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Carbon Dioxide Capture- A Challengefor Green Economy.

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Abstract:

Carbon is a compound of multi-fold enactment. The increased carbon dioxide emissions due to various anthropogenic activities have, in turn, posed various threats and challenges to our environment. Accordingly, an innovative approach linked to themanagement strategies of carbon dioxide is the ardent need of the hour. The literature indicates that carbon dioxide capture and utilization (CCU), is an innovative concept to convert greenhouse gas into a valuable feedstock, and has been gaining much attention in the past few decades. The incremental stratospheric carbon dioxide emission due to various human activities like fossil fuel combustion, improved urbanization, etc. has, in turn, led us towards the search for more sustainable and economic routes of synthesis. With regard to this, instead of developing new strategies for the synthesis of the chemical catalysts, more nature inclined technologies and methodologies should be adopted towards creating a sustainable and clean approach. Over previous centuries, nature has designed and evolved sophisticated mechanisms for carbon concentration, fixation, and utilization manifested through autotrophy. Many photosynthetic and chemolithoautotrophic organisms, display excellent ability towards the assimilation of CO_2 and subsequently followed by the conversion into complex molecules. By adopting various modern and sophisticated technologies like genetic engineering and protein engineering, the spectrum of CO_2-derived bio-based products has been expanding at an alarming rate. A wide spectrum of chemicals can be synthesized biologically like bio-plastics, bio-alcohols, biodiesel,to name a few. Accordingly, continuous research on multiple nichesalong with strong collaboration and synchronizationamong scientists and engineers are required to further develop biological systems into viable chemical production platforms.Thus, the present study aims to explore various opportunities and their associated challenges to be applied for biological systems related to CCU.

Key Words-Carbon capture and utilization (CCU); Carbon dioxide;Protein engineering; Stratospheric Carbon reserve

1. Introduction

Carbon capture and utilization (CCU),indicates a concrete approachtowards the goal of the sustainable chemical industry, along with the concomitantreduction in CO_2 emission into the stratospheric level of the atmosphere. Driven by the various policies as per different National and International bodies, the commitment towards reducing the carbon footprint, many innovative and novel CCU technologies toconvert CO_2 into fuels or value-added chemicals have been adopted[1]. The chemical reactions involving CO_2 are utilized by the useof appropriate catalysts, a few of which are inspired by biologicalsystems. The usage of the physicochemical approaches in CCU has beencomprehensively reported in existing literature[2].Despite various advancements which have been incorporatedtowards the utilizing of CO_2 as a chemicalfeedstock, large-volume CO_2 conversion still requires significant research attention. Few notable instances regarding the industrial utilization of stratospheric CO_2 as per previous literature reports include the production of urea (~70 Mt CO_2 per year) methanol (~6 Mt CO_2 per year), salicylic acid (~20 kt CO_2 per year), and propylene carbonate (a few kt CO2 per year) [3]. In contrast to that, it has been reported that on average, the photosynthetic organisms annually transmute around 100 Gt of stratospheric carbon into respective biomass[4]. In context to that, it can be corroborated that Nature has evolvedhighly sophisticated mechanisms for carbon fixation and utilization;a resource that has remains largely untapped and unexplored which in turn could potentially be adisruptive technology in CCU.The increased carbon dioxide emissions due to various anthropogenic activities have in turn resulted in increased global warming and accordingly have received momentous research attention in the past few decades [5, 6].Although a wide spectrum of CO_2 capture and storage platforms have been proposed,the utilization of captured CO_2 from industrial plants is a progressively prevalent strategy due toconcerns regarding the safety of terrestrial and aquatic CO_2 storage.Another remarkablestrategyinvolves the bio-electrochemical techniques through which electricity can be used as a potential energy source forthe microbial catalytic production of fuels and other organic products from CO_2. As per previously reported studies, this approach has been widely explored as a potential technique to reduce CO_2 emissions. Additionally, such approaches have also been reported to generate a wide spectrum of value-added products[7]. Accordingly, the aim of the present review was framed towards the exploration of both the possible routes of the utilization of CO_2 by adopting biological as well as bio-electrochemical utilization. Within the past, a robust increase in CO_2 emission has been reported in various countries like India and China[8, 9]. The literature also indicated that around 80% of the total global energy requirement is met by fossil fuels which in turn contributes towards a major chunk (more than 60 %) of global GHG emission [10]. It has been reported that the utilization of fossil fuels generates around 3×10^{16} g of CO_2 annually[11].Moreover, the traditional wastewater treatment, along with the degradation of the organic contaminants, alsoresults in the emission of huge quantities of CO_2 emissions into the atmosphere, which is expected to rise as high as 1.21 x 104 td $^{-1}$by 2025 [12].

