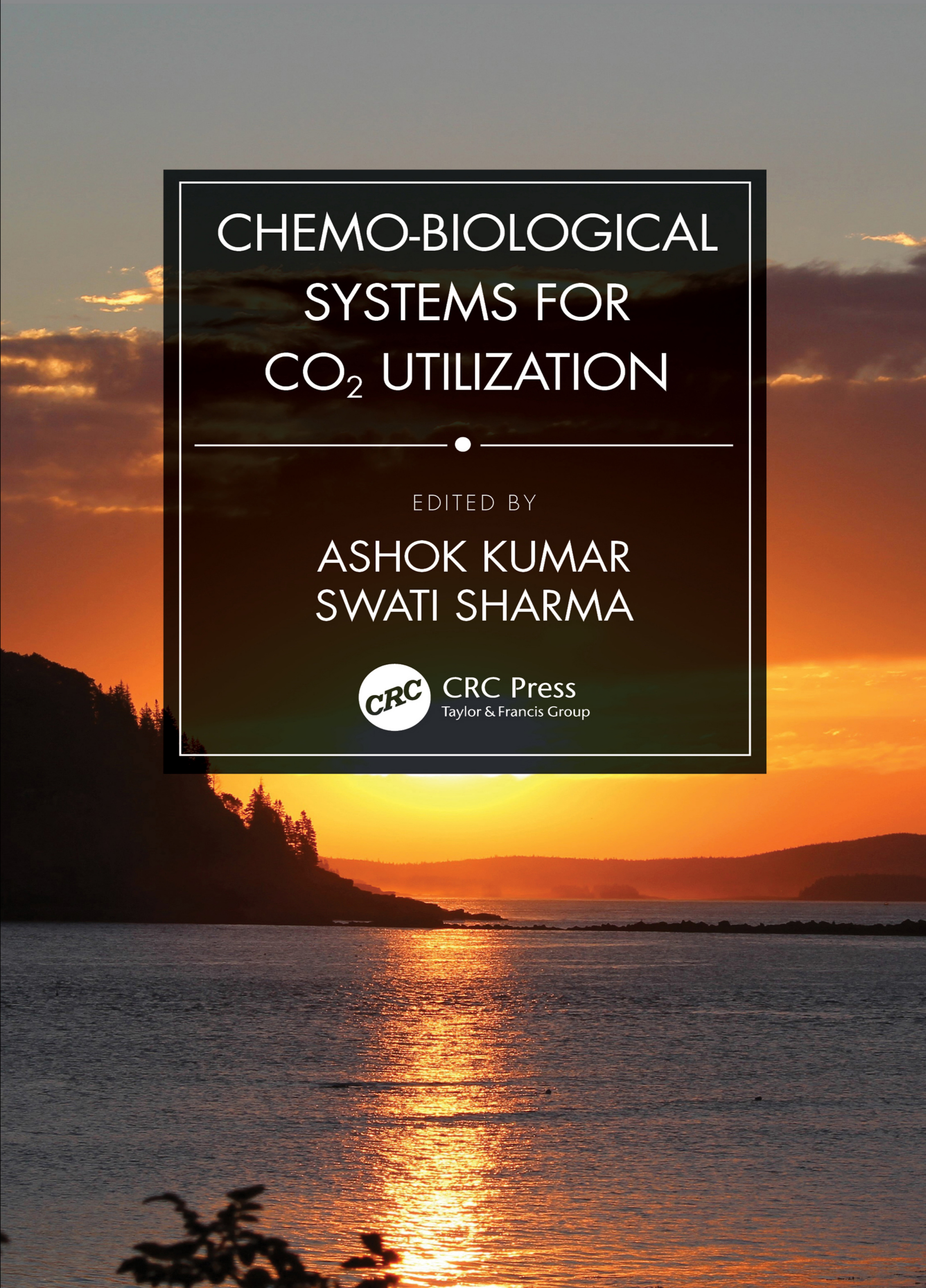

CHEMO-BIOLOGICAL SYSTEMS FOR CO_2 UTILIZATION

EDITED BY

ASHOK KUMAR
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Chemo-Biological Systems for CO_2 Utilization

