Review Article

Flue gas CO_2 supply methods for microalgae utilization: A review

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Abstract: The potential for utilizing flue gas as a carbon source in microalgal cultivation holds great promise. Incorporating flue gas as a carbon source into microalgae culture processes can accelerate the growth rate of microalgae, consequently enhancing the overall economic viability of the integrated process. There are two key sources of flue gas to consider: flue gas from coal-fired power plants, characterized by a CO_2 concentration of 12%–15% w/w, and flue gas from coal chemical processes, boasting a CO_2 concentration of 90%–99% w/w. Additionally, the choice between an open or sealed microalgae culture system can also influence economic efficiency. Thus, there are four distinct microalgal cultivation routes to assess: in-situ open systems, off-situ open systems, in-situ sealed systems, and off-situ sealed systems. The incorporation of flue gas as a carbon source in microalgae cultivation demonstrates significant potential for reducing both environmental impact and costs, rendering it a highly promising and sustainable approach for economically efficient microalgae cultivation. In this review, the in-situ open route is recommended for systems with a high concentration of flue gas CO_2 with the target product of low-margin commodities, while the off-situ sealed route is suitable for systems with low concentration of flue gas CO_2 with the target product of high-value-added products.

Keywords: flue gas; microalgae; biorefinery; photobioreactor; technological process

1. Introduction

The continuous increase in anthropogenic carbon dioxide (CO_2) emissions is widely recognized as one of the primary drivers of global climate change. CO_2 capture technology focuses not only on the sequestration of CO_2 but also on its utilization. The development of this technology enables the continued use of fossil fuels, while maintaining stable greenhouse gas concentrations in the atmosphere[1]. Microalgae, as a biomass with a short growth circle, are widely used in cogeneration, food, chemical industry, and so on, which has attracted significant attention. These organisms have high photosynthetic efficiency, making them ideal for carbon fixation. The cultivation of microalgae consumes external CO_2 and nutrients, which has environmental impacts. Research has identified the CO_2 supplement in microalgae photobioreactor (PBR) systems as the most impactful process, responsible for over 60%–90% of the positive impacts. Flue gas is a waste product that contributes to greenhouse gas emissions and climate change if it is released into the atmosphere[2]. Thus, there have been proposals to use CO_2 emissions gases recovered from flue gas of industrial processes as a source of carbon for microalgae cultivation[3]. This approach has the potential to increase the growth rate of microalgae. Moreover, using flue gas as a carbon source can replace the consumption of commercial carbon sources in the culture process, reducing the associated costs[4]. Studies have shown that using carbon dioxide from flue gas can save up to €0.4 per kg, which represents a significant cost reduction[5]. By using CO_2 emission gases

Received: 1 November 2023 Accepted: 19 December 2023 Available online: 25 December 2023

recovered from industrial processes and recycling part of the nutrients from downstream, this approach can make a significant contribution to reducing the environmental impact and cost of microalgae biorefinery.

There are still challenges to overcome when using flue gas as a carbon source for microalgae cultivation. For instance, the quality and composition of flue gas may vary depending on the source and type of industrial process. Therefore, it is essential to optimize the use of flue gas to achieve optimal microalgae growth and productivity. Except for this, there is a need to scale up microalgae cultivation to an industrial level, while ensuring sustainability and cost-effectiveness.

Despite these challenges, the potential benefits of using flue gas as a carbon source for microalgae cultivation are significant. The use of flue gas can reduce the environmental impact of microalgae biorefinery and provide a low-cost and sustainable carbon source for microalgae cultivation. Moreover, the production of valuable microalgae can have several applications, such as biofuel production, wastewater treatment, and food and feed production. These applications provide a promising future for microalgae biorefinery, and the use of flue gas as a carbon source can significantly contribute to achieving a sustainable and circular economy.

The combination of flue gas and microalgae cultivation processes is a complex system that requires careful consideration of various factors, such as the source of flue gas and the desired microalgae product. Therefore, in the following sections, a more detailed process flow of how to effectively combine these two processes is discussed.

2. Carbon source and culture systems of microalgae

One object of the combined process is carbon sequestration, and the choice of carbon source depends on the source of flue gas. De Assis et al.[6] examined the feasibility of using exhaust gas from gasoline combustion for carbon supplementation and found that the source did not affect treatment efficiency, yield, or biomass composition compared with a high-rate pond supplemented with industrial CO_2. Flue gas from coal-fired power plants has a low CO_2 concentration (usually between 4% and 20%[2]), while flue gas from coal chemical plants has a high CO_2 concentration (usually above 95%[7]). These two sources correspond to two process routes: in-situ mode and off-situ mode.

In the in-situ mode, the microalgae culture facility is located near the gas source, using power plant flue gas with low CO_2 concentration as the carbon source. This gas is less economical to transport, so aeration of the flue gas directly into the nearby microalgae culture sites is a better option. Microalgae strains, such as *Spirulina*[8], *Chlorella*[9], *Chlamydomonas*[10], and *Scenedesmus*[11], can grow rapidly under this concentration of CO_2 aeration.