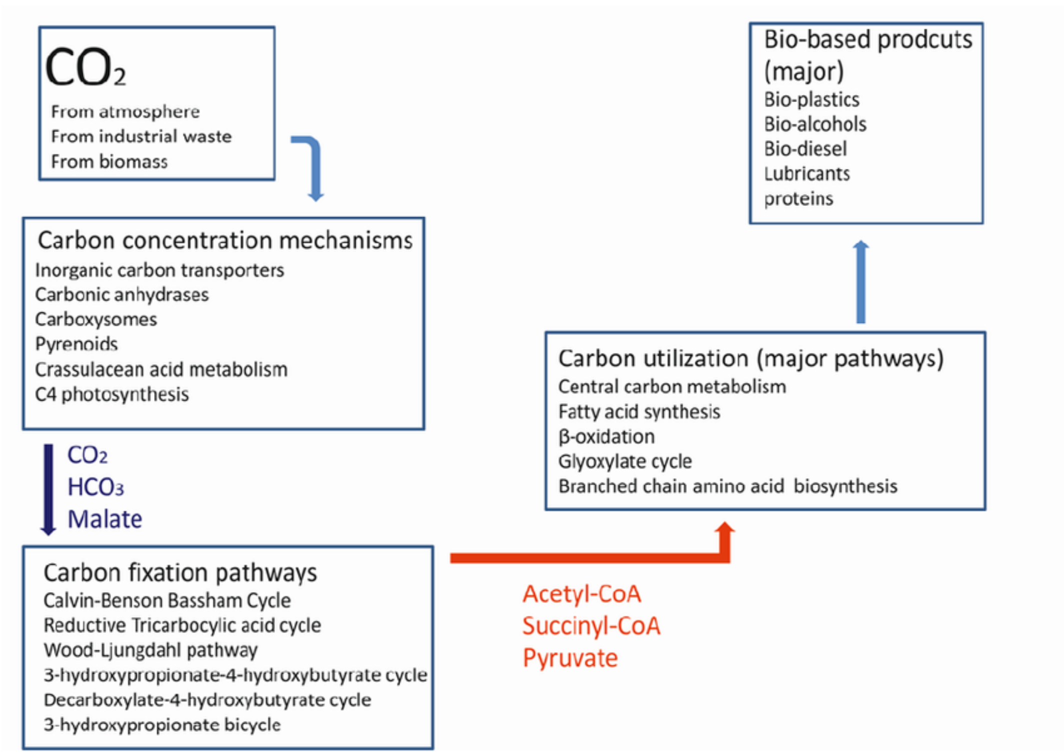

Figure 1.Schematic representation of the carbon concentrating mechanisms, Carbon fixation pathways, and utilization techniques by the autotrophic organism, leading to bio-based products.

2. The spectrum of microorganisms capable of utilizing CO_2

The mechanism of CO_2 assimilation has not only been restricted only towards the photosynthetic organisms (like algae, plants, cyanobacteria, etc.) however as per previous literature reports, such mechanism has also been noted to be involved various autotrophic bacteria and in turn, has received significant research attention [13].Thus, in this section, an attempt has been made to summarize, the microorganisms which are mostly harnessed and accordingly could potentially be scaled up into industrial-scale bioprocess. Few of those microorganisms have already been discussed in the carbon fixation pathway as detailed in Figure 1 along with their possible advantages and disadvantages to be harnessed in the biological CCU. However, this spectrum of microorganisms has not been evolved naturally to be suited for industrial-scale production of the desired products since most of their inherent properties (like growth characteristics, types of metabolites produced, thermostability, and tolerance to inhibitors to name a few) does not suit for such approaches. However, the incorporation of genetic engineering has improved the feasibility to be applied to industrial applications by phenotype improvement for expanding the repertoire of chemical synthesis. Accordingly, an attempt has been made to summarize the recent developments in this particular field of engineering these microorganisms for CCU.

