wide range of products including pigments, cosmetic, functional foods, aquaculture, and animal feed (Chew et al. 2017; González-Fernández et al. 2016; Nagappan et al. 2019b; Nagappan and Verma 2018; Neumann et al. 2015; Park et al. 2013; Perazzoli et al. 2016). Microalgae cultivated in open raceway ponds generate higher biomass at a cheaper cost, and moreover, suitable extraction techniques can further reduce production cost (Nagappan et al. 2019c; Nagappan and Verma 2016a, b). However, neither microalgal biofuel production nor its utilization for waste biomitigation has been commercialized so far. This is primarily due to higher production cost due to harvesting and upstream and downstream processes (Hamid et al. 2014; Hu et al. 2008; Lananan et al. 2016). Moreover, a rapidly growing species with higher carbon dioxide fixation rate is yet to be identified. A possible solution to overcome the abovementioned problem is coupling cement flue gas with microalgal cultivation. This method can provide microalgae with nutrients acquired through industrial plants, thereby lowering upstream expenses. In addition, the efficient aeration strategy, increased solubility of flue gas, selection of proper bioreactor, reduction of cement flue gas toxicity, and moderation of pH are some of the methods that can be implemented to increase the economic viability of microalgal biomass production.

Factors affecting the mitigation of cement flue gas by microalgae

Some of the factors affecting the microalgae-based treatment of cement flue gas are carbon dioxide tolerance and fixation

Table 1 Typical flue gas compositions in coal-fired boiler, waste incinerator, and cement kiln

Flue gas component	Pulverized coal-fired boiler	Waste incinerator	Cement kiln
O_2 (vol%)	4–6	6–15	2–4
CO_2 (vol%)	10–16	5–14	14–33
H_2O (vol%)	5–12	10–18	5–35
CO (ppmv)	10–100	10–100	600–2600
NO (ppmv)	100–1000	100–1000	475–1900
NO_2 (ppmv)	5–50	5–50	25–100
N_2O (ppmv)	< 1–5	< 1	< 1
SO_2 (ppmv)	100–2000	100–300	10–2500
SO_3 (ppmv)	10–40	–	0–30
HCl (ppmv)	1–100	400–1000	1–25
Flue gas temperature (°C)	135–180	180–230	150–230

rate of microalgae, biomass density, flue gas feeding strategies, flue gas flow rate, flue gas solubility, and flue gas temperature (Du et al. 2019; Lara-Gil et al. 2014; Li et al. 2016; Nagase et al. 1997; Pires et al. 2012; Vuppaladadiyam et al. 2018; Yen et al. 2015). Moreover, the carbon dioxide utilization by microalgae is also affected by the type and concentration of nutrients; pH; salinity; dissolved oxygen concentration; the presence of growth inhibitors; and environmental parameters including light intensity, photoperiod, and temperature.

Species characteristics

The selection of microalgal species is an important factor for effective cement flue gas biomitigation, waste biodegradation, and biofuel production (Aslam et al. 2018; Griffiths and Harrison 2009; Klein et al. 2018; Sakai et al. 1995; Yue and Chen 2005). Microalgal species demonstrating a higher tolerance limit toward flue gas components including NOx, SOx, CO_2 and heavy metal positively influences the efficiency of cement flue gas biotreatment and biofuel production (Du et al. 2019; Radmann et al. 2011; Vuppaladadiyam et al. 2018; Yen et al. 2015). Moreover, microalgae that thrive in acidic conditions are more productive in cement flue gas treatment (Varshney et al. 2015).

Several studies have been conducted to identify species with high tolerance to gases such as NOx and SOx (Tables 2 and 3). For example, in a study, *Chlorella pyrenoidosa* XQ-20044 was shown to have a high tolerance to SO_3^{2-} (1600 ppm) and NO^{2-} (368 ppm) (Du et al. 2019). Another study showed that although *Desmodesmus abundans* and *Scenedesmus* sp. UTEX1589 had high nitrite and sulfite tolerance limits (1067 and 254 ppm, respectively), the bisulfite tolerance limit was only 39 ppm (Lara-Gil et al. 2014). The low bisulfite tolerance limit was mainly attributed to the acidic pH of the medium due to flue gas sparging. However, the same study increased the tolerance limit of bisulfite from 50 to 200 ppm using cement kiln dust as a pH buffer. In a screening study, ten different microalgal strains were evaluated for the removal of NOx and SOx species from industrial flue gas (Negoro et al. 1991). Results showed that growth inhibition

Table 2 Comparison of various strains of microalgae related to SOx tolerance

Microalgae	SOx (ppm)			Reference
	SO_2	SO_3^2 –	HSO^3 –	
Chlorella pyrenoidosa XQ-20044	–	1600	–	(Du et al. 2019)
Chlorella sp. XQ-20044	–	1600	–	(Liang et al. 2014)
Botryococcus braunii	–	80	81	(Yang et al. 2004a)
Chlorella fusca LEB 111	400		–	(Duarte et al. 2016)
Mixed microalgal consortia	–	277.2	–	(Aslam et al. 2017)
Nannochloris sp.	50	–	–	(Cuellar-Bermudez et al. 2015)
Nannochloropsis sp.	50	–	–	(Yen et al. 2015)
Chlorella sp.	60	–	–	(Yen et al. 2015)
Desmodesmus abundans	–	254	200	(Lara-Gil et al. 2014)
Scenedesmus sp. UTEX1589	–	254	200	(Lara-Gil et al. 2014)
Nannochloris sp.	–	50	–	(Lara-Gil et al. 2014)
Phaeodactylum tricornutum	–	50	–	(Lara-Gil et al. 2014)
Nannochloropsis sp.	–	50	–	(Lara-Gil et al. 2014)
Scenedesmus obliquus LEB22	–	60	–	(Radmann et al. 2011)
Synechococcus nidulans LEB25	–	60	–	(Radmann et al. 2011)
Chlorella vulgaris LEB106	–	60	–	(Radmann et al. 2011)
Spirulina sp. LEB18	–	60	—	(Radmann et al. 2011)
Chlorella sp. MTF-7 TFRIa	–	90	–	(Chiu et al. 2011)
Chlorella sp. T-1	–	10	–	(Maeda et al. 1995)
Chlorella sp. KR-1	–	250	–	(Lee et al. 2002)
Monoraphidium minutum	–	200	–	(Zeiler et al. 1995)
41 Hawaii fresh/seawater isolates and 13 collection strains	–	3504	–	(Olaizola 2003)

