



Indoor CO₂ direct air capture and utilization: Key strategies towards carbon neutrality

L.R. López^a, P. Dessì^{a,b}, A. Cabrera-Codony^{a,*}, L. Rocha-Melogno^c, N.J.R. Kraakman^{d,e}, M. D. Balaguer^a, S. Puig^a

^a LEQUiA, Institute of Environment, University of Girona, Girona, Campus Montilivi, Carrer Maria Aurèlia Capmany 69, Girona, Catalonia, Spain

^b Department of Agricultural Sciences, University of Naples Federico II, Via Università 100, 80055, Portici, Italy

^c ICF, 2635 Meridian Parkway Suite 200, Durham, NC, 27713, United States

^d Jacobs Engineering, Temple Quay 1, Bristol, BAS1 6DG, UK

^e Institute of Sustainable Processes, University of Valladolid, Dr. Mergelina s/n., Valladolid, 47011, Spain

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ABSTRACT

Direct air capture (DAC) is a promising technology that can help to remove carbon dioxide (CO₂) from the air. One application of DAC is indoor CO₂ direct air capture (iCO₂-DAC). A wide range of materials with unique properties for CO₂ capture have been investigated, including porous materials, zeolites, and metal-organic frameworks. The selection of suitable materials for iCO₂-DAC depends on several factors, such as cost, CO₂ adsorption capacity, and stability. The development of new materials with improved properties for iCO₂-DAC is an active research area. The captured CO₂ can serve as a renewable carbon source to produce biofuels for internal use (e.g., for heating purposes), decreasing the environmental impact of buildings. This review article highlights the importance of iCO₂-DAC to improve indoor air quality in buildings and boost the circular economy. We discuss the available carbon capture technologies and materials, discussing their properties and focusing on those potentially applicable to indoor environments. We also provide a hypothetical scenario where CO₂ is captured from different indoor environments and transformed into sustainable fuels by using an emerging carbon capture and utilization technology (microbial electrosynthesis). Finally, we evaluate the economic feasibility of such an innovative approach in comparison to the use of traditional, fossil-based fuels.

1. Introduction

1.1. Motivation: Improve indoor air quality through indoor carbon dioxide recycling

Indoor environments play a crucial role in modern society by providing shelter from unpleasant and unhealthy outdoor conditions (European Commission, 2003). However, epidemiologic evidence indicates that indoor air pollutants (IAPs) are often present at higher concentrations than outdoor air pollutants (Leung, 2015; Meng et al., 2020). Enclosed spaces, such as public buildings, underground transport, and office buildings, may have a very limited fresh air supply, leading to harmful IAPs levels (Sadriazadeh et al., 2022). The most significant IAPs include particulate matter (PM), biological organisms (fungal spores, bacteria, and viruses), volatile and very volatile organic compounds (VOCs and VVOCs, respectively), and volatile inorganic

compounds (VICs) (González-Martín et al., 2021).

The coronavirus pandemic has significantly increased public awareness of the effects of poor indoor air quality (IAQ). Even before the COVID-19 pandemic, IAQ was an important public health concern, considering that most people spend 80–90% of their time indoors (Boor et al., 2017; Gibson et al., 2019; Kim et al., 2019; Sarkhosh et al., 2021). This means that 80–90% of the average 250 million liters of air that persons breathe during their lifetime are sourced from indoor environments such as houses, workplaces, schools, and transport vehicles (Kraakman et al., 2021; Luengas et al., 2015). Exposure to IAPs can exacerbate a variety of adverse health effects, ranging from mild irritations (Paleologos et al., 2021) to severe diseases affecting the endocrine system (Rudel and Perovich, 2009), the reproductive system (Veras et al., 2010), and the central nervous system (H. Kim et al., 2020). In addition to health problems, exposure to poor IAQ can also impair cognitive performance (Du et al., 2022; Hu et al., 2022; Zhang et al.,

* Corresponding author

E-mail address: alba.cabrera@udg.edu (A. Cabrera-Codony).

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2017).

Removing or minimizing IAPs is a crucial strategy to prevent negative health effects (Allen et al., 2015). However, certain IAPs cannot be limited, such as those related to human metabolism and activities (Zou et al., 2020). Natural or mechanical ventilation can provide adequate air exchange rates to maintain good IAQ. However, ventilation, along with heating and/or cooling, requires energy, contributing to global greenhouse gas (GHG) emissions (Che et al., 2019). The Sixth Assessment Report (AR6) of the Intergovernmental Panel on Climate Change (IPCC) highlights the need of mitigating emissions from the building sector (IPCC, 2022). Enhancing IAQ and ensuring thermal comfort, along with reducing the carbon footprint of buildings, would result in substantial health and economic benefits.