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Preface

This book will describe the various advanced tools and techniques developed in the past decade for carbon dioxide capture and its utilization. In the ecosystem, CO_2 is primarily absorbed by plants, oceans, algae, and soil contents. But in the 21st century, increased industrialization and urbanization result in a tremendous production of CO_2, which is considered as one of the challenging factors of global warming and a dreadful pollutant that affects the human health. Researchers attempted to develop various techniques using chemical, microbiological, enzymatic, and biomolecular systems in order to absorb the increased concentration of CO_2 in the environment and produce some value-added compounds using CO_2. In this context, various biopolymers, nanomaterials, bioinspired surfaces, polysaccharides, organic solvents, chemicals, enzymes, and metal–organic frameworks (MOFs) were developed and utilized for the sequestration of CO_2 into various carbonates. The biomolecular system has its own importance in the conversion of the atmospheric CO_2 into carbonates. Carbonic anhydrase is one of the fastest enzymes and most studied for the conversion of CO_2. Microflora on the Earth present various classes of methanogens, thermophilic bacteria, phosphate-solubilizing bacteria, and acetogens, which play important roles in CO_2 absorption. Amine-based chemical compounds, porphyrins, ionic liquids, eutectic solvents, ceramics, biochar, and organic solvents have also great potential to convert CO_2 into other fine chemicals or useful products. The major advantages such as stability and efficiency of the systems will be discussed in the various sections of this book.

Nowadays, climate change is considered as one of the major issues, and the massive generation of greenhouse gases from automobiles, industries, and carbonaceous fuels have tremendously promoted the alteration in temperature from its normal cycle. This book will emphasize on the energy generation in the form of biofuels, bioelectricity, or biogas from CO_2 using chemicals; nanomaterials; and microbial, enzymatic, and chemo-enzymatic-integrated systems. This book has been divided into four sections. The chapters 1–4 described the importance and utilization of CO_2 in the living system, and various fundamental methods, policies, and techniques involved in CO_2 conversion. The chapters 5–8 focused on the adsorption and fixation of CO_2 using various ionic liquids, organic solvents, amine-based solutions, electrocatalytic reduction, nanomaterials, porphyrins, ceramics, MOFs, and activated carbons (in particular, biological materials). This section draws the insight of various chemical engineers and researchers working across the globe for CO_2 conversion. The chapters 9 and 10 give the emphasis on the production of value-added products using CO_2 and will mainly focus on the production of biomethanol, industrial carbonates, lime, liquid and gaseous fuels, industrially useful precursors, etc. The chapters 11–13 discuss the potential of the microbial system, enzymes involved in the sequestration of CO_2, and CO_2 utilization. The chemo-enzymatic system developed for the utilization of CO_2 will be discussed in the respective chapters of this book. This reliable information from various active researchers and groups will be helpful to find out the alternative methods for clean energy and mitigating the climate change. This book will help the researchers and industrialists to better understand the correlation between microbial, biological, and chemical products and their roles in the conversion of CO_2 into useable energy and related products.

Editors

Dr. Ashok Kumar is working as an assistant professor in the Department of Biotechnology and Bioinformatics, Jaypee University of Information Technology, Waknaghat, India. He holds an extensive 'research and reaching' experience of more than 10 years in the field of microbial biotechnology, with research expertise focusing on various issues pertaining to 'nanobiocatalysis, biomass, and bioenergy', and 'climate change'. He holds International work experiences in South Korea, India, Malaysia, and Republic of China. He worked as a post-doctoral fellow in the State Key Laboratory of Agricultural Microbiology, Huazhong Agricultural University, Wuhan, China. He also worked as a brain pool researcher at Konkuk University, Seoul, South Korea. Dr. Ashok has a keen interest in microbial enzymes, biocatalysis, CO_2 conversion, climate change issues, nanobiotechnology, waste management, biomass degradation, biofuel synthesis, and bioremediation. His work has been published in various internationally reputed journals, namely *Chemical Engineering Journal*, *Bioresource Technology*, *Scientific Reports*, *Energy*, *International Journal of Biological Macromolecules*, *Science of Total Environment*, and *Journal of Cleaner Production*. He has published 60 research articles, 25 book chapters, and 5 books. He is also a member of the editorial board and reviewer committee of the various journal of international repute.

