jResource Recovery
January 2023, Volume 1, 1007
www.resrecov.com

Grand Challenge

# Grand Challenges in $CO_2$ Capture and Conversion

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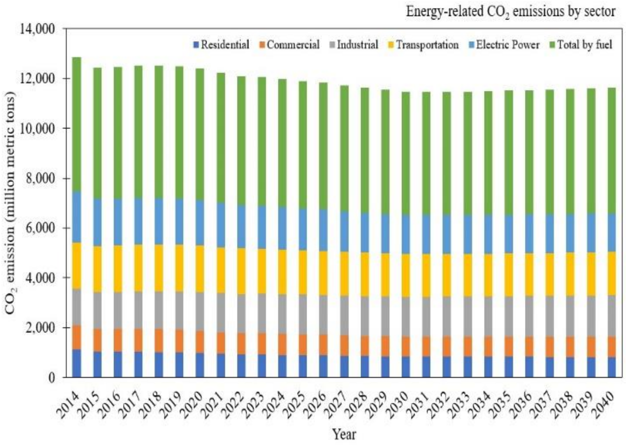
## ABSTRACT

$CO_2$ and its emission control is one of the main challenges in climate change mitigation. There are various methods for $CO_2$ capture, including physical and chemical technologies such as chemical looping, post-combustion, pre-combustion, reduction and bio-technologies. Besides these methods, there are methods to convert $CO_2$ into value-added products. However, both approaches face challenges that limit their commercialization. In this paper, the challenges of $CO_2$ capture and conversion are examined and pros and cons of the methods to remove these obstacles are studied. Here, as a result, four main challenges in $CO_2$ capture and conversion were presented: (1) energy consumption of existing technologies and some alternatives, (2) fixed and operational costs, (3) insufficient activity, sustainability and economics of existing catalysts or microorganisms for $CO_2$ utilization and conversion, and (4) carbon footprint in existing technologies. Also, it was concluded that the need for more reliable life cycle assessment data for zero carbon footprint in existing and future $CO_2$ capture and conversion technologies is one of the most important concerns that should be addressed in future studies to explore creative solutions for this issue.

**KEYWORDS**: $CO_2$ capture and conversion; Climate change; Energy; Costs.

## GRAPHICAL ABSTRACT



## HIGHLIGHTS

- $CO_2$ as a major challenge in climate change.
- $CO_2$ capture and conversion into value-added products.
- Energy, costs, effective catalysts, and carbon footprint as four main challenging issues.

## 1. Introduction

Since the era of industrialization, the amount of carbon dioxide ($CO_2$) emissions has increased and has gradually aggravated climate change (Burkart et al., 2019; Gao et al., 2020). Most $CO_2$ emissions come from the combustion of fossil fuels and can therefore be reduced through the gradual elimination of fossil fuels. However, although fuel eliminations might be possible for the power industry and transportation sector (through substitution with renewable energies), many sectors (e.g., steel/cement production, intercontinental air transport or non-electrical trains) do not have a suitable alternative for carbon-based fuels (Figure 1). Therefore, to achieve net zero greenhouse gas (GHG) emissions, in addition to emission reduction, $CO_2$ capture, and utilization are required in the long term (Castel et al., 2021; Sharifian et al., 2021).

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http://doi.org/10.52547/jrr.2302-1007
Received: 2023-02-27; Revised: 2023-03-28; Accepted: 2023-03-29; Published: xxx