Green buildings are specifically designed to minimize their environmental impact and must fulfil five essential elements, as depicted in Fig. 1., which include sustainable site design, water conservation, and quality, energy and environment, indoor environmental quality (IEQ), and conservation of materials and resources. The five essential elements are defined as follows (Alam and Haque, 2016): 1) Sustainable Site Design stands for the minimization of urban sprawl and needless destruction of valuable land, habitat, and green space, which results from inefficient low-density development; 2) Water Conservation and Quality is the preservation of the existing natural water cycle and design of site and building improvements to closely emulate the site's natural "pre-development" hydrological systems; 3) Energy and Environment stands for the minimization of adverse impacts on the environment through optimized building siting, optimized building design, material selection, and aggressive use of energy conservation measures; 4) Indoor Environment Quality aims to provide a healthy, comfortable, and productive indoor environment for building occupants and visitors; 5) Conservation of Materials and Resources aim to minimize the use of non-renewable construction materials and other resources such as energy and water through efficient engineering, design, planning, and construction debris generation. Furthermore, in the pursuit of addressing climate change and enhancing IAQ, IAPs such as CO₂ can be captured, and ideally transformed into sustainable biofuels for internal use, displacing non-renewable fuels used, e.g., for heating the buildings. to be used in-situ to minimize the inputs of energy and matter. This

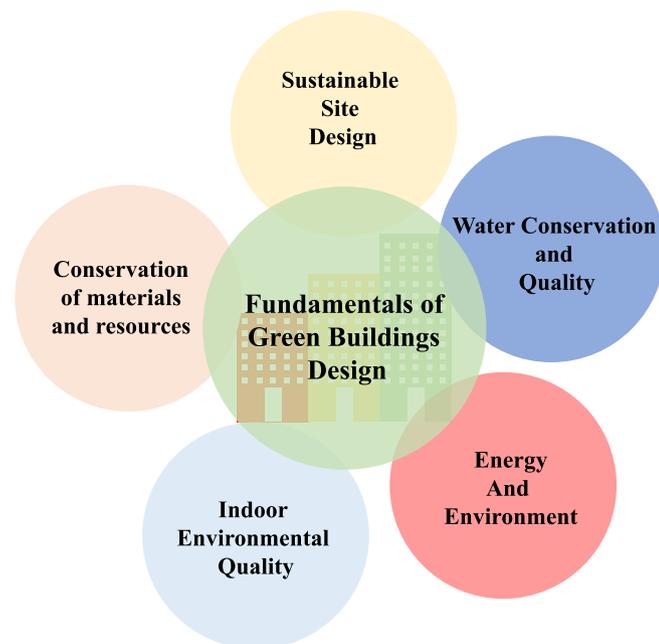


Fig. 1. Fundamental elements of green buildings design. Adapted from (Ragheb et al., 2016). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

approach is in line with two pillars of green building development: which are providing a healthy, comfortable, and productive indoor environment for building occupants and visitors, and as well as reducing the use of non-renewable fuels.

An example of a circular model that includes IAP capture and conversion, against the linear model (take-use-waste) commonly applied until now, is depicted in Fig. 2. In the circular approach, energy savings derive from the reduction of ventilation to control indoor CO₂ concentration, as well as the use of in-situ produced biofuels.

Carbon capture and utilization (CCU) technologies have the potential to convert CO₂ to biofuels, but have been conceived for large punctual CO₂ emission sources (Valentić et al., 2016; Yang et al., 2021). However, it is estimated that between one-third and one-half of CO₂ emissions originate from distributed sources such as commercial and industrial buildings and small sources like transportation vehicles (Ghiat and Al-Ansari, 2021; Rossing and Chiaverina, 2019). Thus, CO₂ direct air capture (CO₂-DAC), where CO₂ is captured directly from the atmosphere, is gaining interest. Indoor CO₂ DAC (iCO₂-DAC), where CO₂ is captured from indoor environments, is thermodynamically favourable than outdoor DAC, as indoor CO₂ concentrations, typically 1000–3000 ppm (López et al., 2022; Shen et al., 2020) with peaks up to 6000 ppm (Cao et al., 2019; Hwang et al., 2017; Park and Ha, 2008) are significantly higher than atmospheric CO₂ concentration (415 ppm) (IPCC, 2022). This review aims to assess the potential of iCO₂-DAC as a renewable carbon source. We present the technological state-of-art of iCO₂-DAC technologies, including the main characteristics, limitations, and future perspectives for the application in the field of such technology. It is important to highlight that a final selection or recommendation of the best iCO₂-DAC technology is not provided in this work, as this selection strongly depends on the type of indoor environment (indoor CO₂ concentration, type of ventilation, air humidity, availability of services), instead critical information to take this decision is provided. Finally, we present a case study examining indoor CO₂ recycling from three different environments: high schools, office buildings, and metro cabins.

