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Introduction

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Of the final products of the combustion of carbon-based fossil fuels, carbon dioxide (CO_2) has the highest oxidation state and is known as the major cause of global warming. Annual CO_2 emissions from anthropogenic activity in 2018 were approximately 33.1 Gton, an increase of 1.7% compared with 2017 [1]. Since the Industrial Revolution, two trillion tons of CO_2 have accumulated in the atmosphere, and the current atmospheric concentration of CO_2 has reached an unprecedented level of over 400 ppm (Figure 1.1) [2]. The anthropogenic emission of CO_2 is associated with energy consumption, i.e. the combustion of carbon-based fossil fuels, which currently account for around 85% of the world's energy.

According to the Paris Agreement of the United Nations, an overall limit on total cumulative CO_2 emissions is crucial for our future development [3, 4]. According to the 2 °C scenario, further cumulative emissions should be limited to below one trillion ton of CO_2. The spread of renewable energy (35%), advances in energy conservation (40%), and carbon capture and sequestration (CCS) technologies (14%) are sure to contribute to addressing the problem (Figure 1.2) [3]. However, it is clear that these methods will not completely solve the issues arising from the vast quantities of emitted CO_2. In 2017, the International Energy Agency (IEA) presented the Energy Technology Perspectives (Beyond 2 °C Scenario: B2DS), which placed a much greater emphasis on the role of CO_2 utilization for reducing emissions [3]. Indeed, in the next decade, we will still rely on carbon-based products for fuels, polymers, commodity chemicals, cosmetics, detergents, and fabrics in modern life. If these chemicals were to be derived from CO_2 instead of fossil oils, a sustainable carbon cycle will be possible.

1.1 Direct Use of CO_2

Apart from chemical applications, already today, CO_2 is used directly in enhanced oil recovery (EOR), beverage carbonation, food processing (e.g. coffee decaffeination and

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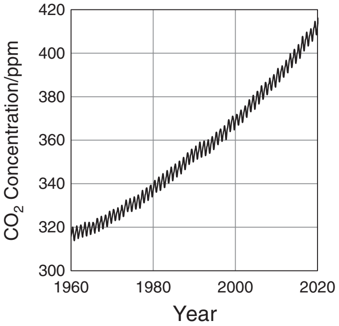

Figure 1.1 Atmospheric CO_2 concentration at Mauna Loa Observatory. Source: Data from National Oceanic and Atmospheric Administration, Global Monitoring Laboratory [2].

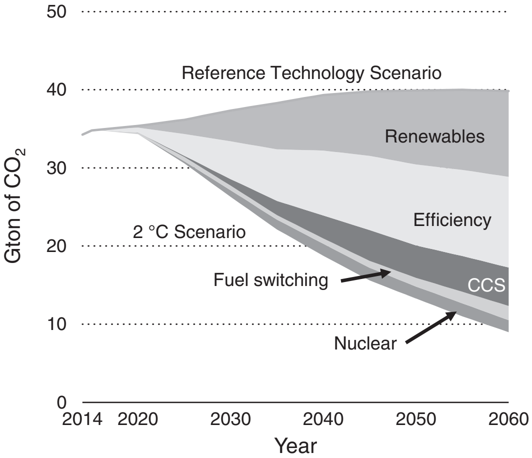

Figure 1.2 IEA 2 °C Scenario (2DS) in Energy Technology Perspectives 2017. Source: Data from Market-driven future potential of Bio-CC(U)S [3].

drinking water abstraction), welding, as a cleaning agent for textiles, and as a solvent in the electronics industry [5]. These approaches are commercially viable. In particular, 70–80 Mton of CO_2 is consumed for EOR in the oil sector. Although such direct utilization of CO_2 addresses a significant amount of CO_2 emissions, these topics are beyond the scope of this book.

1.2 Chemicals from CO_2 as a Feedstock

CO_2 has been recognized as an inexpensive and abundant industrial C1 carbon source. The various chemicals that can be produced by CO_2 conversion are shown in Table 1.1 [6]. The largest chemical use of CO_2 is in the production of urea from ammonia. However, since a

Table 1.1 Chemicals produced commercially from CO_2 .