In the off-site mode, flue gas from coal chemical plants is further processed into other carbon-containing products and transported to the microalgae culture facility as a carbon source. This model enables the microalgae culture site to be located far away from the gas source. If the concentrated flue gas is directly pumped into the microalgae culture medium, the resulting extremely high CO_2 concentration can cause environmental stress, leading to a reduction in the CO_2 sequestration capacity of microalgae cells. Moreover, the concentrated CO_2 supplied for microalgae growth has low utilization efficiency, resulting in high CO_2 supply costs[12]. By contrast, using bicarbonate as a carbon supply significantly benefits *Spirulina* production, with a carbon cost of \$0.359 kg^{-1}, which is much lower than the conventional approach of bubbling CO_2[13]. Processed bicarbonate allows for long-distance and low-cost transportation via vehicles, ships, and other methods[12]. Thus, the economic cost of further processing flue gas from coal chemical plants with high CO_2 concentration is cheaper than that of flue gas from coal-fired power plants.

A key aspect of the off-site model involves converting coal chemical flue gas into bicarbonate, a commonly used carbon source in commercial microalgae cultivation, resulting in a microalgae culture environment with relatively high alkalinity and salinity[14]. The ability to tolerate these conditions is necessary for candidate algal species to be used in this model, though many species have been shown to adapt well to such environments.

Another objective of the combined process is to generate economic benefits by producing microalgal biomass, which makes the choice of the microalgal culture system a crucial consideration. The two main types of culture systems are open systems, which use raceway ponds, and sealed systems, which use PBRs. Open systems typically have lower operating costs, but they also have lower yields and lower product quality. In contrast, sealed systems generally have higher operating costs, but they offer higher yields and better product quality. Therefore, the selection of the appropriate culture system for a specific microalgae product has a significant impact on economic efficiency.

However, only high-value byproduct markets, such as food, nutraceuticals, cosmetics, and pharmaceuticals, can afford the high production costs of current microalgae production systems[15]. Also, it is important to note that the byproducts of microalgae from flue gas utilization are not pure and may contain toxic metals and compounds, making them unsuitable for direct human consumption and animal feed. Therefore, it is necessary to remove toxic metals from flue gas prior to introducing it into the microalgae culture medium to ensure that algal byproducts are free from metal toxicity.

3. Microalgal biorefinery routes

Microalgal CO_2 fixation culture systems can be categorized into four routes based on the two considerations discussed above: in-situ open system, off-situ open system, in-situ sealed system, and off-situ sealed system.

3.1. In-situ open system

In this scenario, low-concentration flue gas from a coal-fired power plant is pretreated to reduce toxic substances and transported through a pipeline to an open microalgae culture facility constructed near the gas source[16]. After the flue gas has cooled, it is pumped into the microalgae culture in an open PBR via a gas distribution unit to support the growth of microalgae. Because the flue-gas-generating plants are near the site of microalgae cultivation, it avoids the need for gas compression. During the aeration process, some of the carbon dioxide in the flue gas bubbles is taken up by microalgae cells, and most of the remaining carbon dioxide is discharged into the environment around the PBR (**Figure 1**).

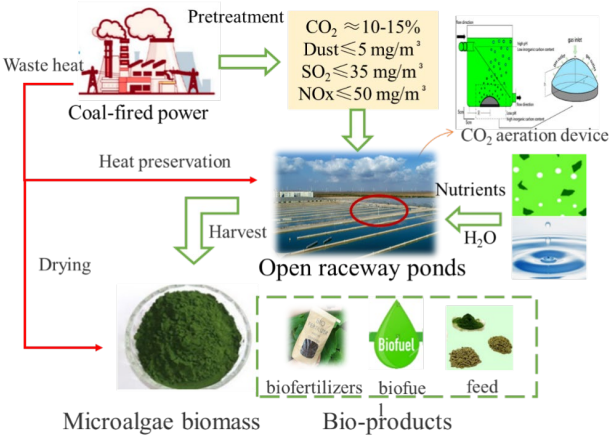

Figure 1. In-situ open route of coal-fired power plant for microalgae culture in open raceway pond.