1.2. Chronological development of indoor CO₂ direct air capture (iCO₂-DAC)

Air purification was introduced in the 1940s and 1950s for use in indoor environments such as submarines and spacecrafts (Sanz-Pérez et al., 2016a; Satyapal et al., 2001; Tepe and Dodge, 1943). However, the introduction of iCO₂-DAC is linked to the development of technologies and materials for atmospheric CO₂-DAC, (Lackner et al., 1999). Originally, the cost of CO₂-DAC was argued to be prohibitively high (Herzog, 2003) as a result of the extremely dilute nature of atmospheric CO₂ (Bui et al., 2018). However, recent reports demonstrated that CO₂-DAC has sufficient technical maturity for economic feasibility (Keith et al., 2018). CO₂-DAC plants are operating worldwide, and at least 130 CO₂-DAC plants are now at various stages of development, where the primary industrial developers are Carbon Engineering (Canada), Climeworks (Switzerland), and Global Thermostat (USA) (IEA, 2023). This significant advance in the field of CO₂-DAC technologies has been achieved thanks to studies on sorbent selection (Azarabadi and Lackner, 2019; Goepfert et al., 2012a; Sanz-Pérez et al., 2016a), unit operation design (Lackner, 2013; Wurzbacher et al., 2016; Zhang et al., 2014a) or basic process development (Bretherton and Bretherton, 1961; Goepfert et al., 2012a; Mazzotti et al., 2013). Nowadays, two major technologies are considered for CO₂-DAC on a commercial scale: absorption using alkaline solutions, and adsorption using amine-functionalized solid sorbents (Schellevis et al., 2021).

In the last decade, significant advances were made in understanding the relationship between adsorption and desorption characteristics of silica-organic hybrid adsorbents as function of important parameters such as temperature, flow rate, polyethyleneimine (PEI) concentration and PEI molecular weight (Goepfert et al., 2014). Together with