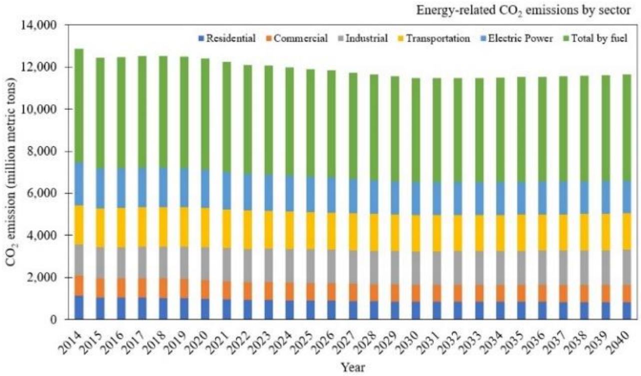

**Fig. 1.** Energy-related $CO_2$ emissions by sector (U.S. Energy Information Administration, 2021).

To combat climate change, an important strategy is to close the anthropogenic carbon cycle through carbon capture and utilization (CCU). Therefore, there is a fundamental need to focus on the effective combination of $CO_2$ capture with further conversion through mineral carbonation, chemical and biological conversion to other chemicals or fuels (Burkart et al., 2019; Gao et al., 2020; Castel et al., 2021). Centralized capture technologies are often grouped into main categories of: (1) chemical looping (combustion), (2) pre-combustion, and (3) post combustion. In addition, $CO_2$ capture technologies, using algae, biochar, charcoal, and nature-based solutions including tree planting (afforestation and reforestation), where $CO_2$ is essentially removed from the atmosphere and is converted into biomass through photosynthesis, has recently been investigated (Sharifian et al., 2021; Galán-Martín et al., 2022).

In general, there are four main challenges in $CO_2$ capture and conversion, including:

1. The main challenge currently limiting $CO_2$ capture and conversion methods is the high energy consumption. Processes such as adsorption and reduction require high energy for adsorbent regeneration and $CO_2$ activation, respectively (Wei et al., 2022).

2. The second challenge is the cost of direct air capture (DAC) through traditional technologies (ranging from $100 to $1000 per tonne of captured $CO_2$) (Sharifian et al., 2021).

3. The development of highly active, stable, and economical catalysts or microorganisms for $CO_2$ utilization is crucial, although difficult and challenging, for practical systems that reduce $CO_2$ emissions (Burkart et al., 2019).

4. CCU's carbon footprint is also an issue (Castel et al., 2021). Carbone neutralizing scenarios are needed in this case. First, advanced separation and sequestration technologies in the short-term development can be developed to mitigate the climate change impact of CCU technologies. Second, substitution of renewable energies or development of fuels or chemicals with equivalent energies and zero emissions instead of the present fossil fuel-based ones is a better scenario that can be considered in the sustainable perspective (Centi and Perathoner, 2023; Chung et al., 2022).

After the $CO_2$ is captured, it can be stored or utilized. Table 1 shows the $CO_2$ products and the relating capture methods. $CO_2$ is an inexpensive, non-toxic, renewable commodity. The $CO_2$ utilization market is projected to grow from 0.23 gigatonnes per year (Gt/year) today to 7 Gt/year by 2030 (Sharifian et al., 2021).

## 2. Challenges in $CO_2$ capture methods

$CO_2$ capture is widely accepted as a vital procedure and presents many challenges (Gao et al., 2020; Gür, 2022). As mentioned before, the most important challenge in CO2capture is the large amount of energy consumption (Kumaravel et al., 2020). For example, in DAC, the energy and material costs of moving large amounts of air through an absorbent structure are also expected to result in high capture costs (Castel et al., 2021).

Separation of $CO_2$ from air, combustion exhausts or other process either from stationary point or distributed sources needs to be investigated and overcome obstacles (Gür, 2022). In general, $CO_2$ capture methods can be classified into chemical and physical categories. Tables 2 and 3 show some challenges in these methods.

**Table 1**
$CO_2$ products and the relating capture methods.

| Technology | | | Production and utilization |
|---|---|---|---|
| $CO_2$ conversion | | | Mineral carbonation (cement and concrete) (Burkart et al., 2019) |
| Industrial synthesis | | | Urea and Salicylic acid (Burkart et al., 2019) |
| Catalytic | | | $C_1$ Products: Carbon monoxide, Methane, Formaldehyde, … (Burkart et al., 2019)<br>Ethylene and Ethanol (Burkart et al., 2019)<br>Cyclic carbonates, Polymers (polycarbonates, …) (Burkart et al., 2019)<br>Hydrocarbons (De Ras et al., 2019)<br>Liquid fuels (Burkart et al., 2019)<br>Higher alcohols (Li et al., 2018)<br>Carboxylic acids (acrylic acid, methacrylic acid, …) (Burkart et al., 2019) |
| Electrochemically | | | Oxalic acid (Burkart et al., 2019) |
| Biological | Photosynthetic | Green algae | Fuels (biodiesel, renewable diesel/gasoline), Protein, Pigment (Burkart et al., 2019) |
| | | Cyanobacteria | Fuels (ethanol, iso-butanol, n-butanol), Fatty acids, Ethylene, … (Burkart et al., 2019) |
| | Non-photosynthetic | Chemo-lithotrophic organisms | Acetogens (Burkart et al., 2019) |
| | | Bioelectrochemical organisms | Succinate, Acetate, Alcohols, Electricity, … (Burkart et al., 2019) |
| | Biorefinery (indirect) | | Biofuels, Bio-materials, Electricity, Biochar, Green hydrogen, … (Galán-Martín et al., 2022) |
| Direct utilization of $CO_2$ | | | Refrigerant, Fire extinguishers, Carbonated beverages, … (Burkart et al., 2019)<br>An excellent working fluid and solvent for many applications such as: power generation, dyeing agent, and food-safe chemical extractions (Valluri et al., 2022) |