Chemical	Scale of production/ton
Anthropogenic CO_2 emissions (2018)	33 100 000 000
Urea [7]	181 000 000
Diphenyl carbonate (Asahi Kasei Process) [8]	1 070 000
Salicylic acid	90 000
Cyclic carbonate	80 000
Polypropylene carbonate	76 000
Acetylsalicylic acid	16 000
Methanol (CRI process) [9]	4000

Source: From Omae [6]. © 2012 Elsevier.

huge amount of CO_2 is emitted during methane steam reforming to supply H_2, urea production does not contribute to carbon sequestration at present.

The catalytic copolymerization of CO_2 with epoxides, which provides a thermodynamic driving force due to the strained three-membered ring, is the most prominent example of the synthesis of CO_2-based polymers without formal reduction of the carbon oxidation state. Another example, the manufacture of diphenyl carbonate from ethylene oxide, bisphenol A, and CO_2 instead of phosgene was developed beginning in 1977 by Asahi Kasei Chemical Corporation to address environmental and safety issues. The first commercial facility started operation in 2002 [8]. This process produces high-quality polycarbonate and high-purity monoethylene glycol in high yields without waste or wastewater. In addition, the phosgene-free process emits approximately 2.32 ton/ton$_{PC}$ less CO_2 than the phosgene process according to life-cycle assessment (LCA). Diphenyl carbonate has a large market (3.6 Mton in 2016) for use in automotive parts and accessories, glazing, and medical devices. The phosgene-free technology has already been licensed to Taiwan, South Korea, Saudi Arabia, China, and Russia.

Since 2011, in Iceland, carbon recycling international (CRI) operated the first commercial plant for methanol production from CO_2 via syngas by the reverse water-gas shift (rWGS) reaction (George Olah Renewable Methanol Plant) [9]. At present, more than five million liters of methanol per year is produced using low-cost electricity and high-concentration CO_2 in the flue gas from an adjacent geothermal power plant. It should be noted that this technology is at present only viable in Iceland; however, if there is a surplus of green electricity in the future from an excess of renewable energy, then this process will be attractive at other places, too.

Notably, the amount of CO_2 utilized by all these approaches, including urea and carbonate production, is very small compared with the magnitude of anthropogenic emissions. Therefore, CO_2 conversion into chemicals is unlikely to significantly reduce emissions. Comparatively, it should be noted that fuels are produced and consumed on a much larger scale than these chemicals.

1.3 Application and Market Studies of CO_2 Hydrogenation Products

Hydrogenation of CO_2 could be an efficient option for developing more environment-friendly products as alternatives to fossil-based ones. In terms of practicality, the distribution infrastructure of carbon-based chemicals is well established. However, their manufacturing is currently several times more expensive than their conventionally produced counterparts, mainly due to the costs associated with H_2 production. Some of the key features of CO_2 hydrogenation products and conventional fuels are given in Table 1.2.

1.3.1 Formic Acid/Formate

Formic acid is the first carboxylic acid and is naturally occurring produced by ants, bees, and some plants. In 2016, the global production of formic acid was 1.02 Mton [10]. The general production process of formic acid involves the formal carbonylation of water in a two-step synthesis via methyl formate. Formic acid and its salts (formate) are valuable chemical products used for silage and animal feed (27%), leather and tanning (22%), pharmaceuticals and food chemicals (14%), textile (9%), natural rubber (7%), and drilling fluids (4%) [11]. Recently, formic acid has been recognized as a promising liquid organic H_2 carrier (LOHC) because of its low toxicity, low combustibility, stability, environmental friendliness, and 4.4 wt% (53 g l^{-1}) H_2 content [12–14]. In addition, compressed hydrogen gas can be supplied only by heating of formic acid using catalysts as a chemical compressor [15]. Therefore, advances in the efficient production of formic acid/formates may eventually lead to their large-scale use as LOHCs (see Chapter 10).