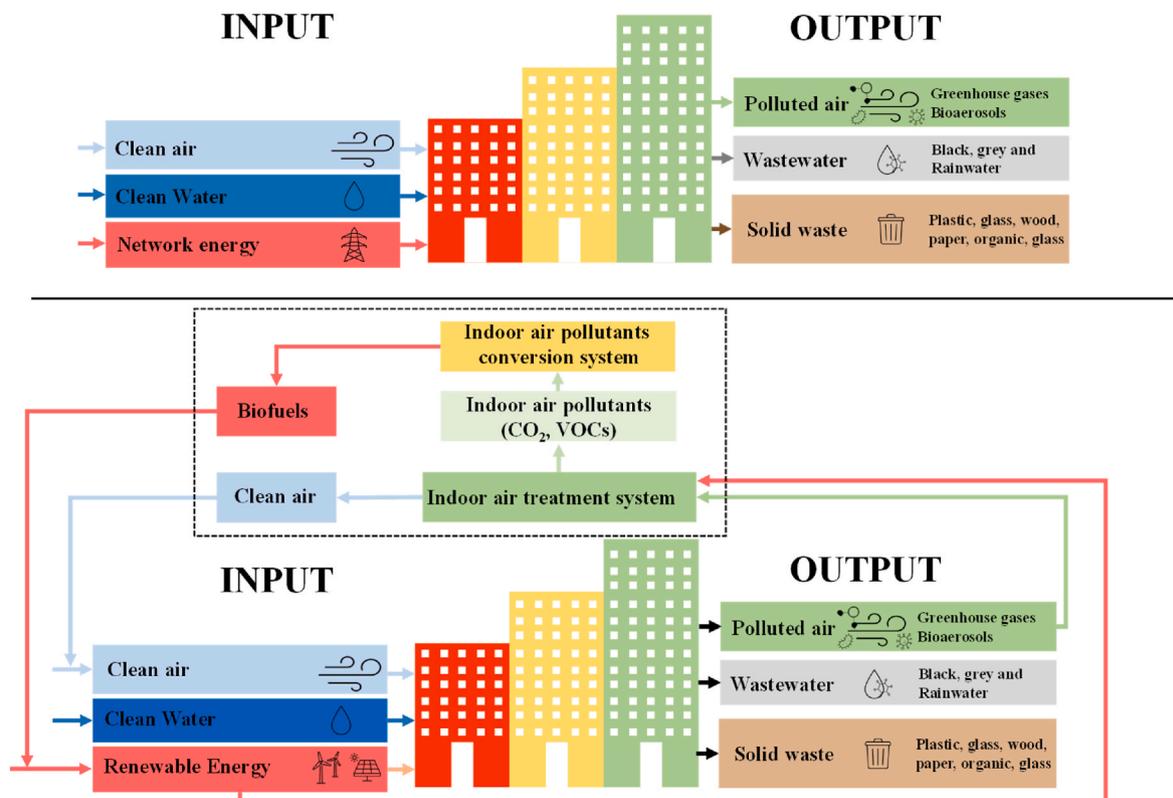


Fig. 2. Coupling of green building fundamentals and IAPs recycling. Indoor CO₂-Direct air capture (iCO₂-DAC) as a strategy to improve IAQ and building's energy efficiency. The top diagram of Fig. 2, shows how current buildings following a linear economic model of take-use-waste, are producers of significant amounts of waste such as wastewater, solid waste, and polluted air. The bottom diagram of Fig. 2 demonstrates the potential of IAPs recycling as a strategy to mitigate gaseous emissions and as a strategy to minimize energy consumption by means of the coupling of technologies capable of transforming IAPs into biofuels that can be consumed in situ, minimizing the external energy inputs of the buildings. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

increasing awareness and understanding of the effects of CO₂ in indoor environments (López et al., 2023) and the increasing need to optimize energy utilization related to ventilation needs to reduce indoor CO₂ and improve IAQ (Beaumont, 2022), promoted the development of modern technologies based on solid-supported amines for iCO₂-DAC (Baus et al., 2023; Baus and Nehr, 2022). Studies carried out by Baus and Nehr (2022), demonstrated through measurements and simulation results that the coupling of Heating, Ventilation, and Air Conditioning (HVAC) and CO₂-DAC in recirculation mode not only improved IAQ, but also provided energy savings through reduced air conditioning requirements enabled by HVAC/DAC-coupling (Baus and Nehr, 2022). In the same direction López et al. (2023), pointed out the potential use of captured indoor CO₂ as renewable carbon source in order to mitigate climate change. As suggested by Modak et al. (2020), technologies developed for atmospheric CO₂-DAC has a great potential of implementation in enclosed environments, which has remained unexploited to date. iCO₂-DAC technologies can only be implemented on a broad scale if CO₂-selective, abundant, sustainable, and low-cost materials are developed toward the conditions and limitations of the building indoor applications.

1.3. Utilization of Indoor CO₂ direct air capture (iCO₂-DAC)

After being captured, CO₂ can be stored, used for enhanced oil recovery, or converted into valuable products such as chemicals, fuels, and cement. Depending on the regeneration techniques used, CO₂ can be recovered in different concentration flows. Lower CO₂ concentration flows can be utilized in applications such as greenhouses, algae cultivation for food or fuel, or microbial cultivation to produce an edible protein (Elfving et al., 2021). Previous studies have identified the

advantages and disadvantages of CO₂-DAC compared to conventional CCS and CCU technologies (Boot-Handford et al., 2014; Goepfert et al., 2012b; Jones, 2011; López et al., 2022; Murdock et al., 2016; Sayari et al., 2016; Socolow et al., 2011). CO₂-DAC is a challenging process that still needs to overcome several limitations to enhance its versatility and applicability. These limitations include the range of suitable locations for implementation, developing new materials, and establishing smart business cases to reduce operating costs (Beaumont, 2022). While CO₂-DAC is not restricted to specific locations, it does require sources of electrical or thermal energy, as well as water. Additionally, the concentrated CO₂ captured needs to be stored in an appropriate outlet storage site (Jones, 2011), such as deep geological formations (Bui et al., 2018; Keith et al., 2018), where the environmental conditions promote mineral carbonation (Gadikota and Park, 2015; Woodall et al., 2019). However, in 2023, as the urgency to minimize or to completely stop the extraction of carbon from the geosphere, it is important to view the CO₂ captured by means of CO₂-DAC as a renewable carbon source rather than a waste to be stored underground.

Another important drawback of CO₂-DAC is the thermodynamic challenge of capturing CO₂ from air at such low concentrations (Lackner, 2013; Lively and Realff, 2016). In order to capture comparable amounts of CO₂ to flue gas processes, large air volumes need to be processed (Jones, 2011). However, this drawback can be mitigated if the CO₂-DAC process is installed in environments with considerably higher CO₂ concentrations than atmospheric, such as indoor environments. Performing iCO₂-DAC can reduce the volume of gas that must be processed, thereby lowering the contactor size and operating costs. Increasing focus has been placed on iCO₂-DAC as a tool to minimize the health impact of indoor CO₂ concentration in environments such as office buildings by coupling the CO₂-DAC process into Heating,