Open PBRs, particularly circular ponds and raceway ponds, are the most commonly used microalgae culture units, accounting for over 90% of global microalgae biomass production[17]. Some researchers have also employed thin-layer PBRs for scaled-up microalgae culture[18–20]. These systems have the advantage of low construction and operating costs and can be easily built on a large scale. For example, B. Zhu et al.[21] cultured *Spirulina platensis* and *Spirulina sp.* in a 605m^2 raceway pond supplemented with 10% CO_2 from flue gas for eight days, and the final average daily biomass dry weight of the two strains reached 18.7 and 13.2 g.m^{-2}.day^{-1}, respectively. However, there are some notable drawbacks, such as high land requirements, low biomass yield, susceptibility to biological contamination, and high sensitivity to environmental conditions. Nonetheless, the production cost of microalgae culture systems that use open PBRs is significantly lower than those that use closed systems. The successful operation of this process route depends on the introduction of CO_2 into the open PBRs, which can significantly increase the biomass production of the entire system. However, the microalgae species used in this process must meet certain requirements. First, they must tolerate the CO_2 concentration in the flue gas of the thermal power plant, the resulting acidification of the medium, and the shear forces on the algal cells due to aeration. Second, they should be able to adapt to changing environmental conditions and fluctuations without significant biomass loss due to foreign organism invasion. Third, they should be effective in carbon fixation[2,22]. Fortunately, there are many algal species that have been shown to meet these requirements both in and out of the laboratory.

Directly exposed to the air, an open system is easy to be contaminated. One solution to this problem in open systems is selecting microalgae that can grow under extreme conditions, such as salinophilic microalgae, which can grow in high salinity media that most invasive organisms cannot adapt to. For example, marine algal strains, such as *Picochlorum maculatum*, *Nannochloris atomus*, and *Nannochloropsis salina*, were cultured in open raceway ponds with seawater medium, of which the results displayed that no biological contamination occurred[23,24].

Open culture systems are not recommended for the production of high-value microalgae products due to external contamination risks, which can compromise the quality of the final product. However, due to their low operating costs and easy scalability, open systems are theoretically suitable for cultivating microalgae for low-margin bulk commodities, such as fuel, feed, and fertilizer. According to Acién Fernández et al.[15], the production cost of biomass was €4.5 kg^{-1} when using raceway PBRs to grow microalgae supplied with freshwater, fertilizers, and CO_2. If flue gas is used as a carbon source, this can save €0.3 kg^{-1} for purchasing commercial CO_2, reducing the biomass cost to €4.2 kg^{-1}. If raceway ponds are replaced by thin-layer cascade PBRs, the cost can be as low as €2.0 kg^{-1}. While this cost is low enough for microalgae products, it is still not competitive with the cost of other products with equivalent functionality, such as fossil diesel (€1.0 kg^{-1}), soybean oil (€0.5 kg−1), and soybean meal (€0.5 kg^{-1})[17].

However, the in-situ model of this process route makes it difficult to implement, since the microalgae culture site has to be located near the thermal power plant. Thermal power plants require large amounts of water to operate and are usually located near natural water bodies, which are often surrounded by agricultural or forestry land[25]. This conflicts with the need for large areas of non-agricultural land for open culture systems. In addition, a mild and stable climate throughout the year is necessary to ensure biomass production to meet the expectation of long-term continuous operation to reduce costs. While some studies claim that the waste heat of flue gas from thermal power plants can be used to ensure that the open system can operate even in cold winters[26], more research is needed to demonstrate its feasibility.

Although coal-fired power plant flue gases were introduced into this process route for the purpose of carbon sequestration[27], many studies have shown that this system does not achieve the expected emission reduction. In outdoor cultivation, after the introduction of 6564 L of flue gas with 538 L of CO_2, the microalgae

biomass increased by only 0.29 g[28]. As the flue gas is fed into the open reactor, most of the CO_2 is actually still exhausted into the air, and so the contribution to carbon sequestration is very small, which means that this process path is more suitable for production where the main purpose is the product rather than carbon sequestration. However, the low CO_2 utilization rate obviously raises the cost of carbon and aeration, which is certainly not conducive to cost control, and so a series of studies have been conducted to improve this process route.

3.2. Off-situ open system

In this scenario, there is an unrestricted distance between the gas source and the microalgae culture facility, which corresponds to fixing high concentrations of CO_2 in the flue gas of the coal chemical plant with microalgae, as described before. The high concentration of flue gas is further processed into the bicarbonate form and fed to the microalgae culture site via long-distance transportation. The bicarbonate does not require any treatment and can be used directly as a carbon source for the microalgae culture. Since bicarbonate is added to the culture facility in the form of fertilizer, unlike the gas aeration method in the previous scenario, additional power to flocculate and settle the algal cells is necessary. During the incubation process, the microalgae will consume some of the bicarbonate in the culture solution while producing carbonate, accompanied by an increase in the pH of the solution. During the subsequent harvesting process, at least half of the carbon will remain in the effluent as carbonate (**Figure 2**).