1.3.2 Methanol

Methanol, the industrial production of which is mainly from syngas, is in high global demand as a fuel and bulk chemical (Figure 1.3) [17]. One ton of methanol produced by the

Table 1.2 Characteristics of various energy vectors.

Compound	Energy density (GJ m^{-3})	Approx. price per energy (US$/GJ)	Boiling point/ melting point (°C)	Ignition point/ flash point (°C)	Vapor pressure at 25 °C (kPa)
Methanol	15.8	15	64.55/−97.68	470/15 (open)	16.9
Formic acid	6.3	100	100.56/8.27	520/59 (open)	43.1
Natural gas (CH_4)	8.1 (20 MPa)	2	−161/−183	537/−188	147 (15 °C)
Gasoline	34.5	30	17–220/≤−40	300/≤−43	50–93 (37.8 °C)
Diesel oil	36.3	23	140–400/−29 to −18	250/40–70	≤0.35 (37.8 °C)
Hydrogen	5.1 (70 MPa)	120	−252.87/−259.14	500–571/—	1.65×10^5

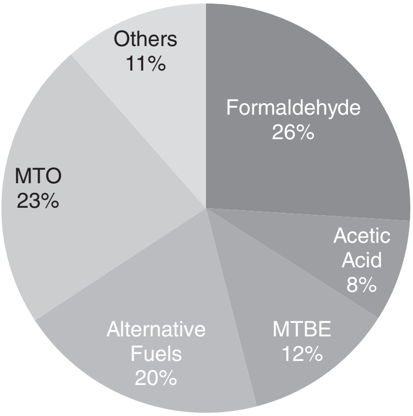

Figure 1.3 Global methanol demand in 2018. Source: Data from Global methanol demand (Methanol Institute) [16].

established process consumes 37.5 GJ of natural gas and emits 1.49 ton of CO_2 [18]. In 2018, the global production of methanol was approximately 91.7 Mton, and since 2015, its production has grown by approximately 16% [16]. Approximately 26% and 8% of the methanol produced worldwide is consumed to produce formaldehyde and acetic acid, respectively, as the conventional demands. Methanol can be used as a fuel for internal combustion engines and fuel cells because it has a comparably high-octane number of 113 and a density approximately half that of gasoline. In addition, methanol can be transformed into gasoline through the methanol-to-gasoline (MTG) process developed by Mobil in the 1970s [19]. Another growing market for methanol is the production of light olefins (i.e. ethylene (152 Mton yr^{-1}) and propylene (103 Mton yr^{-1}) in 2017), [20] which are monomer feedstocks for polyethylene and polypropylene as basic products of the plastics industry [21]. The concept of a so-called methanol economy was independently proposed by Olah and Asinger due to the chemical's promising characteristics for use as an energy vector and chemical feedstock [22–24]. Therefore, the production of methanol by CO_2 conversion is regarded as an attractive and potentially profitable route for CO_2 utilization.

1.3.3 Methanation

CO_2 methanation, also known as the Sabatier process, affords methane by the exothermic reaction of CO_2 with H_2. The commercial methanation of CO_2 is performed at 300–550 °C and above 5 bar. Most CO_2 methanation processes are considered to be a linear combination of rWGS and CO methanation. The process is expected to be a power-to-gas concept for converting renewable electrical energy into methane as chemical energy. In other words, the main goal of methanation is the intermediate storage of renewable electricity in methane as an energy carrier. Since fossil-based natural gas is a common fuel, there would be easy access to existing infrastructure.

Due to the significant interest in CO_2 methanation, the first pilot plant capable of producing 0.5 $Nm^3\ h^{-1}$ of synthetic natural gas was built in Japan [25]. In terms of commercial installations, Audi has an operational CO_2 methanation facility (max. 325 $Nm^3\ h^{-1}$) using renewable H_2 (max. 1300 $Nm^3\ h^{-1}$) from electrolysis (max. 6.0 MW) in Germany [26, 27].