Dr. Swati Sharma is working as an assistant professor at the University Institute of Biotechnology, Chandigarh University, Mohali, India. She has completed her Ph.D. at the Universiti Malaysia Pahang, Malaysia. She worked as a visiting researcher in the college of life and environmental sciences at Konkuk University, Seoul, South Korea. She has completed her master's degree (M.Sc.) from Dr. Yashwant Singh Parmar University of Horticulture and Forestry, Nauni, India. She has also worked as a program co-coordinator at the Himalayan Action Research Centre, Dehradun, India, and senior research fellow at India Agricultural Research Institute, Pusa, New Delhi, in 2013–2014.

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1 Recent Developments in CO_2-Capture and Conversion Technologies

Tanvi Sharma, Abhishek Sharma, Swati Sharma, Anand Giri, Ashok Kumar, and Deepak Pant

CONTENTS

1.1 INTRODUCTION

Carbon dioxide (CO_2) is one of the greenhouse gases causing a threat to the environment at an increased level. The development of technologies for converting CO_2 into value-added products can slow down global warming and reduce the energy crisis too. Thus, the excess amount of CO_2 can be reduced by converting it into industrial products, which are useful for the industries and mankind. Therefore, CO_2 can be considered as a raw substrate for the synthesis of various commercially important products. CO_2 is emitted from different sources such as cement production units; automobiles; and burning of fossil fuels such as oils, coal, and natural gases (Ashley et al. 2012; Billig et al. 2019). An effective method to reduce the CO_2 level is the primary necessity of time. To date, various methods, including enzymatic, chemical, photochemical, and electrochemical systems, have been exploited for CO_2 capture on laboratory-scale and large-scale applications (Figure 1.1) (Wang et al. 2017).

In the literature, various examples have already been given for CO_2 capture using living organisms and synthetic materials. Using plants and algae to reduce CO_2 is a sustainable and green approach to decrease global carbon footprint. Many plants, such as *Pinus radiata* and *Malus domestica*, and algal species, such as *Chlorella vulgaris* and *Scenedesmus quadricauda*, have been widely studied for their remarkable CO_2-capture capacities (Wu et al. 2012; Pavlik et al. 2017). Among the living organisms, bacteria also play a vital role in alleviating the CO_2 level by using their enzymes such as carbonic anhydrase (CA), formate dehydrogenase, and decarboxylase (Chen 2019). Particularly, CO_2 capture using plants, algae, and microbes may be proven as an eco-friendly approach. Nowadays, amine-based solvents and ionic liquids (ILs) have also been used to reduce the CO_2 from industries such as steel manufacturing and fossil fuel power plants. Yet, these solvents have some disadvantages such as high energy demand and low CO_2-capture efficiency (Vega et al. 2018). To overcome these problems, different nanoporous materials, such as metal–organic frameworks (MOFs), nanoparticles, nanotubes, nanosized zeolites, and activated carbon, are seen as a promising alternative with high CO_2-capture efficiency.

Conversion of CO_2 into an industrial product is required for greenhouse gas mitigation. Nowadays, extensive work is being carried out by researchers for the conversion of CO_2 into hydrocarbons such as methane, formic acid, and methanol. It is still a tedious task to convert CO_2 efficiently, economically, and selectively into products of commercial value (Chen and Mu 2019). In this chapter, we emphasize the development of techniques and advances in CO_2 capture and its conversion into valuable fuels and chemicals.

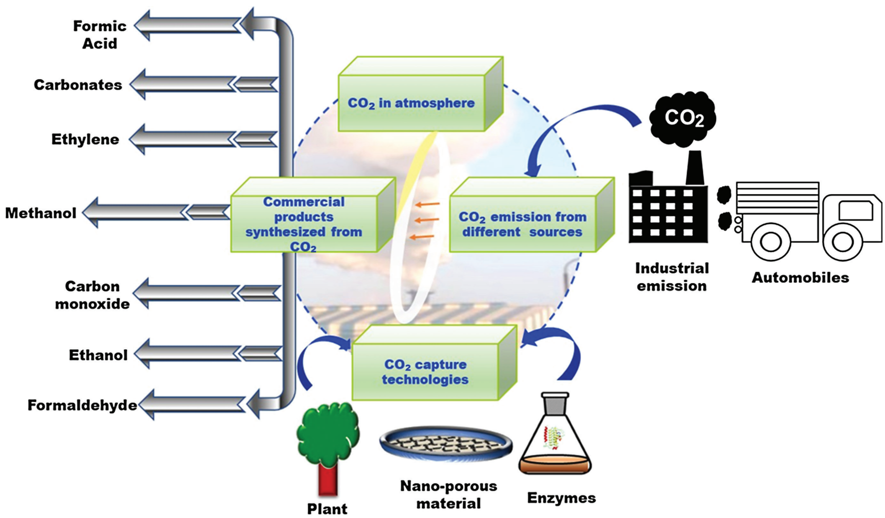

FIGURE 1.1 Schematic representation of various systems used for CO_2 capture and conversion.