2.1.Algae

Algae is considered as one of the major and significant agentsfor photosynthesis and carbon fixation in context to the industrial application[14]. It has also been reported that algae are more diverse and are widely found in various forms including the larger-sizedmacro-algae which are the most commonly studied along with smaller-sized microalgae[15].Furthermore, the micro-algal species can further be subdivided as per structures and habitats into various classes including green algae, diatoms,red algae, yellow-green algae, golden algae, brown algae, and euglenoids[16]. Moreover, it has been reported that algae exhibit prominent photosynthetic organisms [17].The literature suggested that algae canutilize CO_2 through the Calvin-Benson-Bassham (CBB) cycle, by converting the inorganic carbon into complex organic compounds. The key enzyme involved in the CBB cycle is ribulose 1,5-bisphosphate carboxylase/oxygenase (RuBisCO), which catalyzes the carboxylation of ribulose1,5-bisphosphate to give two molecules of 3-phosphoglycerate (3PG)[18].Out of the two molecules thus formed, 3PG is channeled into central metabolicpathways, while the other is utilized in the continuation of the cycle.However, RuBisCO has been reported to have less catalytic efficiency[19]. In addition to that, RuBisCO has also been reported to bind with oxygen due to the presence of oxygenase activity which in turn leads to photorespiration and generation of unwanted products and accordingly are linked to various drawbacks linked with the O_2 and CO_2 concentration in the atmosphere[20].To overcome these complications, algae have undergone various mechanisms linked to carbon dioxide concentrating mechanisms (or CCMs). The literature indicated that there are three major constituents of a CCM; 1) active bicarbonate(HCO^{3-}) uptake transporters, 2) a suite of carbonic anhydrases(CAs) localized strategically within the cells, and 3) a subcellularmicro-compartment within which most RuBisCO is located within thepyrenoids of the chloroplasts [21].The domain of algal transgenics has received significant research cognizance in the past few years[22]. The finest available tools of genetic engineering have been harnessed to develop various model organisms like the green algae*Chlamydomonas reinhardtii* and *Volvox carteri* and the diatom*Phaeodactylum tricornutum.* Furthermore, the use nuclear transformation has also been adopted for many types of algae, including the industrially-relevant speciessuch as the green micro-algae *Dunaliella salina* and *Haematococcuspluvali*[23]. Various strategies have also been developed and thereby adopted for the modification of various green algae [24].The literature reported that RNA silencing has been adopted to down-regulatethe entire gene family which encodes for light-harvesting antennacomplexes of *C. Reinhardtii*[25]. The obtained results indicated that the engineered alga exhibited anenhanced efficiency of cell cultivation under elevated light conditions. It was also noted that upon the application of various modern and sophisticated techniques, the green alga *C. reinhardtii*was reported to expresscomplex mammalian proteins in the chloroplasts, including a full-lengthIgG-1 human monoclonal antibody[26].A recent report has comprehensivelyreviewed the current state of this field of research and its potentialfuture applications[27].

2.2.Cyanobacteria

Photosynthetic prokaryotes can primarily be categorized under five major phyla namely cyanobacteria,proteobacteria, chlorobi, chloroflexi and firmicutes. The literature indicated that cyanobacteria are also referred to as micro-algal species [28]. However, unlike algae, cyanobacteria are prokaryotic in origin and possess their photosynthetic pigment within the cytoplasm rather than specializedorganelles as compared to eukaryotic plants and algae. These organisms also have been reported to fix atmospheric nitrogen by using nitrogenase as well as inorganic carbon. The literature also suggested that cyanobacteria are believed to play a key role in the earlyatmosphere formation resulting in the reduction of CO_2 concentration and therebyelevating the oxygen concentration [29]. Moreover, cyanobacteria are still considered to have a major role towards 20-30 % of earth's primary photosynthetic activity as among the different photosynthetic organisms[30].The literature indicated that the enzyme RuBisCOis primarily responsible for the utilization of carbon in cyanobacteria, and also catalyzes the same reaction similar to algae for the CBB cycle [31].To achieve CCM, the Cyanobacteria primarily rely on carboxysomes. However, cyanobacteria are reported to be more efficient towards atmospheric carbon fixation due to the presence of a relatively simpler structure in comparison to the algae[32]. In addition to that, the biomass yield for cyanobacteria is also lower as compared to that of algae.Accordingly, cyanobacteria have been reported to be harnessed more frequently for inorganic carbon fixation due to the presence of relatively simpler genetic make-up which can be genetically improved using various genetic engineering techniques for better biomass yield and RuBisCO's CO_2 affinityincreased RuBisCO's CO_2 affinityand production of useful [33].Previously reported literature also summarized and reported recent advances in the metabolic engineering of cyanobacteria, includingthe production of ethanol, isobutanol, and isoprene[34].