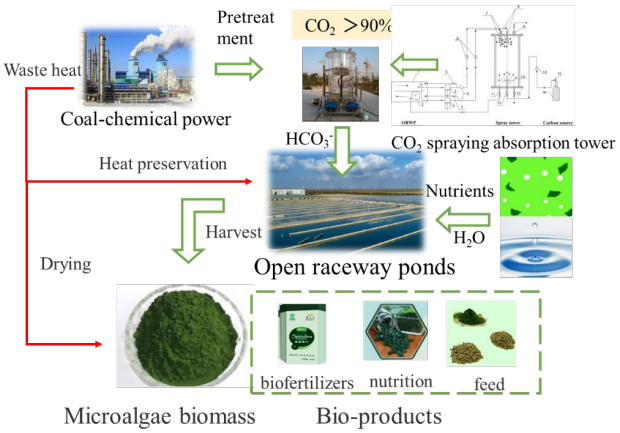

Figure 2. Off-situ open route of coal chemical plant for microalgae culture in open raceway pond.

In this system, it is bicarbonate rather than CO_2 that is the carbon source for microalgae growth, and so the medium in the PBR will be alkaline and the alkalinity of the medium will gradually increase as the microalgae take up bicarbonate and convert it to carbonate. And the cations in the bicarbonate will also give the medium a certain salinity. These place a demand on the tolerance of the microalgae species[29]. According to years of research, it has been found that a wide range of microalgal species can adapt to the high-salt and high-pH environments produced in this scenario, such as *Chlorella*[30], *Desmodesmus*[31], *Dunaliella*[32], *Spirulina*[33], etc. *Cyanobacteria* and *eukaryotic* algae isolated from seawater or saline lakes have better adaptability in this technological route and can be given priority. In a study by Borovkov et al.[34], stable production of *Dunaliella salina* was achieved by culturing two media with sodium bicarbonate concentrations of 2.1 $g.L^{-1}$ and 4.2 $g.L^{-1}$, respectively, in an open rectangular pond outdoors.

In an off-situ open system, the high salinity and alkalinity in the medium effectively inhibit the growth of foreign organisms compared with the in-situ open system, thus making it much less likely to lose yield or reduce quality caused by biological invasion, such as predation by zooplankton or contamination by miscellaneous bacteria. A high-pH environment is also beneficial for preventing contamination with predators

or other photosynthetic microorganisms. The growth of predators, a major threat to microalgae, can be significantly inhibited in the pH range of 8.0–9.0[35].

Microalgae cultivation mainly depends on the availability of sunlight, water resources, and CO_2 supply. Also, the evaporation of water from open systems is an important factor in controlling costs, and so the abundance of local water resources and evaporation rates are also something to consider when selecting a site[36]. In this technological route, the microalgae culture site does not need to be tied to a gas source, which makes the site selection process slightly more relaxed compared with the previous route. However, due to the nature of open systems, important influence factors, such as topography, climate, and water sources, still need to be carefully considered. In particular, the water demand is more urgent than in the previous route because water lost by evaporation can make the already high salinity and alkalinity higher and unsuitable for microalgae growth if not replenished in time[37].

In addition to the conventional terrestrial culture sites, recently some researchers have become interested in growing microalgae on the sea surface, the two most important considerations being the negligible cost of land and the unlimited supply of water. And considering the salinity level of the medium in this route, seawater happens to be a good source of water in this system. The floating open PBR with floating islands at sea has also been proposed and used by researchers for microalgae culture, and one of its features is that it can use the kinetic energy of waves to stir the medium, reducing the cost of additional agitation. As reported in an earlier study by Yang et al.[38], the water footprint for producing 1 kg of biodiesel from marine algae is 399 kg as against 3726 kg for freshwater algae.

From a techno-economic point of view, simply replacing the gaseous carbon source with bicarbonate already allows the off-situ open route to be built and operated at a much lower cost compared with the in-situ open route. First of all, this system no longer requires an aeration unit, which then saves a not insignificant amount of construction costs as well as operation costs, maintenance costs, labor costs, and so on (\$5.0–\$70 kg^{-1} in total[13]) due to aeration. Secondly, the utilization of bicarbonate by microalgae is much higher than in aeration bubbles, and the cost of the carbon source will drop by a large margin. The cost of the carbon source for the *Spirulina* culture with bicarbonate is estimated to be \$0.359 kg^{-1}, which is much lower than the cost of CO_2 aeration (\$1.47 to \$7.33 kg^{-1})[13].

This system is perfectly suited for the production of microalgae products with high added value, as the quality of the biomass products from microalgae can be guaranteed due to low contamination, despite the open culture system. Besides that, under the culture conditions of this technological route, the microalgae can be considered to be under high alkalinity and salinity stress, both of which can lead to the accumulation of lipids, carbohydrates, pigments, and other substances in algal cells, which can enhance the competitiveness of biomass for high-value applications in food, feed, and medicine. Villaró et al.[39] achieved a yield of 30.2 $g.m^{-2}.day^{-1}$ of *Arthrospira platensis BEA005B* in a semi-continuous culture in an $80m^2$ open raceway pond at a high concentration of 16.8 $g.L^{-1}$ of sodium bicarbonate, and the biomass had a high potential for food production.