1.2 CO_2 CAPTURE IN NATURE

Plant, algae, and microbial enzymes capture the CO_2 by carbon-concentrating mechanism and play a significant role in maintaining the CO_2 concentration in the atmosphere (Table 1.1). The conversion of atmospheric CO_2 by the plants and algae occurs by photosynthesis, and in this process, enzyme ribulose bisphosphate carboxylase/oxygenase (RuBisCo) plays a vital role. The CO_2 released from various industries can be used as a source of carbon for the growth of plants and algae (Mistry et al. 2019). But the RuBisCo has a low affinity for CO_2 over O_2, which showed a low CO_2 fixation rate. Many photosynthetic and autotropic bacteria have CA that helps in CO_2 fixation and its conversion. The major benefit of using microorganisms to convert CO_2 is that they have a natural capability to take up CO_2 through their metabolic pathways (Rittmann 2008). CO_2 sequestration using microorganisms not only provides a green approach to alleviate global warming, but also produces numerous value-added products.

1.2.1 CO_2 Capture by Plants

Different terrestrial ecosystems such as agricultural land, forest, and orchards play a vital role in CO_2 capture. For example, *P. radiata* sequesters about 300–500 tonnes of carbon dioxide per hectare. It was also reported that the CO_2 sequestration capacity of the Chinese forest was 41 tonnes of carbon per hectare (Wu et al. 2012; Mistry et al. 2019). The disadvantage of using trees like *M. domestica* for carbon management is that the CO_2 sequestration capability reduces with the age of the tree (Wu et al. 2012).

TABLE 1.1
CO_2 Capture in Nature Using Plants, Algae, and Microbial Enzymes

S. No	Natural System	Examples	Advantages	Disadvantages	Reference
1.	Plants	*P. radiate*, *Pinus sylvestris*, *Cynodon dactylon*	Orchards and agricultural crops can be used to fix CO_2, and they also provide food.	CO_2 fixation capability reduces with the age of the tree.	Luttge (2004)
2.	Algae	*C. vulgaris*, *S. quadricauda*, *Chlamydomonas reinhardtii*, *Nannochloris* sp.	Biofuel production such as biodiesel, biohydrogen, and bioethanol	Require light during growth and poor utility in industrial reaction.	Batista et al. (2015)
3.	Microbial enzymes	CA, formaldehyde dehydrogenase	Economic viability; the growth rate of microbes is faster than that of plants.	Low stability	Long et al. (2017); Kumar et al. (2019)

Various plants such as crassulacean acid metabolism (CAM) plants, C3 plants, and C4 plants use different carbon fixation pathways. In C3 plants, the product of photosynthesis is phosphoglyceric acid. Common examples of C3 plants are *Oryza sativa*, *Triticum* sp, *Hordeum vulgare*, *Pinus sylvestris*, and *Arachis hypogaea*. To decrease the photorespiration, CAM and C4 plants have photosynthetic adaptation, and the rate of CO_2 conversion is higher in these plants as compared to C3 plants (Grodzinski, Jiao, and Leonardos 1998). The C4 plants are the most productive plants, and some examples of these plants are *Sorghum bicolor*, *Zea mays*, *Bouteloua gracilis*, and *Saccharum officinarum*. CAM plants are mostly found in a dry environment, and to reduce photorespiration, these plants use the CAM pathway. Some examples of CAM plants are *Hoya carnosa*, *Ananas comosus*, *Tillandsia usneoides*, and *Crassula argentea* (Mistry et al. 2019; Nimmo 2000; Sage, Sage, and Kocacinar 2012).

Increasing the atmospheric CO_2 level results in enhanced photosynthesis, but after a certain level, the excessive concentration causes a decrease in the nutritional level of the plant (Raven and Karley 2006; Nogia et al. 2016). Thus, to overcome the negative effects of global warming, the researchers are trying to improve naturally occurring photosynthetic reactions by transferring genes from efficient photosynthetic systems, such as C4 plants, microalgae, and cyanobacteria, to inefficient photosynthetic systems, like C3 plants.