1.3.4 Energy Storage

The two most growing renewable energy sources, solar and wind, are intermittent and thus provide highly fluctuating electrical energy. In addition, the region's best suited areas for the production of renewable energy are often far from consumption areas, i.e. cities. These cause the two key problems of storage and transport. Certainly, electrical energy is an effective way to transfer energy within 1000 km and can be stored in batteries. However, low-cost solutions for the large-scale storage and long-range transport of electrical energy must be developed to improve energy security and balance energy prices.

The transformation of excess renewable energy into chemical energy by converting CO_2 is one promising option. CO_2-based compounds, such as methane, methanol and formic acid, can store energy as gas or liquids with comparably high-energy densities. Especially liquids can be easily transported and release energy as H_2 or electricity through oxidation and fuel cells when there is a greater demand. In other words, CO_2 can act as an energy vector between electrical and chemical energy. Recently, the electroreduction of CO_2 to chemical fuels has been receiving increasing attention because it allows for the direct use of renewable electricity without conversion to high-cost H_2 by water electrolysis (see Chapter 9). Much more CO_2 is in demand as a feedstock for fuels than for chemicals and mineralization. In addition, related photo-catalytic processes gain more and more interest.

1.4 Supply of Materials

The CO_2 hydrogenation approach requires H_2, CO_2, and an energy supply. In particular, how much the energy-intensive hydrogenation process contributes to mitigating CO_2 emissions will be dominated by the H_2 source. Obviously, H_2 must be produced with the help of a renewable electricity source such as water electrolysis and not from fossil fuels.

1.4.1 CO_2 Supply

Capture, purification, and transport of CO_2 are essential for its utilization. Table 1.3 lists several large CO_2 sources with their typical amounts and concentrations of CO_2 as well as impurities. In present CO_2 merchant market (approximately 230 Mton, US$7.7 billion), the fermentation process (i.e. bioethanol production) and ammonia production, which provide close to 100% CO_2, are predominantly CO_2 sources [5, 30]. The CO_2 generated from ethanol fermentation commercially supplies roughly 270 000 ton of CO_2 annually for EOR through pipeline from Kansas to Texas [28]. On the other hand, the production of electricity and heat accounts for 41% of global CO_2 emissions (Figure 1.4), and the transport and industrial sectors account for an additional 25% and 19%, respectively [32]. However, suitable sources of CO_2 for use in chemical transformation are limited. The gases contain various impurities, the separation of which is both energy and cost intensive. To supply CO_2 of an

Table 1.3 Concentration of CO_2 and contaminants from various sources.

Source	Amount/Mton	CO_2 concentration/%	Impurities
Ethanol fermentation [28, 30]	50	99	EtOH, MeOH, H_2O, H_2S
Anhydrous ammonia	30	>95	NH_3, CO, H_2, H_2O
Natural deposits	13	90–100	N_2, O_2, He
Power plants	4287	10–15	N_2, H_2O, SO_x, NO_x, CO
Steelmaking	266	18–20	N_2, SO_x, NO_x, O_2
Cement production [31]	220	14–33	SO_x, NO_x, O_2
Atmosphere	3 200 000	0.04	N_2, O_2, SO_x, NO_x

Source: Carbon Recycling International; Capturing and Utilizing CO_2 from Ethanol: Adding Economic Value and Jobs to Rural Economies and Communities While Reducing Emissions (2017); and Greenhouse Gas Inventory Data [9, 28, 29].

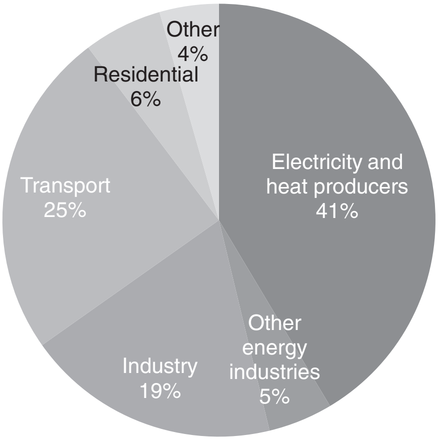

Figure 1.4 CO_2 emissions from fuel combustion. Source: Data from IEA, CO_2 emissions from fuel combustion, 2020 [32].

appropriate quality for use in chemical conversion processes, capture and separation are required (Table 1.4) [33]. The most effective CO_2 capture method as the current industrial standard is chemical absorption in an aqueous solution of an amine-based organic compound. However, the cost (35 US$/ton) and energy consumption ($2.5\,\mathrm{GJ\,ton^{-1}}$) of amine capture must still be reduced to provide economically viable routes from carbon dioxide to fuels [34, 35].