Ventilation, and Air Conditioning (HAVC) systems (Han et al., 2016; Hu et al., 2017; López et al., 2022; Sodiq et al., 2023). This technology stabilizes CO₂ concentrations, as well as humidity levels, inside buildings allowing a higher indoor air recirculation rate, limiting the in-take of fresh air, and reducing the energy input and associated costs on the HVAC system. Hence, HVAC/DAC-coupling in recirculation mode provides energy savings of 20–40% through reduced air conditioning requirements (Beaumont, 2022). Furthermore, the CO₂ captured from the building exhaust air is a renewable carbon source that can be transformed into energy or materials (Baus and Nehr, 2022). Assessing the suitability for integration with buildings HVAC systems and operations are out of the scope of this work. Further information about the potentials and limitations of DAC technologies in the built environment can be found elsewhere (Baus and Nehr, 2022)

2. Materials and methods

This review article adopted a four-step methodology to identify the existing literature relevant to the topic as described in Oloruntobi et al. (2023). Documents focusing on technologies and materials to perform iCO₂-DAC were considered relevant. Scopus, Google Scholar, and Web of Science databases were used as search engines. Then, two main core selection criteria were defined: year of publication (from 2010 to 2023) and topic relevance, selecting articles that explicitly discuss iCO₂-DAC in the context of green buildings, climate change, and circular economy. Some exceptions were made based on literature relevancy. This is the case of highly cited or highly relevant articles that were published before January 2010.

Once the selection criteria were defined, the retrieved documents went through scientific quality evaluation. This consisted of reviewing the scientific quality of the selected studies and evaluating the content for inclusion or exclusion for written in English and from peer-reviewed, conference proceedings and official reports of international entities, such as the European Union and European Commission, were included.

The third step in the methodological process to prepare this review article consisted of the extraction and analysis of the information from the selected articles. This step involved data extraction, analysis, and synthesis of data. The information extracted was information pertinent to fulfil and answer the research objective of this review. Information extracted included important details on the methods, execution, and results of research works in the literature that fulfilled the inclusion criteria. The final goal of the collected information from the selected documents was to synthesize new knowledge and information in the field of indoor CO₂-Direct Air Capture. The review process resulted in the selection of 195 documents that met the inclusion criteria. The final step consisted of applying the so-called “snowball method” to analyse the references within review articles to determine which references were more pertinent to the review of the literature but were not found in the databases searched. This added eleven additional references, resulting in a final selection of 206 references.

3. Suitable materials to perform iCO₂-DAC

Beuttler et al. (2019) defined DAC as a range of technological solutions to extract CO₂ from ambient air at any location on the planet. In particular, the purpose of DAC technologies is to capture CO₂ from air and produce a more concentrated stream of CO₂ for storage or utilization (McQueen et al., 2021). Unlike CO₂ capture from flue-gas, DAC does not operate in the presence of high levels of contaminants (SO_x, NO_x, and mercury), as it occurs in many industry emissions, and does not aim to achieve near-complete CO₂ removal.

As previously mentioned in section 1.2, there are two major technologies are considered for DAC on commercial scale (Schellevis et al., 2021): absorption using alkaline solutions where aqueous KOH is the capture medium and a calcium caustic loop is used to recover CO₂ (Keith et al., 2018; K. Lackner et al., 1999), and adsorption using

amine-functionalized solid sorbents, in which CO₂ reacts with amine-groups on the internal surface of the sorbent and then CO₂ is recovered by a temperature and/or vacuum swing (Bos et al., 2019; Elfving et al., 2021). The majority of DAC studies with sorbents have focused on solid-supported amine materials (Sanz-Pérez et al., 2016a; Schellevis et al., 2021). Aqueous solvent-based approaches typically require high temperatures (over 800 °C) and an oxygen atmosphere to recover CO₂ as calcium carbonate, and to regenerate the alkaline sorbent (Williams and Custelcean, 2020). Furthermore, due to the low CO₂ concentration in air, the energy penalty caused by the latent and sensible heat required to desorb CO₂ from the alkaline medium is higher than from solid sorbents (McQueen et al., 2021; Zhang et al., 2016). DAC using amine-specialized sorbents is less energy intensive, as the desorption process can be carried out at milder temperatures (below 110 °C) (Goepfert et al., 2012a, 2014). The mild operating conditions of amine-specialized sorbents makes them suitable for capturing CO₂ from enclosed spaces such as submarines (Carey et al., 1983; K. Lackner et al., 1999) and spaceships (Carey et al., 1983; Satyapal et al., 2001), which opens the door to explore the application of CO₂ DAC into other types of indoor spaces. As suggested by Modak et al. (2020), DAC has a wide scope for implementation in enclosed environments, which has remained unexploited to date. Although, details about technical (Al-Absi et al., 2022; Goepfert et al., 2012b, 2014; Sanz-Pérez et al., 2016a) and economic feasibility of CO₂-DAC need to be studied further to increase the implementation of CO₂ DAC technologies into indoor environments, further information about technical and economic feasibility can be found elsewhere (Chen and Tavoni, 2013; Daniel et al., 2022; Socolow et al., 2011; T. Wang et al., 2013; Zhang et al., 2014a).