**Table 2**
Challenges in chemical $CO_2$ capture.

| **Capture method** | | | **Challenges** |
|---|---|---|---|
| Chemical looping | Oxy fuel | | • High oxygen production energy costs<br>• High sensitivity to air leakage into the system<br>• Although this is an efficient capture method, it is difficult to retrofit as compared to the post-combustion method.<br>• Special materials are needed to resist the high flame temperature (ca. 3500 °C). However, the recycled $CO_2$ can be used to moderate this temperature (Sharifian et al., 2021). |
| | Calcium (Ca) looping | | • Rapid decrease of limestone (i.e., sorbent) capacity after several cycles of reaction with $CO_2$<br>• Environmental concerns due to limestone mining, waste from Ca-looping (i.e., spent calcium oxide (CaO)) and the need for high temperatures for operation<br>• Need for air separation unit to obtain pure oxygen ($O_2$) for calcination (Sharifian et al., 2021). |
| Post combustion | Absorption | | • High energy requirements (Wilberforce et al., 2021)<br>• The limited $CO_2$ absorbing capacity is caused by the reaction stoichiometry and depends on the absorbent type (Sharifian et al., 2021).<br>• Requires a regeneration step (Castel et al., 2021) |
| | | Amine | • Low $CO_2$ loading capacity of the solvent<br>• Amine degradation in the presence of nitrogen oxides ($NO_x$), sulfur oxides ($SO_x$), $O_2$, and particulate matter<br>• Corrosion in amine equipment causes absorption column and stripper components to degrade over time.<br>• The solvent cannot be completely regenerated.<br>• The recovery step is energy intensive and the waste stream can be hazardous (Spigarelli and Kawatra, 2013). |
| | | Ammonia | • The flue gas must be cooled to 15–27 °C due to volatility of ammonia.<br>• High losses of ammonia vapor during stripping (Spigarelli and Kawatra, 2013) |
| | Adsorption | | • Adsorbent degradation in cyclic operation<br>• Possible decrease in adsorption capacity of the adsorbent after the desorption step (Sharifian et al., 2021)<br>• Cyclic process requires regeneration (Castel et al., 2021). |
| | | Activated carbons | • Low $CO_2$ capacity in mild conditions<br>• The wide variety of raw materials means that a wide range of pore characteristics is often seen between adsorbents.<br>• Negatively affected by $NO_x$, $SO_x$, and water ($H_2O$) (Spigarelli and Kawatra, 2013) |
| | | Amine functionalized | • Degrades at a temperature of about 100 °C<br>• Irreversible reactions with $NO_x$ and $SO_x$ produce unwanted byproducts<br>• A temperature swing approach is required for desorption (Spigarelli and Kawatra, 2013). |
| Absorption | Solvay process | Dual alkali | • High energy demand of calciner and $CO_2$ production from calcination for large-scale $CO_2$ capture (Spigarelli and Kawatra, 2013) |
| $CO_2$ reduction technology (Albo et al., 2021) | Electrochemical reduction (Albo et al., 2021) | | • Improved and/or novel electrocatalytic materials<br>• Improved/novel electrolytes<br>• More efficient electrocatalytic reactors (Albo et al., 2021) |
| | | High-temperature molten carbonate cells | Difficult operating conditions due to:<br>• High temperatures<br>• Corrosion<br>• Sensitivity to the presence of $SO_x$ in the gas mixture (Sharifian et al., 2021) |
| | | Redox active carriers and electrode reactions | • Limitation in terms of both solvents and carriers<br>• Difficult finding a solvent that is inexpensive, safe and electrochemically stable<br>• Difficult to allow high solubility of redox species (Sharifian et al., 2021) |