*2.3.*Genetically modified *Saccharomyces cerevisiae*

Previously conducted studies reported that *Saccharomyces cerevisiae* has been relatively less harnessed as a potential mitigative measure for stratospheric CO_2

emission as compared *E. coli*. The heterologous expression ofprokaryotic RuBisCO from *Thiobacillus denitrificans* and PRK from*Spinacia oleracea* were also demonstrated in *S. Cerevisiae* [35].The improved production of malic acid using *Saccharomyces cerevisiae* by engineering the pathway of CO_2 fixation via carboxylation of pyruvate is another notable example. Engineered *S. cerevisiae* strainhas been reported to produce malate at a titer of up to 59 g/L, with a malate yield of 0.42mole/mole glucose[36].

2.4.Protein engineering for enhanced biological CCU

Protein engineering is an important aspect of genetic engineering and also contributes enormously towards the advancement of biological CCU as indicated in the earlier section. A wide spectrum of enzymes or proteins has not been evolved or designed to be applied for industrial applications. However, those properties can be customized by the application of a wide spectrum of state-of-the-art protein engineering approaches like rational designing, directed evolution, etc. A wide spectrum of properties like enzymatic activity, specificity, selectivity, thermostability, tolerance to organic solvents, and inhibitors are a few properties that can be enhanced using protein engineering [37].

2.5.RuBisCO and RuBisCO activase

The use of engineering tools to improve the selectivity of RuBisCO has been practiced widely[38]. Application of site-directed mutagenesis on RuBisCO from green alga *C. reinhardtii* has resulted in the modification on a particular domain which in turn has resulted in the retrogressed CO_2 selectivity and thereby decreased CO_2 utilization efficiency [39]. Accordingly, this study summarizes the widely accepted hypothesis of the selectivity of the RuBisCo through complex interaction among various amino acid residues apart from their active site. Accordingly, the research niche has beenchanneled toward applying directed evolution to improve the catalytic efficiencyof RuBisCO[40].

2.6.Bioelectrochemical processes for carbon capture andutilization (CCU)