1.2.2 CO_2 Capture by Algae

Algae are one of the most widely researched organisms exploited for CO_2 fixation. The major advantages of algae, including biodegradability, non-toxicity, high CO_2 fixation efficiency, and production of value-added products, make it an ideal candidate for large-scale applications. The CO_2 emitted from various power plants can be used as a carbon source for algal production. Algae use the CO_2 as a carbon source and fix it into organic compounds using the Calvin–Benson pathway (Ullah et al. 2015). These organic compounds can be converted into numerous value-added cellular components such as proteins, lipids, vitamins, and carbohydrates that can be used as feedstock for animal feed, biofuels, and functional foods. Algal species such as *C. vulgaris*, *S. quadricauda*, *Nannochloropsis* sp., *Chlamydomonas reinhardtii*, and *Nannochloris* sp. have been studied to sequester CO_2 (Pavlik et al. 2017). The biofixation of CO_2 and biomass production depend on the characteristics of microalgal species, pH, nutrients, temperature, and CO_2 concentration. The microalgae like *Chlorella* sp., are capable of growing in pH of 5.5–6.0, at the temperature of 30°C, and at 40% CO_2 (v/v). The CO_2 concentration above 5% (v/v) is lethal for algal growth. But the flue gas from the coal-fired plants often includes 10%–15% CO_2 that was reported to use for the cultivation of algae. In flue gas-assisted algal growth, the CO_2 that escapes from microalgal solution due to the high rate of gas flow and low solubility becomes non-toxic to algal species (Zhao and Su 2014; Chen et al. 2014). To improve CO_2 fixation efficiency, screening and domestication of algal species will be major promising strategies.

The most proficient microalgal species such as *C. vulgaris*, *Scenedesmus obliquus, Nannochloropsis oculata*, and *Botryococcus braunii* are used for biofuel production due to their high lipid contents (Kamyab, Chelliapan, Nadda, et al. 2019;

Kamyab et al. 2018). Microalgae like *S. obliquus* is able to produce biohydrogen that is considered as the recyclable and non-pollutant. Meanwhile, *Dunaliella* and *Spirulina* have also been reported as microalgal species widely used for feed production or as functional foods due to their high nutritional value (Song et al. 2019; Kim et al. 2017; Kamyab, Chelliapan, Lee, et al. 2019; Kamyab et al. 2017).

1.2.3 CO_2 Capture Using Microbial Enzymes

The enzymatic conversion of CO_2 can be applied not only for efficient CO_2 utilization but also for capturing waste CO_2 from power plants' flue gas. Generally, enzyme immobilization or absorption technologies have been employed for capturing and long-term storage of CO_2. CA converts CO_2 into bicarbonate ions, which can be further converted to calcium carbonates in the presence of calcium ions. $CaCO_3$ is the product formed during CO_2 conversion that can be separated easily and used as a raw material for many industrial applications such as ceramics, cement, sugar refining, steel, iron, and glass production units (Sharma, Sharma, Kamyab, et al. 2019). In search of potent CA, various strategies such as the isolation of bacteria from different habitats, the use of protein engineering tools, and immobilization of enzyme on various matrixes to achieve high CO_2 conversion become essential (Sharma, Sharma, et al. 2018; Sharma, Sharma, Sharma, et al. 2019; Kumar et al. 2020; Kumar, Wu, et al. 2018; Thakur et al. 2018). Various bacterial genera having CA, such as *Serratia* sp., *Pseudomonas* sp., *Bacillus* sp., *Vibrio* sp., and *Lactobacillus* sp., have been studied for the conversion of CO_2 into $CaCO_3$ (Kumar, Sundaram, et al. 2018; Kumar et al. 2019).