**Table 2**
*Continued.*

| | | | |
|---|---|---|---|
| | | Proton coupled electron transfer (PCET) active agents | • Slow electrode kinetics<br>• Low solubility of PCET organics<br>• The sensitivity of the process to the impurities in the flue gas such as $O_2$, water and sulfur (Sharifian et al., 2021) |
| | Photochemical reduction | | • Innovative photoactive materials<br>• Efficient photocatalytic reactors (Albo et al., 2021) |
| | Photoelectrochemical reduction | | • Development of photoactive materials<br>• Efficient photocatalytic reactors (Albo et al., 2021) |

**Table 3**
Challenges in physical $CO_2$ capture.

| Situation | Capture method | Technology | Challenges |
|---|---|---|---|
| Post-combustion | Membrane | | • Low concentration of $CO_2$ in fuel gas leads to higher energy consumption (Wilberforce et al., 2021).<br>• Sensitivity to moisture (i.e., lower selectivity) (Sharifian et al., 2021)<br>• The high temperature of flue gas degrades organic membranes. The gas must be cooled below 100 °C.<br>• Membranes must be resistant to flue gas impurities, aging and plasticization (hardening).<br>• Single-stage membrane systems are not capable of high $CO_2$ capture efficiency; a second stage is required (Spigarelli and Kawatra, 2013).<br>• The low concentration of $CO_2$ in the fuel gas leads to high separation energy and the need for membranes with high selectivity. Therefore, it is not economical in terms of scale (Castel et al., 2021; Wilberforce et al., 2021; Spigarelli and Kawatra, 2013).<br>• Trade-off between permeability and selectivity in polymeric membranes (Sandru et al., 2022)<br>• Ion exchange extra energy and membranes require thermal stability is a limitation at high temperature (Sharifian et al., 2021). |
| | Adsorption | Zeolite | • The presence of impurities ($NO_x$, $SO_x$ and $H_2O$) significantly affects performance.<br>• It is time and energy consuming for complete regeneration as a temperature swing approach is required (Spigarelli and Kawatra, 2013). |
| | | Metal-organic frameworks (MOFs) | • Negatively affected by $NO_x$, $SO_x$, and $H_2O$<br>• Low $CO_2$ selectivity in carbon dioxide/nitrogen ($CO_2/N_2$) gas streams<br>• Lack of experimental data on performance after multiple adsorption/desorption cycles<br>• Pressure and temperature swing desorption approaches have not been adequately investigated (Spigarelli and Kawatra, 2013).<br>• By increasing the porosity of MOFs, their mechanical stability is compromised.<br>• The instability of MOFs may cause a phase change in them.<br>• The cost of $CO_2$ capture through MOFs is high (Younas et al., 2020). |
| | Cryogenic distillation | | • Very high energy requirement for DAC<br>• The water content of the feed stream must be removed to avoid equipment clogging due to ice formation.<br>• Buildup of solid $CO_2$ reduces the efficiency of the evaporator over time.<br>• High capital cost of equipment<br>• High cost of the refrigerant used to cool the system (Spigarelli and Kawatra, 2013) |
| Pre-combustion | Absorption | Selexol process | • Process is most efficient at elevated pressures (Spigarelli and Kawatra, 2013). |
| | | Rectisol process | • The solvent is capable of absorbing trace metal compounds, such as mercury to form amalgams at low operating temperatures.<br>• Solvent cooling leads to high operating costs.<br>• Complex operating scheme leads to high capital costs (Spigarelli and Kawatra, 2013). |
| | | Fluor process | • High solvent circulation rates (increasing operating cost)<br>• Solvent cost (Spigarelli and Kawatra, 2013) |

**Table 3**
*Continued.*

| | | |
|---|---|---|
| | Purisol process | • Additional compression is required after the water-gas shift reaction (Spigarelli and Kawatra, 2013).<br>• Retrofitting of existing plants is expensive and more difficult compared to oxy-fuel and post-combustion.<br>• Syngas must be dried before $CO_2$ capture.<br>• For non-gaseous feed stocks (e.g., coal or crude oil) the syngas stream must be cleaned of impurities in the gasification material.<br>• The integrated gasification combined cycle (IGCC) system has high investment and operational costs (Spigarelli and Kawatra, 2013). |

Note that many of the physical solvents used in pre-combustion $CO_2$ capture can be used in post-combustion $CO_2$ capture if the gas stream is properly clean and pressurized, so the challenges are the same (Spigarelli and Kawatra, 2013).

## 3. Challenges in $CO_2$ conversion methods

### *3.1. Challenges in conventional $CO_2$ conversion methods*

The production of synthetic hydrocarbon fuels and chemicals often requires high purity $CO_2$ feeds (Sharifian et al., 2021). In these carbon-based fuels, $CO_2$ is ultimately released into the atmosphere elsewhere in the value chain (Galán-Martín et al., 2022).