The exhaustion of fossil fuels due to various anthropogenic activities has, in turn,led to their immense shortage, and accordingly, their priceis expected to rise exponentially. To circumvent such critical issues, renewable energy sources for energy procreation have received significant research attention in the past few decades[1]. One of the widely adopted routes for such development is the conversion of CO_2 into such compounds having the potential to store energy. The adoption of such practices will not only promote the replacement of fossil fuel but will also promote and contribute towards attaining overall sustainability. Methane is one such universally recognized compound as a promising alternative that can store energy adopting various chemical routes[41]. Methane is being generated through the anaerobic respiration pathway of various anaerobic microbes. The literature indicated that methane can be harnessed for generating energy which in turn can be used for the genesis of electricity. Additionally, methane could also be used as the precursor for the generation of biodiesel and related products[42]. Accordingly, the route of methane procreation involves the utilization of CO_2 for the subsequent conversion into a clean source of energy. There are two predominant pathways of methane genesis namely biotic and abiotic. The biotic route of methanogenesis involves microbial interference whereas the abiogenic route involves the thermal fissure of kerogen. It has been reported that methane production through the biogenic route is widely available. More than 20% of the total reserve of natural gas is reported to be produced by microbes[43].The potential of the electromethanogenesis process has been investigated globally. The potential of microbes to generatemethane from CO_2 reduction via an electrode, which is used asa direct electron donor, has been earlier illuminated in the last few decades[44]. The production of methane has been observed with a cathode potential of −0.7 V against Ag/AgCl (equivalent to −0.5against standard hydrogen electrode (SHE). At a potential difference of−1 V, 96% of current has been reportedto be captured into methane. The obtained currentdensities and the small amounts of abiotic hydrogen generationindicated that methane was directly produced from current, andnot from hydrogen gas. Alternatively, it was claimed that a smallamount of methane was directly generated via accepting electronsfrom the electrode, while the remaining portion was biologically generated byhydrogenophilic methanogenesis, consuming abiotic H_2which was generatedfrom the reduction of water molecules[45].Similar results (−0.2 V against the SHE) were achieved in other literature reports. The anodic compartment of a dual-chambermethane-generating microbial electrolysis cell (MEC) fed with acetate, as the main source of carbon, inthe anaerobic medium. The cathodic segment was continuously fed with a $CO_2$and N_2 gas mixture for pH adjustment and carbonate supply. The obtained results indicated 94% removal of acetate at the anode chamber via anaerobic oxidation with a columbic efficiency of more than 90%. The obtained electric power was mainly recovered in the form of methane[46]. In addition to that, the literature also indicated that activated sludge can also be a potential substrate for methane procreation using CO_2 by various methanogens via an anaerobic route. The production of methane was reported to increase by 70-fold via dynamic methanogens after 72 hours in the presence of CO_2 and H_2. The electrochemical conversion Bioelectrochemical systems (BESs), including microbial fuel cells (MFCs), microbial fuel cells (MECs) are reported to be the most promising modes for renewable energy procreation[47].

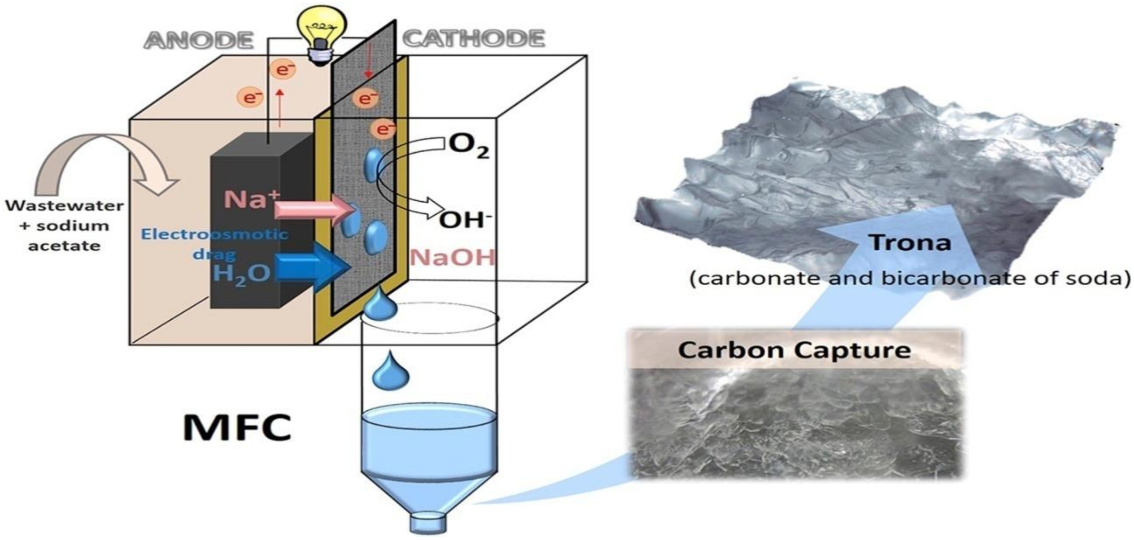

Figure 2A typical representation of electrolysis cell.

The MFCs are reported to generate bio-electricity using organic wastewater as the feedstock using anodic electrophilic bacteria, whereas MECs mimicphotosynthesis, by electro-synthesizingthe value-added chemicals using CO_2 via the cathodic growth of microorganisms as electrocatalysts [48].The MECs are reported to be more superior to MFCs in context to CO_2 capturing potential and thereby promotingoverall environmental security. This to electric storage in the form of valuable products, the dependence on non-renewable energy sources will also decrease[1]. Accordingly, the MECs have grabbed significant research attention in the last few decades, as a novel tool for fuel generation like acetate, formate,hydrogen, or alcohols [45].