Table 3 Comparison of various strains of microalgae related to NOx tolerance

Microalgae	NOx (ppm)	Reference
41 Hawaii fresh/seawater isolates and 13 collection strains	328	(Olaizola 2003)
Botryococcus braunii	230	(Yang et al. 2004b)
Chlorella sp. KR-1	300	(Lee et al. 2002)
Chlorella sp. MTF-7TFRIa	80	(Chiu et al. 2011)
Chlorella sp. T-1	150	(Maeda et al. 1995)
Chlorella vulgaris LEB106	100	(Radmann et al. 2011)
Chlorella fusca LEB 111	400	(Duarte et al. 2016)
Chlorella sp.	60	(Cuellar-Bermudez et al. 2015)
Chlorella sp. C2	4060	(Li et al. 2016)
Desmodesmus abundans	1067	(Lara-Gil et al. 2014, Lara-Gil et al. 2016)
Marine strain NOA-113	300	(Yoshihara et al. 1996)
Mixed microalgal consortia	209.2	(Aslam et al. 2017)
Monoraphidium minutum	150	(Zeiler et al. 1995)
Nannochloropsis sp.	300	(Lara-Gil et al. 2014, Lara-Gil et al. 2016)
Scenedesmus obliquus LEB22	100	(Radmann et al. 2011)
Scenedesmus sp. NIER10060	300	(Jin et al. 2008)
Scenedesmus sp. UTEX1589	1067	(Lara-Gil et al. 2014, Lara-Gil et al. 2016)
Spirulina sp. LEB18	100	(Radmann et al. 2011)
Synechococcus nidulans LEB25	100	(Radmann et al. 2011)
Chlorella pyrenoidosa XQ-20044	368	(Du et al. 2019)

and cell death was observed when SO_2 concentration was increased to 50 and 400 ppm, respectively. Similar to the above studies, the decline in growth at 400 ppm SO_2 concentration was attributed mainly to the change in the pH. In such cases, pH moderation can be an effective solution to increase the tolerance limit of the species. Apart from pH moderation, the species tolerance limit has been demonstrated to increase after the initial acclimation process. In a study, microalgae *Nannochloris* sp. was shown to grow normally under a higher concentration of NOx concentration (300 ppm) after a longer lag period (Negoro et al. 1991). In a study by Liang et al. (2014), *Chlorella* sp. XQ-20044 was shown to exhibit high sulfite tolerance (1600 ppm). The high sulfite tolerance was achieved by maintaining the conditions that rapidly transform sulfite to sulfate. The parameters that influenced such rapid conversion of sulfite to sulfate included temperature (35 °C), sodium bicarbonate concentration (6 g L^{-1}), light intensity (300 μmol m^{-2} s^{-1}), cell concentration (0.8 optical density), and pH (9–10). Thus, the above studies highlight the importance of using robust strains and the manipulation of growth parameters for effective flue gas treatment.

Extremophiles that display tolerance to stressful conditions are efficient in the treatment of cement flue gas. The supplementation of flue gas with high carbon dioxide content decreases the pH level of the culture medium, deterring cell growth. Moreover, the delivery of hot flue gas raises the culture medium temperature, inhibiting the growth of mesophilic microalgae. The research was able to tackle the above issue by identifying acidophilic and thermotolerant *Cyanidium caldarium* as an effective species for flue gas biomitigation. Similarly, the use of microalgal species that are tolerant to the high concentration of carbon dioxide can overcome the above problem (Table 4a). For instance, it has been reported that microalgae can grow even at a carbon dioxide concentration of 100% in the culture medium (Bhakta et al. 2015; Hanagata et al. 1992; Kassim and Meng 2017). In particular, species like *Chlorella* and *Scenedesmus* were able to adapt to 80 to 100% carbon dioxide (Salih 2011).

Studies had shown that the cultivation of diverse microalgal species increases the biomass production as well as effectively mitigate carbon dioxide present in flue gas (Cardinale et al. 2006; Gross and Cardinale 2007; Worm et al. 2006). This is due to the fact that one among the diverse species can very well tolerate the local conditions and can lead to an overall higher microalgal biomass production (Fox 2005). Moreover, the cultivation of diverse microalgal species can overcome the effect of foreign species invasion as well as cross-contamination due to complementary effect (Corcoran and Boeing 2012). In addition, the number of products synthesized by diverse species is more than monoculture. Also, the diverse microalgal species possess varied light-harvesting mechanisms as well as a wide range of pigments which make

Table 4 Comparison of various microalgae based on (a) carbon dioxide tolerance and (b) carbon dioxide efficiencies using different reactors

(a)

Microalgae	Carbon dioxide tolerance (%)	Reference
Chlorella sp.	50	(Cuellar-Bermudez et al. 2015)
Chlorella sp.	100	(Li et al. 2011)
Chlorella T-1	100	(Varshney et al. 2015)
Chlorococcum littorale	50	(Cuellar-Bermudez et al. 2015)
Cyanobacteria G. membrancea	15	(Mohsenpour and Willoughby 2016)
Euglena gracilis	10	(Cuellar-Bermudez et al. 2015)
Mixed microalgal consortia	11	(Aslam et al. 2017)
Nannochloris sp.	15	(Cuellar-Bermudez et al. 2015)
Nannochloropsis sp.	15	(Cuellar-Bermudez et al. 2015)
Phaeodactylum tricornutum	15	(Cuellar-Bermudez et al. 2015)
Scenedesmus strain K34	100	(Varshney et al. 2015)