About the operating costs of the CO₂-DAC process range from 94 to 232 \$/t-CO₂ (Keith et al., 2018), considerably higher than conventional CCS technologies. The high energy footprint of CO₂-DAC is principally related to the energy associated with adsorbent materials regeneration. To understand how iCO₂-DAC can be made affordable, it is essential to assess the differences between CO₂-DAC and CCS from punctual sources.

Extraction and concentration of CO₂ from gas streams (concentrated or diluted) are commonly the first steps of CCS, followed by compression, transportation, and finally sequestration in a permanent storage. Approximately 75% of the total cost falls into compression and transportation (Jones, 2011; Xu et al., 2005). In CO₂-DAC, the low CO₂ concentrations and the necessity to operate close to room temperature and ambient pressure rule out most separation and concentration technologies used in CCS (Goepfert et al., 2012b; Jones, 2011). For example, most physical CO₂ adsorbents, commonly used in CCS, such as zeolite (13 X and 5A), activated carbon, mesoporous silica, and metal-organic frameworks (MOFs), require large pressure and/or temperature gradient between the adsorption and desorption stages to enable efficient CO₂ capture and complete desorption (Alessandro et al., 2010; Sayari and Belmabkhout, 2010). Zeolites are typically employed at elevated pressures (above 2 bar), and the regeneration of the material is typically carried out at very high temperatures exceeding 300 °C (Alessandro et al., 2010). Activated carbon, MOFs, and zeolites are not efficient materials in CO₂-DAC due to their low heat of adsorption that leads to shallow adsorption isotherms with low adsorption capacities, and perform poorly at low CO₂ partial pressures (Choi et al., 2009; Zhang et al., 2014a). Thus, while suitable to treat industrial gases containing 10–30% CO₂, such as those released in cement and steel production plants (Dessi et al., 2021), their capacity and selectivity drops when treating low-concentration CO₂ streams. In addition, these types of adsorbent can easily adsorb moisture and other gases. Water competes with CO₂ for binding and leads to an overall decrease in the number of reactive sites (Murdock et al., 2016; Sayari et al., 2016). As a result, several stages of separation are required to get a pure CO₂ stream, resulting in high operating costs in practical CO₂-DAC applications (Xu et al., 2005).

In contrast, chemisorbent materials have demonstrated higher performance at low adsorbate pressure, making them better suitable for

CO₂-DAC (Sayari et al., 2016; Zhang et al., 2016). The sorbent-based iCO₂-DAC processes are described in the following sections. Alternative approaches such as anionic exchange, permeable or semipermeable membranes, ionic liquids, electrochemical approach, and mineral carbonation, (Goepfert et al., 2012b; Murdock et al., 2016; Stern et al., 2013; Yuan et al., 2016) are out of the scope of this review.

3.1. Inorganic chemisorbents for CO₂-Direct air capture (CO₂-DAC)

Inorganic chemisorbents can be divided into inorganic chemisorbents in solution, also known as aqueous hydroxide sorbents, and solid inorganic chemisorbents, also known as solid alkali carbonates.

3.1.1. Aqueous hydroxide sorbents

Aqueous hydroxides are one of the earliest and most widely studied sorbents for CO₂ DAC (Goepfert et al., 2011; K. S. Lackner et al., 1999; Stolaroff and Keith, 2008; Zeman, 2007). Already in the 1940s and 1950s, the feasibility of extracting CO₂ from diluted gases, such as air, through aqueous basic absorption processes has been reported (Blum

et al., 1952; Tepe and Dodge, 1943). In CO₂-DAC with aqueous hydroxide sorbents, an aqueous solution of sodium hydroxide (NaOH) or calcium hydroxide (Ca(OH)₂) is used to skim CO₂ from ambient air (Mikulčić et al., 2019). NaOH and Ca(OH)₂ have high binding energy with CO₂, leading to the formation of either sodium carbonate (Na₂CO₃) or calcium carbonate (CaCO₃) (Yuan et al., 2016). The whole process is summarized in Fig. 3 (Yuan et al., 2016).