#### *3.1.1. Membrane technology*

The increased purity (selectivity) obtained by the high-performance membrane material is associated with significantly lower productivity (permeability) (Ebadi Amooghin et al., 2019; Ebadi Amooghin et al., 2022; Bandehali et al., 2021; Mashhadikhan et al., 2021; Nematolahi et al., 2022). The increase in selectivity in fact systematically induces the need for a larger membrane surface area due to the faster decrease in driving force with increasing permeate purity (Castel et al., 2021).

#### *3.1.2. Sorbent*

In high-temperature solid sorbents for $CO_2$ capture (in pre-combustion applications), several challenges arise at the adsorbent, reactor, and system scales (Gao et al., 2020).

#### *3.1.3. Heterogeneous catalyst*

For heterogeneous catalyst, there are important challenges related to the development of practical catalytic systems for $CO_2$ utilization. These challenges include the limited range of products due to their thermodynamic stability, the large energy barriers due to their kinetic stability, and the restraining by the produced water of the hydrogenation reaction (Burkart et al., 2019).

### *3.2. Challenges in $CO_2$ conversion through reduction methods*

The major problem with many systems using this technology is that only a small amount of $CO_2$ (< 20%) is converted to product and the $CO_2$ reacts with hydroxide ($OH^-$) in the electrolyte to form carbonates (Burkart et al., 2019).

#### *3.2.1. Photoelectrochemical technology*

The following challenges must be addressed for large-scale production through photoelectrochemical (PEC) $CO_2$ conversion (Kumaravel et al., 2020):

*• Stability*

Some inexpensive electrode materials are unstable and scale up procedures are expensive.

*• Light absorption*

Some additional studies are needed for precious metals to improve selectivity and product efficiency.

*• Yield and selectivity*

1. The main issue is bond-breaking/making reactions at the electrode−electrolyte interface.

2. Ionic liquids are one of the promising electrolytes with high product selectivity, but they are very expensive.

### *3.3. Hybrid methods*

#### *3.3.1. MOF-Catalyst*

• For direct solid-gas phase conversion of $CO_2$, the selectivity of the MOF-based catalyst should be further improved because water and $N_2$ can compete with $CO_2$.

•The instability of MOFs in the presence of SOx and NOx (Younas et al., 2020).

#### *3.3.2. Photo-Catalyst*

Designing a photo-catalyst active in visible light and at a same time having favorable properties for the selective reduction of $CO_2$ remains a challenge (Gao et al., 2020).

## 4. Concluding remarks

Grand challenges in $CO_2$ capture and conversion were investigated. Challenges in chemical and physical $CO_2$ capture methods were discussed and summarized in Tables 2 and 3. Moreover, conventional, advanced, and hybrid $CO_2$ conversion methods and the related challenges were studied. In general, the four main challenges in $CO_2$ capture and conversion that need to be solved are: (1) the energy consumption of existing $CO_2$ capture technologies, especially at low $CO_2$ concentrations, (2) fixed and operating costs, (3) the need to development of highly active, stable, and economical catalysts or microorganisms for $CO_2$ utilization, and (4) carbon footprint in carbon capture and utilization technologies.

Along with the research in the development of existing technologies for $CO_2$ capture and conversion, as a perspective for future studies, the life cycle assessment and value chain analysis in the separation, reuse or conversion of $CO_2$ as a carbon source, need further investigations. It is recommended to further study the engineering aspects of the carbon footprint of $CO_2$ capture and conversion technologies to explore creative solutions to the issue.

| Abbreviations | Definition |
|---|---|
| Ca looping | Calcium looping |
| CaO | Calcium oxide |
| CCU | Carbon capture and utilization |
| $CO_2$ | Carbon dioxide |
| DAC | Direct air capture |
| GHG | Greenhouse gas |
| Gt/year | Gigatonnes per year |
| $H_2O$ | Water |
| IGCC | Integrated gasification combined cycle |
| MOFs | Metal organic frameworks |
| $N_2$ | Nitrogen |
| NOx | Nitrogen oxides |
| $O_2$ | Oxygen |
| $OH^-$ | Hydroxide |
| PCET | Proton coupled electron transfer |
| PEC | Photoelectrochemical |
| SOx | Sulfur oxides |

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