(b)

Microalgae	Type of reactor	Carbon dioxide fixation efficiency	Reference
Chlorella sp.	Raceway pond	46	(Ramanan et al. 2010)
Chlorella sp.	Bubble column reactor	50	(Doucha et al. 2005)
Chlorella sp.	Bubble column reactor	58	(Chiu et al. 2008)
Chlorella sp.	Bubble column reactor	27	(Chiu et al. 2008)
Chlorella sp.	Bubble column reactor	20	(Chiu et al. 2008)
Chlorella sp.	Bubble column reactor	16	(Chiu et al. 2008)
Chlorella sp.	Airlift photobioreactor	63	(Chiu et al. 2009b)
Chlorella sp. MTF 15	Bubble column reactor	25	(Kao et al. 2014)
Chlorella sp. MTF 7	Bubble column reactor	60–70	(Jiang et al. 2013)
Chlorella vulgaris	Bubble column reactor	74	(Keffer and Kleinheinz 2002)
Desmodesmus abundans	3-L glass bioreactor with a working volume of 1 L (for simulations)	50	(Lara-Gil et al. 2014).
Scenedesmus dimorphus	Bubble column reactor	75.6	(Praveenkumar et al. 2014)
Scenedesmus Obliquus WUST4	Airlift column	67	(Li et al. 2011)
Scenedesmus sp.	Raceway pond	94	(De Godos et al. 2014)
Spirulina platensis	Tubular reactors	70	(Watanabe and Hall 1995)
Spirulina sp.	Bubble column reactor	37.9	(De Morais and Costa 2007)
Spirulina platensis	Open pond reactors	39	(Pires et al. 2012)

them effectively harness the solar energy (Dubinsky and Stambler 2009). In addition, microalgal species with high lipid and carbohydrate productivity are promising ones for microalgal biodiesel and biogas/bioethanol production, respectively (Chen et al. 2013; Nagappan and Verma 2016b).

Carbon dioxide fixation rate

The carbon dioxide utilization by microalgae is a vital parameter to consider for the integration of microalgae with a cement production facility for biomitigation purposes. Several studies had reported carbon dioxide tolerance of microalgae in the range of 1 to 100% of the total supply (Verma and Srivastava 2018). However, in many cases, the optimal carbon dioxide concentration conducive for microalgal growth is estimated to be only 10–15% (Ishida et al. 2000; Salih 2011). Moreover, the maintenance of the carbon dioxide level is crucial for achieving the desired lipid content in microalgae. It has been suggested that supplying carbon dioxide at lower levels of 5–20% increases the lipid production while a higher concentration of 20% or more leads to decreased lipid production (Chiu et al. 2009a; Yoo et al. 2010). This is probably due to the negative effect of high-concentration carbon dioxide on microalgal growth, subsequently leading to decreased lipid production.

Although carbon dioxide fixation rate is an intrinsic characteristic of a microalga, the carbon dioxide utilization can be improved using a specialized reactor in which the mass transfer of carbon dioxide to liquid is maximum (Doucha et al. 2005; Jiang et al. 2013). Additional factors affecting carbon dioxide fixation by microalgae are environmental and nutritional requirements of algae including light intensity, photoperiod, temperature, pH, carbon dioxide level, and the nitrogen and phosphorus level in the media (Grierson et al. 2013; Li et al. 2009). Table 4b presents the carbon dioxide fixation rate of several microalgae cultivated in different conditions.

Solubility of gas

The flue gas can be directly supplied into the culture medium for microalgae biomitigation. However, the flue gas solubility, particularly that of carbon dioxide in the culture medium, is estimated to be very low (Lam et al. 2012). In most cases, the difference in the actual solubility of carbon dioxide and the amount of carbon dioxide needed for ideal growth is significant, influencing the process of microalgal biomitigation. In the case of open ponds, lesser solubility of flue gas leads to excess loss into the atmosphere (De Godos et al. 2014). But the addition of captured carbon dioxide such as bicarbonates into medium rather than supplying flue gas directly can solve the above problem (Chi et al. 2011). However, the conversion of carbon dioxide in the flue to dissolve inorganic carbon can be a disadvantage since a higher amount of chemicals such as sodium hydroxide and potassium hydroxide will be required, leading to additional cost as well as problems associated with wastewater generation. In this case, the generation of value-added products along with biofuel and biodegradation purposes can justify the additional cost.

Flue gas toxicity

The cement kiln dust and gas are two major components of cement flue gas (Talec et al. 2013). The dust is usually composed of various residues from clinker, un-reacted and partially reacted raw material remaining after the combustion process. The cement kiln dust containing soot, trace metals, Al_2O_3, and SiO_2 are reported to inhibit microbial growth (Jiang et al. 2009; Matsumoto et al. 1997; Wang et al. 2009). Microalgal growth inhibition is also caused by a change in media pH due to supplementation of flue gas having a high concentration of CO_2, NOx, and SOx (Talec et al. 2013).

The gas component mainly contains nitrogen oxides, sulfur oxides, and carbon monoxide which are considered to be harmful to microalgal growth when present beyond a threshold value. The concentration of these toxic compounds depends on the type of raw material, fuel, and technology used in particular cement industry (Van Oss and Padovani 2003). Flue gas contains a significant concentration of NO_x. Nitric oxide and nitrogen dioxide are two predominant NOx gases present in the flue gas (Lara-Gil et al. 2014). These components form photochemical smog when emitted into the atmosphere and also causes acid rain. Although the above species do not contribute to global warming, it has been reported to have a negative impact on human health (Boningari and Smirniotis 2016). While NO_x at higher concentration has an inhibitory effect, the same component at lower concentration can be used for microalgae cultivation.