In the first step (Eq. 1), carried out in the absorber unit, CO₂ is chemically absorbed using any metal aqueous hydroxide solution (LiOH/NaOH/KOH/Ca(OH)₂) to produce a carbonate, which is cyclically recycled back to the hydroxide-based solution. When NaOH is used as the aqueous hydroxide absorbent, the resultant Na₂CO₃ solution is mixed with Ca(OH)₂ to recover NaOH and CaCO₃ in a reaction known as causticizing (Eq. 2) (Zeman and Lackner, 2004). Calcium carbonate (CaCO₃) is then separated, dried and calcined at temperatures above 700 °C to form lime (CaO) (Eq. 3), releasing a concentrated CO₂ stream (Murdock et al., 2016). To complete the cycle, calcium hydroxide is regenerated by hydration in a slaker (Eq. 4) and reused in the causticizer. This process is typically carried out in packed columns, open

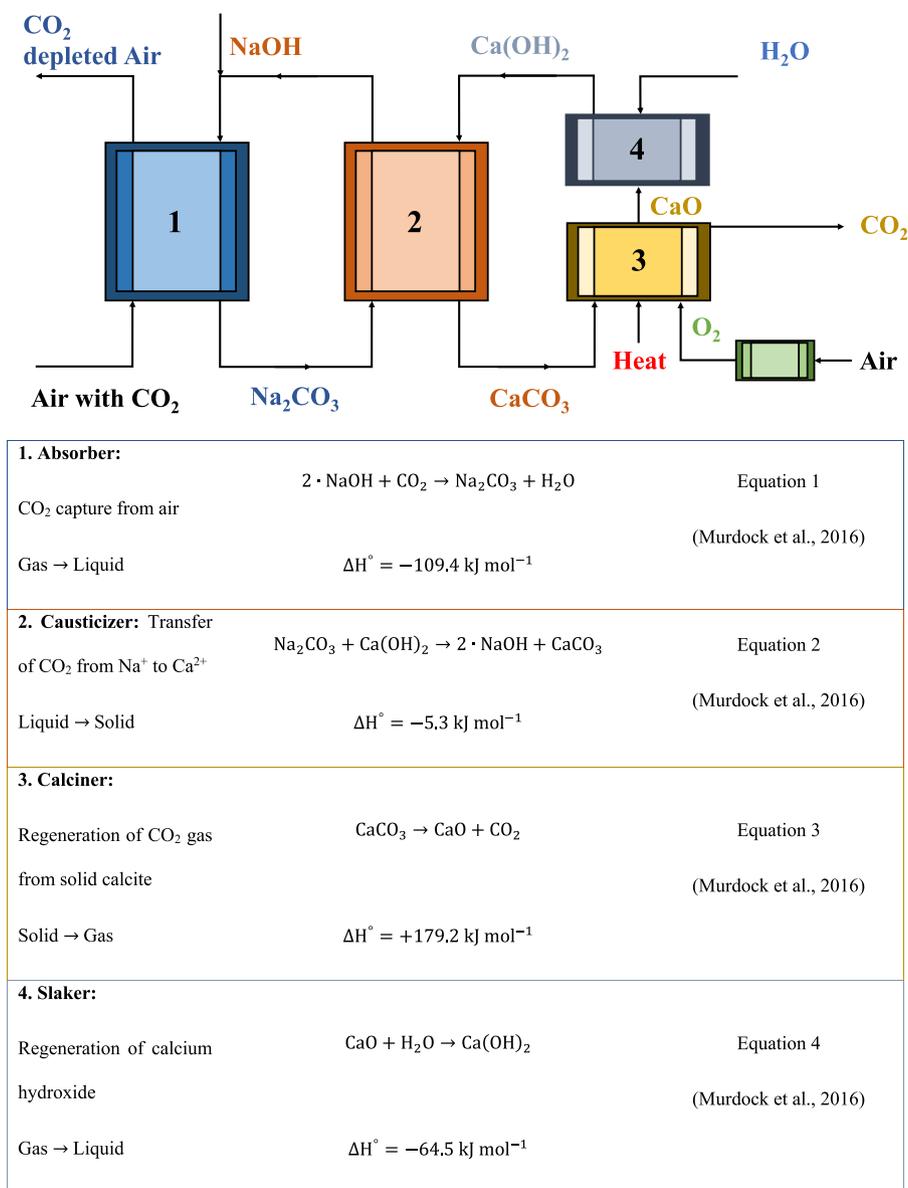


Fig. 3. Process diagram and equations of CO₂ extraction from ambient air using alkaline liquid sorbent. Adapted from Zeman and Lackner (2004) and Murdock et al. (2016).

towers, and pools, where CO₂ reacts with the aqueous hydroxide solutions (Mahmoudkhani et al., 2009; Qing et al., 2011) and numerous unit processes have been installed and implemented in the pulp and paper industry (Goepfert et al., 2012b; Spínola et al., 2021; Zeman and Lackner, 2004).