Besides CA, several other types of decarboxylases have been explored for the conversion of CO_2 into eco-friendly chemicals. For example, 4-hydroxybenzoate decarboxylase purified from *Enterobacter cloacae*, *Chlamydophila*, *Pneumoniae* sp., and *Clostridium hydroxybenzoicum* catalyses the reversible carboxylation of phenol with CO_2 to yield 4-hydroxybenzoate (Shi et al. 2015). Another promising route for the reduction of CO_2 into methanol using a multienzymatic system such as formaldehyde dehydrogenase, formate dehydrogenase, and alcohol dehydrogenase has been gaining attention. Conversion of CO_2 into methanol is very beneficial as it is cheaper to produce, is less inflammable, and is advantageous in many industries (Aresta et al. 2014). Methanol is gaining popularity as an alternative to petroleum-based fuels and is beneficial for a safer and cleaner environment. Another product formed from the enzymatic conversion of CO_2 is formic acid, which can be used in textile finishing, paper production, animal feed additive, and chemical intermediates (Alvarez-Guerra, Quintanilla, and Irabien 2012; Rees and Compton 2011). Formate dehydrogenase from *Pseudomonas oxalaticus* is used in the reduction of CO_2 into formate using oxidized methyl viologen as an electron relay (Lu et al. 2006; Long et al. 2017). In this regard, the enzymatic conversion of CO_2 into various products using a free or immobilized biocatalyst seems to be a promising approach.

1.3 CO_2 CAPTURE BY SYNTHETIC MATERIALS

Various chemical solvents (e.g. diglycolamine (DGA), monoethanolamine (MEA), diethylenetriamine) and solid absorbents (nanoparticles, graphene oxide, MOFs) have been used to capture CO_2. The CO_2-capture efficiency of these solvents and solid absorbent materials is high. However, the implementation of CO_2-capture plant at industrial levels such as in iron and steel manufacturing, fossil fuel power plants, and cement production requires novel solvent and solid absorbent formulations with highly equipped machinery (Sharma, Sharma, Kamyab, et al. 2019). The nanoporous material and chemical solvents will be a key solution to enhance CO_2 capture.

1.3.1 CO_2 Capture by Nanoporous Materials

Nanotechnology can provide a viable material for the global reduction of CO_2. Nanoporous materials such as metal and metal oxide nanoparticles, nanotubes, nanosized zeolites, nanosheets, and MOFs have been used as adsorbents for CO_2 capture (Hedin, Chen, and Laaksonen 2010; Sharma, Verma, et al. 2018; Ghodake et al. 2019). Nanoporous materials are one of the best options for capturing CO_2 due to their large surface area, easy surface functionalization, and low cost (Yu et al. 2019). Due to the acidic nature of the CO_2 molecule, the large surface area of nanoporous materials is essential for CO_2-capture applications. Copper oxide nanoparticles show enhanced CO_2 absorption capacity as CO_2 molecules have the electron acceptor property and these nanoparticles have the electron donor property, which results in an efficient CO_2 capture (Kim, Cho, and Park 2010).

Nowadays, MOFs are gaining attention due to their three-dimensional structure, large pore volume, large surface area, and good potential for CO_2 capture. Copper porphyrin-based MOFs are emerging tools as they mimic photosynthesis and offer a method for designing a photocatalyst for CO_2 reduction and its capture (Liu et al. 2013). Various nanostructures have been studied for CO_2 capture; among them, nanotubes, nanofibers, and nanosheets have CO_2 adsorption capacities between 0.26 and 4.15 mmol g L^{-1} (Rodriguez Acevedo, Cortes, and Franco 2019; Wang et al. 2014). Recent advances in nanomaterials like chemically or physically introducing a nanoparticle into a base solvent can obtain liquid nano-absorbents having high CO_2-capture efficiency, improved solvent stability, and decreased solvent vapour pressure (Agarwal, Qi, and Archer 2010). These nanoporous structures have enhanced CO_2 absorption capacity as compared to conventional absorbents.

1.3.2 CO_2 Capture by Graphene Oxides

Recently, graphene, chemically modified graphene, and graphene oxide have been widely used in gas separation, desalination, organic separation, and water filtration processes. Due to their outstanding chemical stability, thermal stability, and mechanical strength, graphene-based materials have attracted the attention of researchers. As such, graphene-based materials are recognized for their potential in capturing CO_2 from the combustion of fossil fuels (Ali, Razzaq, and In 2019; Chowdhury and Balasubramanian 2016). It was found that graphene hydrogel prepared via the hydrothermal treatment of graphene oxide, having high surface area, three-dimensional structure, and large pore volume, showed high CO_2 adsorption capacity (Sui and Han 2015). In another study, it was reported that polyethylenimine loading on 3D graphene led to an increase in CO_2-capture capacity (Liu et al. 2015). Moreover, the nanoporous graphene (NPG) membrane has the ability to separate CO_2 from CO_2/CH_4, CO_2/O_2, and CO_2/N_2. The performance of the NPG membrane used for the separation of gaseous impurities is governed by its monoatomic thickness and high permeability for gas molecules (Lee and Aluru 2013). Although great progress has been achieved on the benefits of graphene-based membranes for CO_2 capture, there are still some challenges that need to be addressed, such as resistance against impurities (SO_2 and NO_2), temperature, pressure, and graphene production cost.