2.7.Bio-based products from CO_2

The wide spectrum of CO_2 utilizing microorganisms has been studied. The ability to genetically modify microorganisms and the use of protein engineering to tailor enzymatic/proteinproperties have vastly expanded the repertoire of bio-basedproducts that can be synthesized directly from CO_2which has been listed in Table-1.

Table 1 Representative bio-based products derived from CO_2.

Bio-based products	Organisms	Species	Chemical	Productivity
Bio-plastics	Algae	Phaeodactylum tricornutum	Poly-3-hydroxybutyric acid	PHB accumulated to 10% of algal dry weight
	Cyanobacteria	Synechococcus elongates PCC 7942	Ethanol	Ethanol production rate of 0.18 µg/L/h from CO_2 and water
	Cyanobacteria	Synechococcus elongates PCC 7942	Isopropanol	26.5 mg/L of isopropanol after 9 days
Bio-alcohols	Cyanobacteria	Synechococcus elongates PCC 7942	Isobutyraldehide, isobutanol	Productivity of isobutyraldehyde of 6230 µg/L/h was achieved and 450 mg/L of isobutanol was produced in 6 days
	Cyanobacteria	Synechococcus elongates PCC 7942	n-butanol	n-butanol accumulation reached 14.5 mg/L in 7 days
Biodiesel	Algae	Nannochloropsis oculata	Lipids	The maximal biomass and lipid productivity in a semi-continuous system were 0.480 & 0.142 g/L/d with 2% CO_2 aeration

	Algae	Chlorella vulgaris	Lipids	The maximal biomass and lipid productivity were 3.83 g/L and 0.157 g/L/d with CO_2 aeration rate of 0.5 vvm
	Algae	Porphyridium aerugineum	Polysaccharide	~2.5 mg/ml in 20 days
Other chemicals	Cyanobacteria	Synechocystissp PCC 6803	Isoprene	Accumulation of ~50 µg isoprene/g of dry cell weight per day
	Cyanobacteria	Synechocystissp PCC 6803	Sesquiterpene β-caryophyllene	3.7 µg of β-caryophyllene/g of dry cell weight/week
	Yeast	Trichosporonmonilliforme	Salicylic acid	Phenol was converted to salicylic acid with a 27% (mol/mol) yield at 30°C for 9 hours.

3. Challenges

The literature indicated that the industrial cultivation of algae is mainly carried out in open ponds, raceways, or photobioreactors[49].Although, the open ponds for the cultivation of algae have relative ease in their approach, however, there are certain limitations,such as larger land requirement, high cultivation cost, more contamination probability, and lower productivity[50].To circumvent such issues, the use of closed system cultivation using photobioreactor has received significant research attention. The alleviation of the global CO_2reserve represents concurrent global opportunityand thereby helps to attain better and sustainable strategies related to the environment and energy utilization. Such strategies include various environmental-friendly processes linked with CO_2reductions, the generation of industrially value-added chemicalsfrom CO_2,and CO_2 recycling techniques integrated with sustainableenergy. MES technology has currently been considered as one of the mostpotent approaches to convert CO_2 into valuable chemicals. Accordingly, in context to this, microalgae have been counted among the most productive biologicalplatforms for carbon fixation and biomass production.Theintroduction of microalgae in the biological carbon emission mitigationprocess is associated with dual benefits, where CO_2 could be directlysupplemented towards the growth of microalgae growth and indirectly employedin extraction and transesterification processes. In addition to that, the harvested microalgal cells have also the potential to be utilized for the generation of renewable energy, food products, value-added chemicals, nutrients, and more.Recent advances in the field of bioprocess engineering of algae havebeen fostered in the past few decades, both in the perspective of scope and diversity. Ahuge advancementhas been achieved inthe various spheres of large-scale algal cultivation, right from CO_2 supply to product extraction. In addition to that, it is noteworthy that, life cycle analysis of biodiesel production using algae as the feedstock material using the commercially available data [51].