If the production of high-value products is the goal, the focus of algal species selection must be on having a high content of the target product in addition to the above-mentioned need to adapt to the culture conditions, such as a high protein content of the algal cells when used as feed or food. In addition, considering the need for easy digestion or extraction, algal species lacking cell walls, such as *prokaryotic Spirulina* or e*ukaryotic Dunaliella salina*, are choices that should be favored[40]. Another example is that if sold as a microalgae human food, FDA-approved algae species belonging to *Spirulina* sp., *Chlorella* sp., *C. Reinhardtii*, *Haematococcus* sp., and *Dunaliella* sp.[41] are necessary candidates.

For CO_2 fixation, this process route is capable of much higher carbon sequestration rates than the in-situ open route due to the use of bicarbonate, which has a relatively low risk of leakage. Kim et al. reported that cultures of *Dunaliella salina* with 5 $g.L^{-1}$ $NaHCO_3$ obtained the highest carbon utilization efficiency of 91.4%, but only 3.59% was achieved in CO_2-based cultures[12]. And due to the low environmental impact of the open system, this route should be the most environmentally friendly production process of the four routes. Also, Guo et al.[42] cultured *Arthrospira platensis* in a 660m^2 raceway pond and supplemented the medium with a Na_2CO_3/$NaHCO_3$ mixture as a carbon source, showing a maximum microalgal growth rate of 39.9 $g.m^{-2}.day^{-1}$, which is an example of large-scale cultivation of *Arthrospira platensis* using the off-situ open system.

3.3. In-situ sealed system

Flue gas from coal-fired power plants in this scenario is used as a carbon source for microalgae in a sealed PBR. In this scenario, the microalgae culture site is still located near the gas source, i.e., a coal-fired power plant. However, the microalgae are not grown in an open pool but in a sealed PBR. The flue gas is also transported through a pipe and then pumped under pressure into the PBR, where it mixes with algae cells and exchanges gas in the form of aeration bubbles. The bubbles lose some of their carbon dioxide during the aeration process and they are then enriched at the PBR outlet, and due to the sealed nature of the PBR, these residuals can be reused back at the inlet to increase the carbon fixation rate (**Figure 3**).

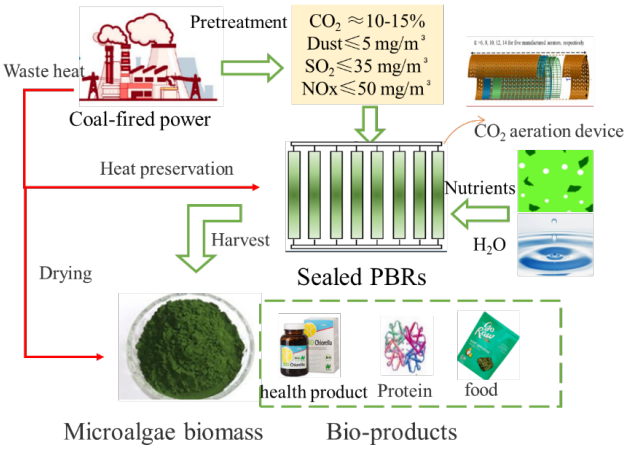

Figure 3. In-situ sealed route of coal-fired power plant for microalgae culture in sealed PBRs.

Compared with open culture systems, sealed PBRs have many advantages in microalgal biomass production, with high yields and quality of output microalgal products. Two *Scenedesmus* strains were cultivated in flat-panel PBRs (55 L) and outdoor raceway ponds (2300 L) using a CO_2 concentration of 1.5% (v/v). Results showed that flat-panel PBRs had an average biomass productivity of 19.0±0.6 $g.m^{-2}.day^{-1}$ compared with 6.62±2.3 $g.m^{-2}.day^{-1}$ for open raceway ponds[43]. However, their excessive production costs limit their application scenarios. The costs of PBRs are approximately 10.0–100 times higher than those of open raceway ponds[13]. There are many kinds of common PBRs applied in microalgae culture, such as flat panel PBRs, airlift PBRs, tubular PBRs, and so on. Among them, the airlift PBR is the most suitable to be used in combination with the aeration process for its excellent gas-liquid mass transfer efficiency[44], which is crucial to achieve emission reduction by making full use of the CO_2 in the flue gas. But from an economic point of view, the plate PBR seems to be the best choice because it has the lowest construction cost[5,45], which is the most important aspect in determining the cost of microalgal biomass production (up to 66.62%–90.29%[46]). However, there are also many studies that have made efforts to improve gas-liquid exchange efficiency, which seems to be a reasonable solution that can balance the economics and carbon sequestration rate of PBRs.