Recently, the direct capture of CO_2 from ambient air, called direct air capture (DAC), has received increasing attention [36]. One of the advantages of DAC is that it can be located anywhere, because it is unnecessary for CO_2 transport. However, from both engineering and chemistry views, there remains much room for improvements to the sorbents and processes. Additionally, thorough techno-economic analyses of DAC processes are necessary [37].

Table 1.4 CO_2 capture technologies.

Capture technology	**Technical principle**
Chemical absorption	Chemical reaction between CO_2 and absorbent by a temperature swing.
Physical absorption	Dissolution of CO_2 into a liquid, the efficiency of which depends on the solubility of CO_2 in the liquid.
Solid absorption	Absorption into solid absorbents, which include porous materials impregnated with amines for low-temperature separation or other solid absorbents for high-temperature separation.
Physical adsorption	Adsorption onto porous solids such as zeolites by a pressure or temperature swing.
Membrane separation	Permeation through a membrane with selective permeability for different gas species.

Source: Based on Styring [33].

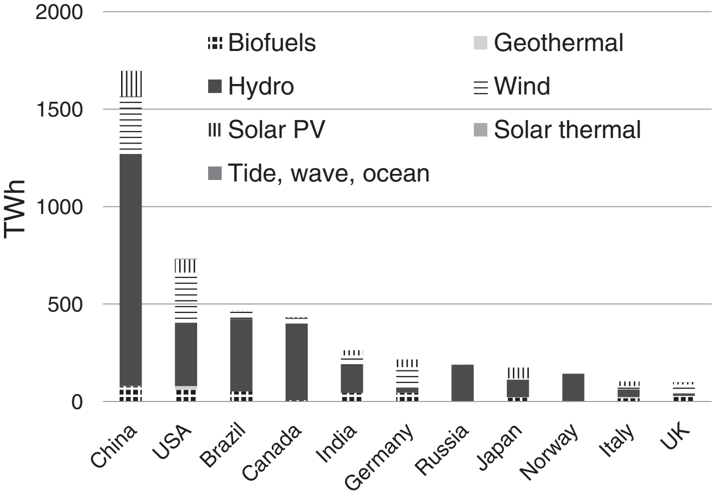

Figure 1.5 Low-carbon electricity generation by source in 2017. Source: Data from explore energy data by category, indicator, country or region (IEA) [38].

1.4.2 Energy and H_2 Supply

Another consideration is the energy required to capture and convert CO_2, which must certainly be derived from renewable sources (Figure 1.5) [38]. If this energy comes from fossil oils, much more CO_2 will be emitted than separated. Fortunately, the renewables now account for over 25% of global power output (hydro: 16%, wind: 5%, PV: 2%), [1] and the costs of PV and wind power become even lower than that of fossil fuels (natural gas and coal) (Figure 1.6) [39]. Thus, electricity from renewable sources can be converted into H_2 by water electrolysis, which can be performed on an industrial scale. Nevertheless, H_2 produced by electrolysis systems (2.5–6 US$/kg$_{H2}$) is at present more expensive than that from