4. Future prospects and conclusion

The principles of green chemistry have become firmly entrenched in academia as well asindustry. Such principles serve as the blueprint for guiding the design and thereby screening and developing environmentally sustainable processes. Various researchers have put forward various strategies to improve productivity and thereby promote green chemistry. The data summarized and presented in Table 2 indicated good synchronization between the idea of applying biological systems to achieve CCU along with the philosophy of green chemistry.A possible biological route for chemical production does notalways guarantee its eventual translation into a viable industrialprocess. If we consider the principles of green engineeringagain summarized perfectly using a mnemonic “IMPROVEMENTS”, there are challenges that need to be addressed according toour assessment, should we adopt a biological route for CCU.

Table 2 The 24 Principles of Green Engineering and Green Chemistry: “IMPROVEMENTS PRODUCTIVELY”.

Principles of Green Chemistry*	Biological CCU

P	Prevent wastes	Recyclable bio-wastes
R	Renewable materials	Light/ H_2 as an energy source and CO_2/ flue gas as carbon source
O	Omit derivatization steps	CO_2 converted via integrated biochemical pathways
D	Degradable chemical products	Biodegradable products
U	Use safe synthetic methods	Use of non-pathogenic organisms
C	Catalytic reagents	Specialized enzymes, micro-compartments, or organelles
T	Temperature, pressure ambient	Mild cultivation conditions
I	In-process monitoring	Process control for bioreactors or fermenters is available
V	Very few auxiliary substances	Other non-carbon nutrients derived from biomass
E	E-factor, maximize feed in product	Yield optimization via strain selection, genetic engineering, and synthetic biology
L	Low toxicity of chemical products	Biocompatible products
Y	Yes it's safe	Generally safe. Cautions in large-scale H_2/O_2/syngas utilization
	Principles of Green Engineering*	**Challenges of applying biological CCU**
I	Inherently non-hazardous and safe	The use H_2/O_2/syngas presents explosion safety challenges to large-scale production
M	Minimize material diversity	Less of a biological problem
P	Prevention instead of treatment	Bio-wastes are inevitable in fermentation
R	Renewable material and energy inputs	Concentration, composition, temperature, and pressure of CO_2 sources have a direct impact on organismal growth and productivity. The same applies to energy sources (Example- Light intensity and wavelength etc.)
O	Output-led design	The design of a biological system is not trivial and requires

		sound knowledge at both the molecular and system level. The robust genetic tool is lacking for modification of some organisms
V	Very simple	The biological system is inherently complex, highly integrated, and regulated
E	Efficient use of mass, energy, space, and time	Energy and carbon source are channeled into cell growth and biomass accumulation, instead of chemical production. Low productivity is an issue. The biological membrane could be a barrier to mass/energy transfer. Some enzymes display promiscuous activities (moonlighting). Maintaining strict anoxia for anaerobic cultivation, sparging, and cell stirring can be costly and energy-intensive.
M	Meet the need	Less of a biological problem
E	Easy to separate by design	Most organisms or enzymes are not tolerant to solvents used in product separation.
N	Networks for exchange of local mass and energy	Less of a biological problem
T	Test the life cycle of the design	Less of a biological problem
S	Sustainability throughout the product life cycle	Less of a biological problem

Biological CCU is not likely to be a stand-alone technology and accordingly couldpotentially be coupled to other well-established chemical processes suchas gasification and water gas shift (WGS) reaction etc. Various biomass feedstocks likewood and straw contain a huge portion of indigestible chunksthat cannot be degraded and fermented by microorganisms.Accordingly, an excellent alternativein this regard would be biomass gasification, partial oxidation of carbonaceouscompounds into a mixture of CO, CO_2, and H_2.Additionally, the technical aspects described in this review, the advancement of biological CCU is highly dependent on other crucialfactors such as R&D funding commitment to reducingcarbon footprint, governmental policies (*e.g.*, carbon tax,cap-and-trade system), and incentives for CCU (*e.g., a* tax credit forrenewable energy and for developing/deploying energy-efficientequipment/technologies). Often, these factors trigger much discussionand debate, at both national and international levels. Accordingly, we conclude by remarking that scaling-up and industrial implementation of biological and bioelectrochemical utilization ofCO_2can only be realized by close collaboration between scientists and engineers.

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