The SOx is another predominant gas found in the cement flue gas. Sulfur dioxide and sulfur trioxide are dominant species in the flue gas (Boningari and Smirniotis 2016). Even though SOx does not contribute to global warming, the species have a negative impact on the environment, including acid rain formation and destabilization of ozone. Similar to NOx, SOx affects microalgal growth at higher concentration. In a low buffered medium, SO_2 is converted to bisulfite; when consumed by cells, bisulfite is converted to toxic oxygenated gas that damages membrane and pigments, causing cell death (Negoro et al. 1991; Yang et al. 2004a). However, upon aeration, SOx and also NOx are oxidized into sulfate and nitrate and can thus serve as a nutrient for algae. Carbon monoxide is less tolerated by algae, but tolerance limit as high as 3 ppm has been reported in the literature (Doucha et al. 2005).

Another toxic compound for microalgal growth present in cement flue gas is heavy metal. Exposure to heavy metals, even at lower levels, is known to cause damage to several organs. They are likewise named human cancer–causing agents as indicated by the International Agency for Research on Cancer and US Environmental Protection Agency. As a result, strict emission laws have been implemented by several countries. For instance, the emission of mercury in newly constructed and existing cement plant is limited to 4 and 10 $\mu g\ Nm^{-3}$ in the USA by the environmental regulation agency (Zheng et al. 2012). In conclusion, an effective treatment is required to overcome the flue gas toxicity and further use in the cultivation of microalgae.

Flue gas biomitigation strategies

Flue gas aeration and feeding strategies

One of the problems associated with flue gas supplementation into the microalgal culture medium is the lower mass transfer rate of flue gases. Carbon dioxide especially has a higher mass transfer coefficient, causing slow dissolution in the culture medium (De Godos et al. 2014). Moreover, in the case of open ponds, easy loss of carbon dioxide to the atmosphere occurs. Therefore, in order to increase the flue gas, including carbon dioxide mass transfer rate, techniques including high aeration rate can be adopted (Grobbelaar 1994). This not only increases the carbon dioxide gas dissolution but also enhances the removal of oxygen in the culture medium generated by photosynthesis. The presence of oxygen causes growth inhibition of microalgae (Sousa et al. 2012). Another method that has been reported is the installation of pumps with a novel design. For example, the use of ÒsumpÓ device—a pump with an innovative design—along with intermittent flue gas supply was employed, leading to an increase in contact time between the flue gas and liquid phase (Pawlowski et al. 2014). In another study, careful positioning of the sump, maintenance of fixed pH of 8, and reducing the flow rate of flue gas to 0.005 vvm had led to carbon dioxide removal efficiency of 94% in flue gas (De Godos et al. 2014). In addition to increased gas dissolution, a higher rate of aeration improves the homogenous distribution of nutrients, algal biomass, and temperature in the medium.

Gas feeding strategies have been successfully optimized in several studies to increase the efficiency of flue gas treatment by microalgae. Rather than supplying flue gas continuously, an intermittent supply was proven beneficial for growth and carbon dioxide fixation of a *Chlorella* species (Chiu et al. 2011). Furthermore, a combination of intermittent flue gas supply with supplementation of calcium carbonate in the medium for pH regulation was shown to improve the efficiency of carbon dioxide fixation and overcome the flue gas toxicity (Jiang et al. 2013). Another study showed that the dilution of flue gas reduced the adverse effects of high concentration of carbon dioxide and high temperature typically associated with flue gas on a microalga, *Chlorella* sp. (Kumar et al. 2014). A unique strategy was adopted in a study where supplying flue gas in a mixotrophic media during the dark period resulted in highest biomass and biodiesel productivity of 561 and 168 mg L^{-1} day^{-1}, respectively (Praveenkumar et al. 2014). Control of flow rate was also shown to be an effective method to increase both microalgal biomass production and carbon dioxide fixation rate. This was demonstrated in an earlier study where a lower flow rate favored higher carbon dioxide mitigation (Li et al. 2011).

The continuous flue gas aeration rapidly changes the medium pH, adversely affecting microalgae. Addressing this issue, a study involving *D. abundans* showed that intermittent supply of flue gas was better than the continuous supply in overcoming pH toxicity (Lara-Gil et al. 2014). However, in this case, the growth rate was maintained for only 60 h after which 60–70% decrease was observed. Similarly, a study showed that an intermittent supply of flue gas with 400 ppm SO_2 resulted in an enhanced growth of *Scenedesmus dimorphus* with a cell density of 3.2 g L^{-1} (Jiang et al. 2012). In another study, the aeration rate was moderated using an electromagnetic valve, which was turned on or off by a pH controller (Du et al. 2019). This setup overcame the toxicity of the flue gas component by maintaining pH in the range of 7 to 8. Effective aeration of flue gas enables SOx, NOx, CO_2, and trace metals to serve as nutrients for microalgae, increasing the biomitigation potential.