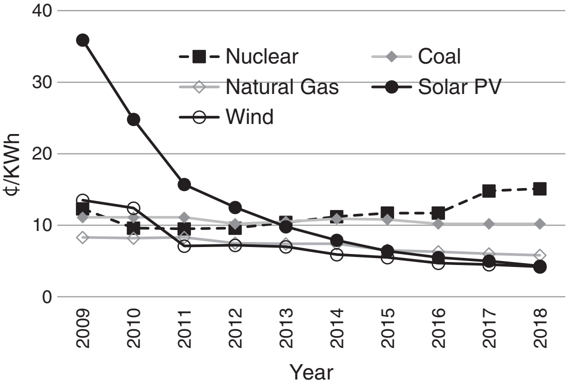

Figure 1.6 Levelized cost of energy comparison: Renewable energy versus conventional generation. Source: Data from Lazard.com, Lazard's levelized cost of energy analysis [39].

current industrial production based on conventional fossil sources, like natural gas reforming and coal gasification (<1 US$/kg H_2) [40, 41].

1.5 Political Aspect: Tax

The future prospects for CO_2 utilization on large scale will mainly depend on policy support. The carbon tax, a fee imposed on the burning of carbon-based fuels (coal, oil, gasoline, and natural gas), is one policy for reducing the use of fossil fuels. To reduce CO_2 emissions, as many as 29 countries have implemented carbon taxes as of 2019. Tax rates, including energy taxes, differed according to use and fuel type in 2017. For example, high tax rates are imposed on gasoline in every country, from y_i^{SOLL} in the United States to $\left(y_i^{SOLL} - y_i^{IST}\right)$ in the Netherlands [42]. On the other hand, there are also significant differences in tax rates for the industrial sector depending on the country (Figure 1.7). An increasing of the tax rates of carbon-based fuels seems to be necessary to motivate our societies to switch to clean energy.

1.6 Conclusion and Perspectives

CO_2 utilization will play a crucial role in achieving the internationally agreed climate and energy goals. In particular, the conversion of CO_2 to fuels and chemicals will be of significant importance. However, these technologies are still in their infancy, and the following issues require consideration and technological improvements:

(i) Supply of H_2 and power from renewables.
(ii) Cost reduction, mainly for the supply of low-carbon H_2.
(iii) Political support to shift from a fossil-based to CO_2-based economy.
(iv) Highly efficient catalysts to minimize energy usage for the valorization of CO_2.

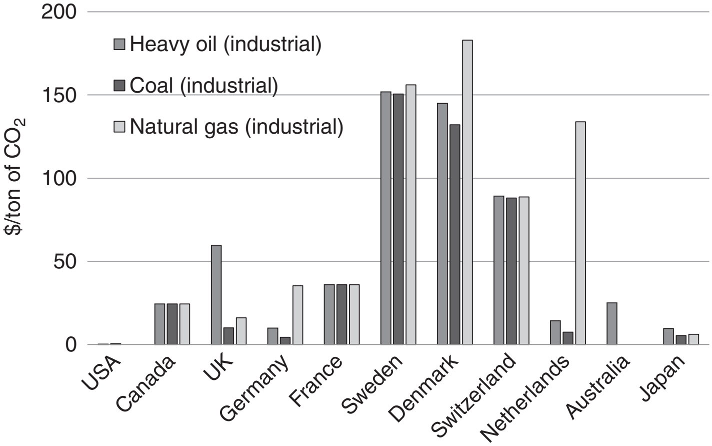

Figure 1.7 Carbon tax rate per ton of emitted CO_2. Source: Data from the Ministry of the Environment of Japan [42].

In addition, critical evaluation from an LCA perspective will be necessary.

In any case, the spread and expansion of renewable energy are essential, which, in turn, require energy storage and transport. CO_2-based fuels produced by CO_2 hydrogenation will contribute to these needs. Therefore, further research into CO_2 hydrogenation is necessary from a standpoint of both fundamental science and application. In this respect, we believe the focus of this book on the hydrogenation/electroreduction of CO_2 to formic acid and methanol as chemicals and fuels using homogeneous and heterogeneous catalysts will be of interest to many scientists. It will serve as motivation for studying the development of catalysts for the hydrogenation of CO_2 as a fuel and bulk chemical. In addition, the challenge of activating unreactive CO_2 will stimulate the curiosity and creativity of chemists.

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