Unlike open systems, sealed systems are less influenced by external factors, such as climate and topography, that limit site selection. However, there are some details to consider. In a sealed PBR, the temperature of the microalgae culture solution will gradually increase with the culture process until it is unsuitable for cell growth. Therefore, suitable cooling measures are necessary to guarantee the operation of the PBR. From this point of view, and in order to reduce the energy consumption in this part, this route is less suitable for high latitudes or high altitudes, where the average temperature is low, than for low latitudes, where the heat dissipation is unfavorable. In addition, sufficient water resources are a condition for cooling water to be secured, but for this route the incubation site is bound to a thermal power plant that also needs cooling water, so it does not need to be considered. However, in warm climates, internal PBR temperatures would be excessive and cooling is uneconomical at scale[43]. In cooler regions, temperature regulation may not be necessary, enabling the use of outdoor PBRs.

On the one hand, the construction cost per unit area of a sealed PBR is very high, about 3–30 times higher than that of an open culture pond and is estimated at \$50–\$500 m^{-2}[41]. The design of one CO_2 bubbling device for each PBR significantly increases its fabricating cost, resulting in a high production cost of at least \$1.0 kg^{-1} caused by PBR depreciation[47,48]. But on the other hand, microalgae may produce ten times higher biomass in a sealed PBR than in an open pond, so the final cost per unit weight of biomass may be the opposite, i.e., it is economically more cost-effective to use a PBR to culture microalgae. It is estimated that the unit cost of culturing microalgae using tubular or plate PBRs may be as low as \$0.68–\$0.7 kg^{-1} compared with a minimum of \$1.28 kg^{-1} for microalgae cultured in open ponds[48]. Another study concluded that the production cost of culturing microalgae in a plate PBR may be as low as €1.6 kg^{-1}[5].

In this route, the system has no special requirements for microalgal algae species because all culture conditions are artificially controllable. In contrast, some microalgae with a high content of high value-added products should be noted in order to be able to produce microalgal products of sufficient value to offset the high cost of running the system. In addition to this, there are some special requirements, such as if the cultured algae strains are able to tolerate temperatures up to 45 °C, then this leaves €0.3 kg^{-1} in the cooling costs[5]. Also, if colder regions are chosen as culture sites in order to reduce cooling costs, then obviously a cryophilic strain is the way to go for screening[49]. Sung et al.[10] studied the acclimation of four microalgae strains (C*hlamydomonas reinhardti*i, *Chlorella sorokiniana*, *Neochloris oleoabundans*, and *Neochloris oleoabundans*) to flue gas using a polycarbonate PBR, which showed that they were all able to grow continuously at this level of CO_2. YY Choi et al.[50] cultivated *Haematococcus pluvialis* in a tubular vertical bubble column PBR aerated with 10% CO_2 and found that the microalgae showed a good growth rate with KOH buffering.

The high cost of PBRs in this route results in higher production costs, ranging from €2.90 kg^{-1} to €290 kg^{-1} in different studies[47,51,52]. This system has the potential to produce low-cost high-yield bulk products. However, considering that the stable and controlled environment in the PBR offers the possibility of producing high-value products, it is clearly more economical to use this system for the production of high-value-added products, such as food, nutraceuticals, and even pharmaceuticals. It has also been suggested that the price of microalgae biomass should not be less than €20 kg^{-1} if the economics of culturing microalgae in a sealed PBR is to be achieved even at a large scale[17]. In a scientific study, *Nannochloropsis gaditana* was cultivated using flue gases on demand from a coal-fired power plant, and the biomass analyzed complied with Spanish regulations for use as animal feed[53].

The carbon sequestration rate of sealed PBRs is much more substantial compared with that of open culture systems, and most of the supplemental carbon source can be converted into microalgal biomass, mainly because the gas passing through the medium can still be collected and re-pumped into the PBR. Moreover, the

carbon source in this route is taken up by microalgae cells in gaseous form, and this uptake is much faster than that of bicarbonate in ionic form, and the final macroscopic manifestation is that this pathway has a fast and sufficient carbon sequestration capacity.

Many studies have conducted large-scale cultivation of microalgae using in-situ sealed routes. For instance, Ye et al.[54] cultivated *Arthrospira* sp. cells in 900L tangential spiral-flow column photobioreactors using CO_2. SY Choi et al.[55] proved the feasibility of large-scale cultivation using in-situ sealed systems. The engineered *Cyanobacteria* strains were cultured in a sealed 100L scalable serial column-shaped PBR using 5% CO_2 from flue gas.

3.4. Off-situ sealed system

In this route, microalgae are cultured in a sealed PBR with a carbon source derived from bicarbonate processed from coal chemical flue gases. The bicarbonate is transported and added to the microalgae medium as a nutrient supplement, which circulates in the PBR under the action of a power unit, such as a peristaltic pump, which is supplemented by the lack of power provided by aeration. During the circulation process, the microalgae take up bicarbonate to proliferate and convert it to carbonate. Thus, the system effluent still has a large amount of carbonate and bicarbonate, which can be harvested and continue to be replenished to restart the cycle in a new medium (**Figure 4**).