1.3.3 CO_2 by Chemicals

Among various technologies employed for CO_2 capture, chemical absorption using aqueous amine solutions such as DGA, methyl diethanolamine (MDEA), MEA, and pentamethyldiethylenetriamine (PMDETA) is one of the most widely used methods (Dutcher, Fan, and Russell 2015; Mazari et al. 2015). During CO_2 capture by an amine, one CO_2 molecule reacts with two amine molecules to form a stable carbamate ion and protonated amine, limiting the loading capacity to 0.5 mol CO_2/mol amine (Stowe and Hwang 2017). MEA is one of the highly efficient amines employed for CO_2 absorption with 90% efficiency. These amine solutions possess some drawbacks such as high energy consumption in regeneration, high equipment corrosion rate, and low CO_2-capture efficiency. So, the researchers are focusing on novel solvent blends because they possess high thermal stability and are resistant to corrosion and solvent degradation; moreover, these solvent blends require low energy for solvent regeneration (Lee et al. 2012). These novel solvent blends consist of fast kinetic solvents like MEA and other slow kinetic solvents like MDEA (Tong et al. 2013). These blends improve individual CO_2 absorption capacity.

Nowadays, IL has engrossed the attention of researchers due to its unique properties such as non-volatility, high thermal stability, non-toxicity, high polarity, and low energy requirement for energy regeneration (Yu, Huang, and Tan 2012; Cullinane and Rochelle 2004). IL selectively absorbs acidic gases, and these ILs are organic salts with low vapour pressure and increased boiling point. Although IL possesses higher selectivity and CO_2 solubility for CO_2-capture application, some researchers stated that amino-functionalized IL has more CO_2 absorption capacity. Recently, amino-functionalized ILs such as triethylenetetramine lysine and diethylenetriamine lysine were used, and CO_2 uptake capacity of this system was found to be 2.59 and 2.13 mol CO_2/mol, respectively (Jing et al. 2018). But the viscosity and production cost of these ILs are high, which may be one of the concerns in their practical applications.

1.4 SYNTHESIS OF INDUSTRIAL PRODUCTS BY CO_2 CONVERSION

Transformation of CO_2 into useful chemicals/fuels is a promising strategy as such a system not only provides value-added products by consuming CO_2, but also alleviates global warming. Additionally, the CO_2 conversion into industrial products also possesses the potential to fulfil the energy demand in a sustainable manner. CO_2 is a notorious greenhouse gas and a cheaper source of carbon too (Ali, Razzaq, and In 2019; Wang, Yu, and Huang 2018). The development of a new system for converting

CO_2 into hydrocarbon and alcohol is an attractive subject. Various methods are employed for CO_2 conversion, including photochemical, enzymatic, chemical, and electrochemical methods. These methods will vary in the economic value of the product, CO_2 fixation time, and volume of CO_2 utilized (Long et al. 2017). Therefore, CO_2 concentration in future technologies and processes needs to be reduced in a controlled manner that allows the conversion of CO_2 into industrial products.

1.4.1 Methanol

Biomethanol is an alternative fuel to petrochemical industries, and it can be produced by the reduction of CO_2. Firstly, the electrochemical reduction of CO_2 into methanol using formate dehydrogenase, methanol dehydrogenase, and pyrroloquinoline quinone (PQQ) as a cofactor was reported. This approach provides a facile route for the production of methanol from CO_2 under mild conditions (Shi et al. 2015). The catalytic hydrogenation of CO_2 and H_2 is the basis of the syngas process. Various homogeneous and heterogeneous catalytic systems are used for catalytic hydrogenation of CO_2 into methanol. For over 40 years, $Zn/Cu/Al_2O_3$ catalyst system is used for the industrial synthesis of methanol (Yang et al. 2017). Carbon Recycling Inc (CRI) plant in Iceland produces low-carbon-intensity methanol that is utilized for gasoline blending and biodiesel production (Pontzen et al. 2011; Sharma, Shadiya, et al. 2019). CRI is the first company to capture CO_2 from flue gas and to be involved in the large-scale conversion of CO_2 into methanol.