Gas solubility enhancers

In order to increase the solubility of the gas in a liquid medium, substances including amines, carbonates, bicarbonates, and sodium hydroxide can be added (Fernández et al. 2012; Kim et al. 2011). A study had shown that the addition of monoethanolamine in culture medium increased both the carbon dioxide fixation efficiency and biomass productivity (Choi et al. 2012). In contrast, another study displayed that the addition of methanolamine and 2-amino-2-methyl-1-propanol in culture medium resulted in lesser biomass production compared to the control (Kim et al. 2013). This was attributed to increased carbamate production as intermediate in alkanolamines supplied culture medium; higher carbamate levels adversely affect microalgal growth (Fernández et al. 2012). The NO is a flue gas component which dissolves poorly in the culture medium and is considered a rate-limiting step in the microalgal reactor system (Jin et al. 2005; Nagase et al. 1997). Addressing the above problem, a study showed that NO solubility was increased and 85% NO removal efficiency was achieved with the addition of 5 mM Fe–EDTA using *Scenedesmus* species (Santiago et al. 2010). The removal of NO also depends on the concentration of oxygen in flue gas. In another study, the NO removal efficiency was improved by nearly 96% when the proportion of air in the flue gas was increased to 85% (Nagase et al. 1998). Therefore, the addition of supplements at an optimum concentration increases the solubility of flue gas and thus leads to efficient flue gas biomitigation.

pH moderation

Microalgae can tolerate flue gas toxicity by the maintenance of optimal pH. The reduction in the pH levels due to flue gas can be moderated using alkalis such as $CaCO_3$ and NaOH, as well as buffers. The concentration of alkali required to neutralize the acidic medium sparged by flue gas will depend on the concentration of flue gas component including CO2, NOx, and SOx. Moreover, strategies including dilution or dosing of flue gas (Jiang et al. 2013; Lee et al. 2002; Maeda et al. 1995), aeration (Chiu et al. 2011; He et al. 2012; Lee et al. 2002), high initial biomass density (Jin et al. 2008), and acclimatization of microalgae to acidity (Jiang et al. 2012) are additional options available for moderating the medium pH. Besides, a high concentration of CO_2 can also be used to regulate the pH. The CO_2 dissolves in water to form carbonic acid and subsequently dissociates into hydrogen ion and bicarbonate (Gao et al. 1991). Therefore, concentration of hydrogen ion increases in the media and could thus be used for pH regulation. However, the choice of strategy should be in accordance with the cost of the product. A low-value product such as biofuel requires a low-cost strategy to maintain pH.

The decrease in the pH can also be neutralized using alkali. This was demonstrated in a study where sodium hydroxide was used for pH neutralization leading to an increase in the tolerance limit of SOx from 50 to 400 ppm (Matsumoto et al. 1997). A study demonstrated that cement flue gas with high carbon dioxide concentration (12–15%) was well tolerated by several microalgal species including *Skeletonema marinoi*, *Tetraselmis* sp., and other natural isolates from the Baltic sea (Olofsson et al. 2015). The reason for tolerance as well as enhance biomass productivity was attributed to the salts present in the seawater which acted as a natural buffer, preventing the pH increase due to the carbon dioxide present in the flue gas.

A low-cost technique to moderate pH is the use of cement kiln dust (CKD) in media. Studies have shown that CKD addition had benefited microalgal growth (Lara-Gil et al. 2014; Lara-Gil et al. 2016). Typical composition of CKD is CaO, SiO_2, K_2O, MgO, Na_2O, SO_3, Fe_2O_3, and heavy metals (Borkenstein et al. 2011; Talec et al. 2013). CKD is produced around 15–20% per ton of cement generated through the calcinations process. In general, CKD is disposed of in landfills and only limited use has been demonstrated so far. In this regard, CKD can be used as buffer and also offers additional advantages. It is highly water-soluble and contains valuable trace metal essential for growth (Lara-Gil et al. 2014).

Heavy metal bioremediation

The heavy metal could be removed from the flue gas using conventional methods including physico-chemical cleaning of raw material and fuel, combined technique of oxidation and wet scrubbing, sorbent injection before fabric filtration, and use of sorbent bed (Liu et al. 2017; Yang et al. 2016; Yu et al. 2016). However, several drawbacks are associated with conventional methods. For example, the high moisture content in cement flue gas reduces the efficiency of the sorbent injection method (Chen et al. 1999). In the case of physicochemical methods, the cost involved in cleaning the cement flue gas decreases the viability of the treatment process (Stathi et al. 2007).

An alternative for conventional heavy metal removal process in flue gas treatment is the use of microalgae. Several studies have demonstrated that microalgae can be used for effective bioremediation of heavy metal (Kumar et al. 2015; Zeraatkar et al. 2016). The reason for the efficiency is attributed to the higher metal binding capacity of cell walls (Travieso et al. 1999). The cell wall of microalgae, which is made up of polysaccharide, lipid, and proteins, has charged functional groups that could effectively sequester heavy metals. Factors including the concentration of heavy metal, type of microalgal species, biomass density of microalgae, pH of the medium, and other physiochemical parameters strongly influence the bioremediation efficiency of heavy metals (Kumar et al. 2015). Studies have shown that microalgae are advantageous over conventional methods such as scrubber and activated carbon in the removal of heavy metal from flue gases (Chen et al. 1999; Rio et al. 2007). The biosorption of heavy metal by microalgae, although promising, has some bottlenecks which require further research. One of the problems with the biomass comprising heavy metals is that it cannot be used in the cosmetics, animal feed, aquaculture, and functional foods sectors. Moreover, these biomasses cannot be disposed of in landfills directly. However, biofuel can be produced from the heavy metal–containing microalgal biomass.

Biomass density

Another strategy adopted to improve microalgal flue gas treatment is the use of high biomass density. This overcomes the problem of toxicity and high temperature associated with the flue gas (Chiu et al. 2008; Chiu et al. 2011). However, the growth of microalgae in medium containing high cell density can be severely limited due to the fact of self-shading of cells (Hewes 2015). The light penetration is difficult in self-shading conditions, and alternative strategies have to be adopted to overcome the problem. A possible solution is truncating the antenna size which is responsible for controlling the light-harvesting process in photosynthesis using genetic engineering methods (Ort et al. 2011). Another solution is decreasing the chlorophyll content in microalgae so that light penetration is enhanced, possibly leading to increased biomass production.