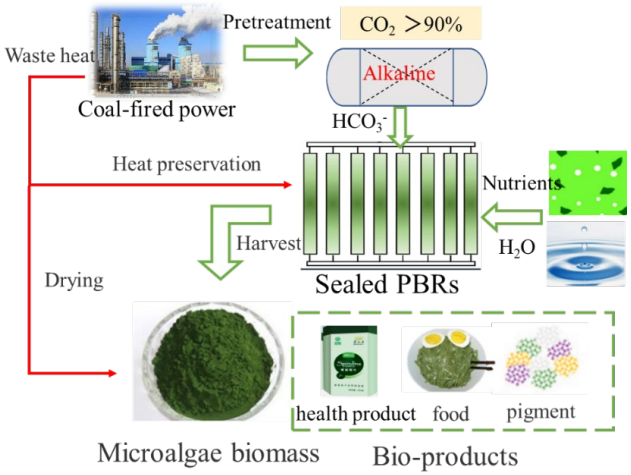

Figure 4. Off-situ sealed route of coal chemical power plant for microalgae culture in sealed PBRs.

This route takes a sealed PBR and does not require an aeration strategy, and so the range of candidates is broader than the previous routes. However, considering the need to utilize additional power to keep the medium circulating, some PBRs with larger vertical heights, such as column PBRs, seem to increase their operational costs due to the need to overcome the large gravitational potential energy of the medium. Horizontal tubular PBRs, flat plate PBRs with horizontal deflectors, photo-stirring tanks, or sealed raceway pond PBRs may be more economically efficient. However, the restriction on vertical height inevitably results in an increase in construction area, which is reflected in an increase in construction costs. For PBR selection, the trade-off between increased construction costs and reduced operating costs requires additional data analysis. Although efforts can be made to reduce all construction and operating costs of a PBR, the PBR is still a very large part of the product cost in this system. Therefore, this system can only be used to produce high-value products that are not cost-sensitive, such as food and pharmaceuticals.

In addition to the above-mentioned traditional PBRs, a new type of PBR, namely the floating PBR on the sea surface, has received increasing attention in recent years[56], and based on it, a promising technological

route for bicarbonate culture of microalgae has been developed. This PBR wraps microalgae culture medium in a bag made of transparent membrane material, which floats on the offshore sea surface. Depending on the purpose of the culture, the PBR can be made with semi-permeable membranes, reverse osmosis membranes, or plain PVC membranes, and the cost can be very low (approximately \$7.00 m^{-2}[57]). This PBR naturally has lower land costs, water costs, and light costs. Supplied with sodium bicarbonate as the carbon source, floating PBRs have higher biomass yields than those of raceway ponds[57]. A simple floating PBR without mixing or without the use of an aeration device was developed based on a bicarbonate carbon supply, and a peak biomass productivity of 3.10 $g.m^{-2}.day^{-1}$ was achieved with this PBR in the cultivation of *Dunaliella tertiolecta*[58]. In its design concept, the waves can provide mechanical energy to agitate the algal cell suspension, which saves the cost of power in a terrestrial PBR. In addition, the heat released in the PBR due to the vital activity of the algal cells can be transferred to the seawater in time, which saves the cost of dedicated cooling in land-based PBRs. The characteristics of this PBR do not lend themselves to an aeration strategy, and bicarbonate solves the problem of its carbon source supply.

Microalgal species suitable for adoption in this route need to meet the basic requirements for bicarbonate as a carbon source, i.e., tolerance to alkalinity and salinity. In addition, since this system uses a sealed PBR, strains tolerant to high temperatures can reduce cooling costs, as previously mentioned. However, the microalgae products of sealed PBRs are preferably high-value-added products, which are less cost-sensitive. Therefore, the yield of the target product remains the most preferred condition to be met by the algae strain under this route. The incubation conditions of the floating system are not very controllable and will eventually be reflected in lower biomass yields, and so this system can be used to produce cost-sensitive, low-cost bulk products, such as feed and fertilizer.

Since the land cost of this technology route is not very high for either land-based PBRs or offshore floating PBRs, site selection is not the primary consideration in commercialization, but rather the availability of feedstock and subsequent processing of the product is more important. For example, for high-value product-driven PBR systems, a location that optimally balances the cost of transporting carbon sources and algal products is most appropriate, while offshore waters near mariculture plants are a logical choice for floating PBR systems where feed is the primary product[56].

In this technological route, the sealed PBR restricts the carbon source to flow only between the medium and the algal cells, and then all the consumed bicarbonate will be converted to microalgal biomass. According to C. Zhu et al.[56] the carbon utilization efficiency of the system can reach 104±2.6% with 8.4 $g.L^{-1}$ of $NaHCO_3$ as the carbon source for *Spirulina platensis* in a floating PBR. Besides that, microalgae convert every two bicarbonates to one carbonate, and because the PBR is sealed, the carbonate cannot be converted back to bicarbonate with the addition of atmospheric CO_2. However, as many researchers have suggested, the medium remaining after harvesting the algal cells can be regenerated using exogenous CO_2, and in this cyclic mode, microalgae can theoretically achieve complete utilization of the carbon source in this route.