1.4.2 Carbon Monoxide

Carbon monoxide (CO) is considered as waste gas, yet it is a vital feedstock for the synthesis of many fuels and chemicals. Mostly, the conversion of CO_2 to CO occurs in the presence of a catalyst, which results in a decrease in reaction rate and an increase in reaction velocity. Recently, the rhenium tricarbonyl catalyst attracted the attention of researchers for the conversion of CO_2 to CO. Carbon monoxide dehydrogenase from *Moorella thermoacetica* was the first reported biological catalyst for the electrochemical reduction of CO_2, and it exhibits no overpotential. However, the direct electrochemical reduction of CO_2 requires 1–2-V overpotential. Carbon monoxide dehydrogenase is another important metalloenzyme that contains nickel and iron in its inactive site, which catalyses the reversible oxidation of CO to CO_2. Several metal-centred catalysts such as nickel, iron, cobalt, and ruthenium catalyse the conversion of CO_2 into a value-added product (Agarwal et al. 2012; Shin et al. 2003). Further insights in this field are required to develop an effective catalytic system in terms of selectivity and overpotential.

1.4.3 Methane (CH_4)

Methane is one of the most efficient means to store electric energy. The most common methods for methane synthesis are Sabatier reaction and Fischer–Tropsch process, but both of these methods are expensive. For a catalytic transformation of CO_2 to methane, $Rh/\gamma\text{-}Al_2O_3$ catalyst is one of the promising compounds, as this works at low pressure and temperature as compared to other conventional methods that require higher temperatures (Beuls et al. 2012). It was reported that nitrogenase containing molybdenum iron protein also catalyses the reduction of CO_2 to methane (Yang et al. 2012). Nowadays, research has been going for implementing TiO_2 nanotubes for CO_2 conversion; this technique is eco-friendly as it depends on solar energy as an input.

1.4.4 Formic Acid

Formic acid can be used as animal feed additive, silage preservation, fuel for low-temperature fuel cells, and textile finishing, and in paper and pulp industry. The various homogeneous and heterogeneous systems have been studied for the reduction of CO_2 into formic acid (Alvarez-Guerra, Quintanilla, and Irabien 2012). In a homogeneous system, Formate dehydrogenase (FDH) is used for the hydration of CO_2 into formic acid. The catalyst uses NADPH as an electron donor and is embedded in alginate–silica hybrid gel nanostructures; this process occurs at low temperature and neutral pH. In addition to the homogeneous system, several heterogeneous systems have also been developed in the past decades (Lu et al. 2006). The catalytic hydrogenation of CO_2 into formic acid using $Ru(II)Cl(OAc)(PMe_3)_4$ as a catalyst in the presence of alcohol and base shows the high turnover frequency for formic acid production (Munshi et al. 2002). Various other metal complexes such as Ni, Rh, Pd, and Cu have been studied for formic acid formation with excellent yields. Climostat Ltd. (Cheshire, UK) has filed a patent application for the conversion of CO_2 and methane into formic acid using enzymatic catalysis (Fothergill et al., 2014).

1.5 CONCLUSION

Worldwide, CO_2 capture seems to be one of the serious issues. There is a critical need to capture CO_2 from the atmosphere, in order to prevent climate change. Although various efforts have been made to capture CO_2, there are still some challenges that need to be addressed. Various impurities in the flue gases may affect the activity and stability of chemical solvents and absorbent materials. Conversion of captured CO_2 into industrial products at large scale is still needed to be explored.

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2 Heterogeneous Catalytic Hydrogenation of CO_2 to Basic Chemicals and Fuels

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CONTENTS

2.1 INTRODUCTION

Due to economic development, energy consumption, including fossil fuel, is increasing all around the world. The use of fossil fuels for energy production in houses, power plants, and vehicles is the main source of anthropogenic CO_2 emissions. In 2017, CO_2 concentration in the atmosphere reached 405 ppm (Liu et al., 2019). It is predicted that by the end of this century, CO_2 concentration in the atmosphere will continue to rise to 570 ppm. Unfortunately, it is not possible to propose a truly low-carbon technology to the public market in the short term. The best approach will be CO_2 utilization (Araújo et al., 2014).