Choice of the reactor

Microalgal treatment of cement flue gas involves cultivation in either open or closed system. The main advantage of an open system is the low cost; however, it has problems including contamination and foreign species invasion (Whitton et al. 2015). Moreover, open pond conditions cannot be controlled as conditions depend on local weather pattern and the geographical location of the cultivation site (Narala et al. 2016). However, the selection of species that has a higher tolerance to stress condition and that adapt to local environmental condition is critical for the open pond's commercial viability. For example, species including *Dunaliella* having high salinity tolerance, *Spirulina* having high alkaline tolerance, and *Chlorella* having high nutrient tolerance have been successfully cultivated in open ponds (Mobin and Alam 2017; Odjadjare et al. 2017).

The photobioreactor, in contrast, has culture conditions that can be easily controlled and also does not suffer from the problem of severe contamination (Wang et al. 2013). Various configurations of photobioreactor exist, including vertical column reactors, also called as bubble columns or airlift, tubular reactors, and flat-plate reactors. Due to low hydrodynamic shear, the vertical column reactor has great potential for biotechnological applications (Carvalho et al. 2006). The photobioreactors can be operated in both outdoor and indoor conditions. In the case of an outdoor setup, it can be configured in different positions allowing maximum harvest of sunlight. The flat-plate reactors produce more biomass than other reactors (Endres et al. 2018). Advantages of flat-plate reactor are that it consumes less power and has a higher mass transfer.

So far, different photobioreactors with sizes ranging from 8 to 400 L have been successfully demonstrated to treat flue gas and support the growth of several species including *Chlorella*, *Scenedesmus*, *Spirulina*, *Cyanidium caldarium*, *Galdieria partita*, *Chlorococcum littorale*, *Prasinococcus capsulatus*, *Cyanidioschyzon merolae*, *Thermosynechococcus*, *Euglena gracilis*, and *Nannochloropsis* (Chen et al. 2012; Chiu et al. 2008; Choi et al. 2018; Doucha et al. 2005; Kim et al. 2011; Narala et al. 2016; Ouyang et al. 2015; Vergara et al. 2016; Yadav et al. 2015; Yoshihara et al. 1996). Studies have shown that maximum carbon dioxide fixation rate can be achieved using closed photobioreactors including bubble column and airlift bioreactor (Chiu et al. 2011; Douskova et al. 2009; Duarte et al. 2016). In comparison to open ponds, biomass productivity is higher in a photobioreactor (Yen et al. 2015). However, as mentioned earlier, the higher capital and operational cost remains a challenge for this class of system. In addition to capital cost, the treatment efficiency and flue gas pretreatment step determine the economic viability of the entire process. For example, the counter-flow type airlift reactor had nearly three times higher NO removal efficiency than the bubble column reactor (Nagase et al. 1998). In this context, the production cost of algal biofuel will be justifiable if biorefinery products could be additionally generated and waste could be biodegraded.

Integration of carbon capture and storage technologies with microalgae biomass production

The carbon dioxide emitted from the cement industry is captured using various techniques including physical adsorption, chemical absorption, oxy-fuel technology, gas separation membrane, wet method, dry method, and cryogenic distillation (Lam et al. 2012; Leung et al. 2014; Thomas et al. 2016). The captured carbon dioxide can be compressed to liquid, which can further be transported or simply stored (McCoy and Rubin 2008; Svensson et al. 2004). Mineralization and ocean and geological storage are some options that are available for storing the captured carbon. These techniques are together known as carbon capture and storage (CCS) technologies. The CCS technologies facilitate the reduction of carbon footprint and thus contribute toward the prevention of global warming. However, CCS technology causes an economical burden to the cement industry due to high capital and operational cost (Stewart and Hessami 2005). Furthermore, no valuable products are obtained using CCS technology. In this regard, a combination of CCS technologies with microalgae cultivation can be beneficial.

Various CCS technologies have been reported for the treatment of flue gas containing carbon dioxide. One of the recent additions is oxy-fuel technology which involves supplementation of pure oxygen instead of air for combustion (Schneider et al. 2011). Supply of oxygen rather than air facilitates flue gas emission with a higher concentration of carbon dioxide that could be easily recovered. However, one of the main challenges associated with oxy-fuel technology is the prevention of air intrusion during combustion. Another method is mineralization which is achieved by the mixing of flue gas containing carbon dioxide with brine containing calcium and magnesium resulting in the production of carbonate (Matter et al. 2016). The efficiency of the abovementioned carbon capture process has been estimated to be 70–90%. Wet methods including the use of mopfan, wet electrostatic precipitator, and wet scrubbers, as well as dry methods including cyclone separator and dry electrostatic precipitator, have been employed and well documented for the mitigation of carbon dioxide (Singh and Shukla 2014). The use of solid adsorbents for carbon dioxide sequestration has been well documented in the literature (Lee and Park 2015). So far, materials including zeolite, activated carbon, hydrotalcites, calcium oxide, carbon nanotubes, and molecular sieves have been tested for the physical adsorption of carbon dioxide. In addition, various membranes have been reported for carbon dioxide mitigation. Polymeric membranes including polysulfone, and polyamide, and inorganic membranes such as palladium and silver alloys, alumina, silicon carbide, zirconia, and glass, have been shown to separate carbon dioxide gas (Powell and Qiao 2006; Yang et al. 2008). However, further research is required to overcome the limitations of membrane technology. This includes corrosion of membrane, lesser stability, and lower selectivity and permeability of gaseous molecules.