4. Perspective

Thanks to the diversity of algal species and microalgal products, all four of these process routes theoretically have a sufficient number of candidates to achieve economic viability. The four process routes also have their characteristics, which are listed in **Table 1**.

Table 1. Some features of four technology routes of combined process.

		In-situ open route	**Off-situ open route**	**In-situ sealed route**	**Off-situ sealed route**	**Ref.**
Utilization of flue gas		CO_2 bubbles	Bicarbonate	CO_2 bubbles	Bicarbonate	
CO_2 fixation rate	Microalgae strains	*Nannochloropsis oculata*	/	*Arthrospira* sp. cells	*Chlorella vulgaris*	[54,59,60]
	Cultivation condition	1191m^2 raceway ponds	/	900L tangential spiral-flow column PBRs	Vertical bubble column glass reactors with a working volume of 500 mL	
	Data	40.7 $g.m^{-2}.d^{-1}$	/	0.665 $g.L^{-1}.d^{-1}$	0.408 $g.L^{-1}.d^{-1}$	
Carbon utilization efficiency	Microalgae strains	*Scenedesmus acutus*	*S. platensis*	*S. platensis*	*Dunaliella salina*	[12,61–63]
	Cultivation condition	Raceway ponds with area of 5.6 m^2 and a volume of 900 L; use of membrane carbonation to deliver CO_2	800m^2 raceway pond supplied with NH_4HCO_3 and $NaHCO_3$	4L helical PBR	250mL baffled culture flasks containing 220 mL of either Modified Johnsons medium with 5 $g.L^{-1}$ of $NaHCO_3$	
	Data	78% ± 55%	70.50% ± 4.76%	50%–69%	91.40%	
Biomass productivity	Microalgae strains	*Staurosira* sp.	*Chlorella sorokiniana str.* SLA-04	*Arthrospira* sp. cells	*Trebouxiophyte*	[25,54,57,64]
	Cultivation condition	Raceway ponds with area of 400 m^2 and depth of 15 cm	Raceway ponds with area of 4.2 m^2 and depth of 17.8 cm	900L tangential spiral-flow column PBRs	Bubble column PBR with 5cm diameter and total of 600 mL of working volume supplied with 300 $mmol.L^{-1}$ of bicarbonate	
	Data	21.1 $g.m^{-2}.d^{-1}$	18.0 ± 1.8 $g.m^{-2}.d^{-1}$	0.29 $g.L^{-1}.d^{-1}$	0.80 g L^{-1} d^{-1}	
Biomass production cost		€5.0–11.0 kg^{-1}	\$3.27 kg^{-1}	€3.1–6.0 kg^{-1}	Extra \$5.0–\$70 kg^{-1} cost than those of raceway ponds	[5,13]
Products		Biodiesel, biofertilizer, animal feed, etc.	Biofertilizer, animal feed, human food, etc.	Human food, medicine, cosmetics, etc.	Human food, medicine, cosmetics, etc.	
Product market size		Large	Middle/large	Small	Small	

Finally, for one of the two main purposes of the above-combined processes, in terms of economics, so far, except for a few cultures with specific high-value products as target products that have been commercialized, more experimental and field data are needed for the other process routes to advance. And, if significant profits are to be realized, large-scale industrial cultivation for the production of low-value, low-margin bulk commodities, such as biodiesel, biofertilizer, or animal feed, is the way to go. From this point on, the open route with lower operating costs and easier to scale up is the one that needs to be focused on.

In terms of carbon sequestration, the most promising process route is the in-situ sealed route, but this also implies a higher cost of sequestration. Moreover, in order to achieve significant carbon sequestration, large-scale farming is inevitable, and this route also requires high construction costs. In order to solve this contradiction, the production of microalgae products with high enough value to be economically profitable, carbon taxes in the carbon trading market, and local government subsidies for green industries can also be included in the economic perspective.

5. Conclusion

The combination process of using flue gas as a carbon source in microalgae biorefinery is able to achieve CO_2 fixation and economic benefits. Four microalgal CO_2 fixation and biomass biorefining technology routes, namely in-situ open systems, off-situ open systems, in-situ sealed systems, and off-situ sealed systems, have

been proposed. They are suitable for different CO_2 sources, microalgal species, and target biomass products. Choosing the appropriate technical route for the scale-up of microalgae CO_2 fixation and biomass biorefining can obtain good environmental benefits and product economic benefits. The in-situ open route is recommended for systems where the CO_2 concentration of flue gas is low and the target products are low-margin commodities, such as fuel, feed, fertilizer, etc., while the off-situ sealed route is recommended for systems where the CO_2 concentration of flue gas is high and the target products are high-value-added products, such as food, health products, cosmetics, etc.

Acknowledgments

This paper was supported by the National Natural Science Foundation of China (52206270).

Conflict of interest

The authors declare no conflict of interest.

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