Solvents including monoethanolamine, polymethylmethacrylate, tetraethylenepentamine, and potassium carbonate can also be used for carbon dioxide mitigation (Chi et al. 2013; Kim et al. 2013). A study showed that the addition of monoethanolamine up to 100 mg L^{-1} in culture medium significantly improved carbon dioxide fixation by *Scenedesmus dimorphus* (Sun et al. 2015). Although the solvent absorption technique is easy to implement, there are certain limitations. For instance, the complete desorption of carbon dioxide from solvents including monoethanolamine, polymethylmethacrylate, and tetraethylenepentamine requires high temperature and pressures in the range of 120–150 °C and 1–1.5 MPa, respectively, thus increasing the energy demand (Brilman et al. 2013; Lam et al. 2012; Yeh et al. 2005). A cost-effective method of carbon dioxide desorption from the solvent has to be developed for the economical viable treatment of flue gas using microalgae. However, drawbacks of the sole implementation of CCS technologies including lack of production of valuable products and high cost can be solved by integration with microalgal biomass facility. Therefore, the integrated CCS–microalgae biomass production is a potential biomitigation method for flue gas treatment.

Strain improvement

Strain improvement is an effective tool to produce strains with higher carbon dioxide biomitigation potential. Strain improvement can be performed using two strategies. One is directed evolution involving random mutation and subsequent selection of species with desired phenotype, whereas the rationale screening approach involves specific planned mutation and species selection (Banerjee et al. 2016; Bassalo et al. 2016). Although the latter technique saves cost and time, the unavailability of species-related genetic information deters its implementation. However, in well-documented species, it had been readily adopted by several studies (De Bhowmick et al. 2015). Figure 1 summarizes various microalgal strain improvement techniques. Various characteristics have been successfully shown to be improved by both directed evolution and rationale approaches. In the study by Cheng et al. (2019), *Chlorella* sp. Cv mutant, which was selected using directed evolution experiment, displayed increased carbon fixation rate of 1.2 g L^{-1} day^{-1}, biomass production of 2.7 g L^{-1}, and carbohydrate content of 69% compared to wild strain. The problem of a high temperature of the medium caused by the direct injection of hot flue gas was also solved by the production of thermotolerant mutant strains that were also able to fix CO_2. For instance, mutant strains of *Chlorella*, cyanobacteria *Chlorogloeopsis* sp., and *Chlorella sorokiniana* were selected and demonstrated for growth at high temperatures of 40, 50, and 42 °C, respectively (Ong et al. 2010; Ono and Cuello 2007; Sakai et al. 1995). Moreover, mutant strains that are tolerable to high carbon dioxide concentration, low pH condition, heavy metals etc. have been produced by various strain improvement techniques (Ibuot et al. 2017). To conclude, strain improvement is rapidly evolving and is very much an essential strategy for successful flue gas treatment in industrial setups.

Perspective

Current cement flue gas treatment methods significantly increase the operation cost. Similarly, biomass production from microalgae is not economically viable due to the high production cost. In this scenario, microalgae cultivation, if combined with CCS technologies, can be a possible solution to the abovementioned problem. Moreover, an integrated facility will save valuable land space. For example, most cement plants that already exist find difficulty in allocating a vast area for the construction of a microalgal plant. In such cases, carbon could be captured and

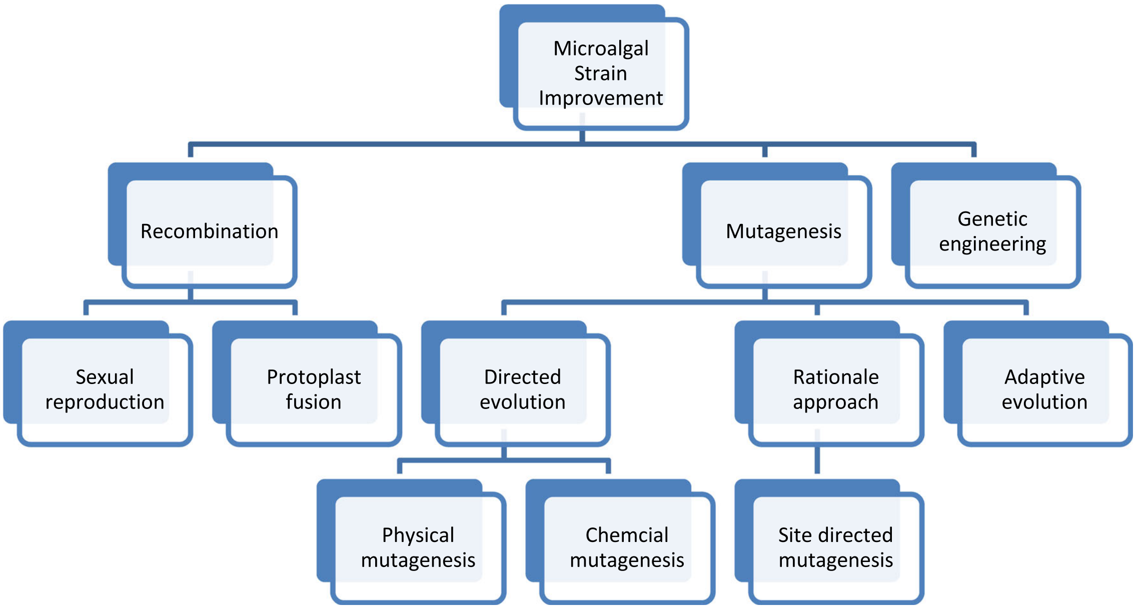

Fig. 1 Summarization of various microalgal strain improvement techniques

subsequently transported to the site of algal cultivation using CCS technologies. The cost-effectiveness of the above-integrated approach will ultimately depend on the type of CCS technique used. For example, chemical absorbent can be chosen over physical adsorbent since carbon could be delivered directly into the medium without any pre-treatment in the former case, whereas desorption of carbon from physical adsorbent involves high energy, leading to increased operating cost.

Similar to the integration of CCS with the algal plant, the industrial treatment of flue gas can also be combined with microalgae cultivation. In general, the industrial treatment of flue gas includes electrostatic precipitation, desulfurization of the gas, heat exchange, etc. For instance, SOx can be removed by chemical desulfurization process. However, removal of NOx is a potential problem since it has a lower solubility in the liquid phase. In this case, the introduction of microalgae can solve the problem; industrial treatment also reduces the toxicity of flue gases, helping microalgal growth. The supply of flue gas and wastewater to microalgae cultivation is another potential option to simultaneously reduce environmental problems and produce biomass at low cost. Wastewater generated from various industries, agricultural runoff, municipal waste stream, etc. can be used for this purpose. Thus, the above approaches can lead to flue gas biomitigation, and waste bioremediation, as well as biofuel production.

The economics of cement flue gas biomitigation using microalgae depends on the concentration of carbon dioxide in the flue gas. More dilute carbon dioxide source consumes high energy due to increased time of sparging in the cultivation media. Moreover, if the carbon dioxide–emitting plant is not at close proximity to the algal plant, the transportation cost also increases (Davis et al. 2011). In addition, concentration and compression of flue gas for transportation and distribution further increases the operational cost. For instance, it is estimated that the cost of transportation of low-pressure flue gas over 100 km through the pipeline is $86.5 per metric ton of CO_2. However, in such cases, the transportation of compressed flue gas rather than low-pressure flue gas has shown to reduce the operation cost. Further, the economics of the flue gas mitigation by microalgae is also determined by the carbon dioxide utilization efficiency of microalgae. The species with efficient carbon capture biochemical mechanism have proven to improve the process economics of such treatment plants (Somers and Quinn 2019).

Many countries are realizing the imminent threat of global warming due to industrialization. For example, India had emitted 2.8 gigatons of carbon dioxide equivalent in 2013 (Dhankar et al. 2017). The total emission of carbon dioxide equivalent increased annually at a rate of 5.57% for the past 10 years. At this rate, India realizing the vulnerability to climate change has introduced the flue-gas mitigation framework in the current and future plants. Apart from climate change, India is also affected by poor air quality. India has joined the Climate and Clean Air Coalition recently and set the reduction target for fine particulate air pollution by 20 to 30% by 2024. Likewise, the government of India has also set a target year to replace fossil fuel completely with renewable energy by 2030. In such a scenario, carbon capture by microalgae could effectively meet the policy requirement of India by meeting both strict flue gas emission norms and the renewable energy target.

Many countries' policies, including India, require the manufacturing of biofuels using wasteland (Saravanan et al. 2018). However, popular feed stocks of biofuel including jatropha and sugarcane face various hurdles in wasteland-based cultivation. The biomass productivity in wastelands is very low. Moreover, in the case of sugarcane, the price of molasses, which is used to produce biofuel, has been steadily increasing in recent years. In this scenario, algal fuel has features that can overcome the above obstacles including the utilization of wasteland and waste stream, high biomass productivity, efficient carbon dioxide sequestration, and synthesis of biorefinery products. A techno-economic analysis based on several studies revealed that the supplementation of nutrients from the waste stream and flue gas from industrial plants including the cement industry could reduce the production cost of algal fuel in the range of 35 to 86% (Judd et al. 2017). In summary, close proximity of emission source to algal plant, high concentration of carbon dioxide in flue gas, and algae with high carbon dioxide fixation efficiency can both improve the economic viability of cement flue gas biomitigation using microalgae and help in achieving the emission target set by the government.

Various biofuels can be produced from microalgae. The most prominent fuel is biodiesel due to the higher lipid content of microalgae. The biodiesel has chemical properties similar to regular diesel and could therefore be used directly in engines without any modification (Knothe 2010). Even though microalgal biodiesel has been unsuccessful so far, optimization of stress conditions and generation of genetically altered strains can possibly increase the biodiesel productivity in the future. Although microalgae are well known for biodiesel, other fuels can also be produced from this versatile source. For instance, biohydrogen can be produced using microalgal species such as *Chlamydomonas reinhardtii* through a combination of the aerobic and anaerobic pathway (Kosourov et al. 2002). However, current hydrogen production from microalgae is not feasible and therefore improvements in the solar energy conversion efficiency through genetic alteration can achieve economic viability. Biogas can also be generated from carbohydrate-rich microalgal biomass. The main limitation of microalgal biogas production is the identification of species and conditions related to carbohydrate accumulation. Here again, stress application, bioprocess optimization, and strain improvement techniques can play an important role.

The optimization of reactor conditions as well as adopting strategies to overcome flue gas toxicity increases the viability. Future studies should attempt to improve carbon dioxide retention time since it promotes higher biomass productivity. In addition, novel methods facilitating rapid carbon dioxide mass transfer between gaseous and liquid phase should be developed. For example, strategies including the installment of novel pumps, aeration regime, and control of flow rate can be implemented. Moreover, designing reactors with an efficient light distribution system can further increase microalgal biomass production. Furthermore, the adoption of various cultivation mode including semi-continuous, fed-batch, and continuous, along with genetic engineering of microalgae in future studies, could lead to efficient microalgal cement flue gas biomitigation, waste biodegradation, and biofuel production.

Conclusion

In comparison to other industries, the cement sector emits flue gas with high concentrations of CO_2, NOx, SOx, suspended particulates, and heavy metals, which impacts the environment as well as human beings. Fortunately, the elements of flue gas were found to have a positive impact on the production of microalgae biomass. In addition, several important precursors can be generated from microalgae, which could then be used to produce biofuels, feed livestock, nutraceuticals, and cosmetics. Moreover, the efficient sequestration of heavy metals by microalgae is an added advantage. However, the economic viability of microalgae-based flue gas treatment can be achieved only by the selection of species with high growth rate, increasing the dissolution of flue gases and the development of low-cost processing technologies for the generation of various products including biofuel. In summary, the utilization of microalgae rather than conventional methods for the off-gas treatment in the cement industry serves the dual benefit of reducing the toxicity of flue gas and generation of biofuel and related biorefinery products.

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