The regeneration step is energy intensive due to the high energy involved in the CO₂ binding with the aqueous hydroxide solution (Goepfert et al., 2012b; Murdock et al., 2016). Zeman (2007) assessed the overall energy requirements for CO₂ DAC using an aqueous hydroxide absorbent and obtained that nearly 58 % (256 kJ/mol) of the total energy required is used during the regeneration step. In contrast, a theoretical minimum of 109.4 kJ/mol is required for the conversion of calcium carbonate to calcium oxide (Murdock et al., 2016). In addition to energy requirements, water loss is of concern in CO₂-DAC systems because, due to the low CO₂ concentration in ambient air, a large interaction between the gas and liquid phases is required (Stolaroff and Keith, 2008). Operating costs can be reduced by improving contact tower design and by using more efficient packing materials, liquid, and gas distributors, and operation. Costs can also be reduced by using renewable energy sources to achieve the endothermic regeneration of the adsorbent, thus making the process more sustainable.

1. Absorber: CO ₂ capture from air	2 • NaOH + CO ₂ → Na ₂ CO ₃ + H ₂ O	Equation 1 (Murdock et al., 2016)
Gas → Liquid	ΔH° = - 109.4 kJ mol ⁻¹	
2. Causticizer: Transfer of CO ₂ from Na ⁺ to Ca ²⁺	Na ₂ CO ₃ + Ca(OH) ₂ → 2 • NaOH + CaCO ₃	Equation 2 (Murdock et al., 2016)
Liquid → Solid	ΔH° = - 5.3 kJ mol ⁻¹	

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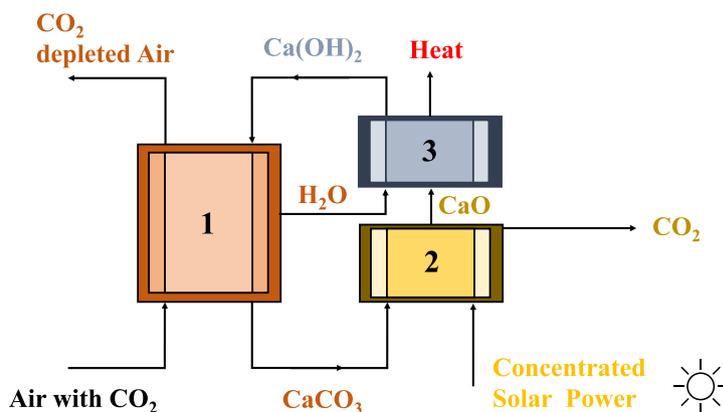
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3. Calciner: Regeneration of CO ₂ gas from solid calcite	CaCO ₃ → CaO + CO ₂	Equation 3 (Murdock et al., 2016)
Solid → Gas	ΔH° = + 179.2 kJ mol ⁻¹	
4. Slaker: Regeneration of calcium hydroxide	CaO + H ₂ O → Ca(OH) ₂	Equation 4 (Murdock et al., 2016)
Gas → Liquid	ΔH° = - 64.5 kJ mol ⁻¹	

3.1.2. Solid alkali sorbents

CO₂ adsorption using solid alkali is based on a carbonation reaction in which gaseous CO₂ reacts with solid metal oxide (MO) to yield a metal carbonate (MCO₃). The adsorbent can be then thermally regenerated by heating the metal carbonate beyond the calcination temperature (650–890 °C) (Gupta and Fan, 2002). However, the carbonation reaction using metal oxides is kinetically hindered. After an initial rapid and chemically controlled period, the reaction undergoes a much slower regime controlled by ion diffusion mechanisms (Dennis and Hayhurst, 1987). Moreover, Ca-based sorbents exhibited a decrease in their CO₂ uptake capacity with the number of cycles under both atmospheric and pressurized conditions, making this technology unsuitable for CO₂-DAC (Kuramoto et al., 2003).

The kinetic limitations can be overcome by using solid hydroxide adsorbents such as Ca(OH)₂ (Balen, 2005). CO₂ adsorption from air using a solid hydroxide adsorbent is thermodynamically feasible in the range of 298–1019 K, and is sustainable, particularly when using Ca(OH)₂ as an adsorbent and solar power for the regeneration step (Nikulshina et al., 2006) as shown in Fig. 4. Further studies examined the carbonation rates of CaO and Ca(OH)₂ by thermogravimetry at low



3. Carbonator:	Ca(OH) ₂ + CO ₂ → CaCO ₃ + H ₂ O	Equation 5
Carbonation reaction		(Nikulshina et al., 2006)
Gas → Liquid	ΔH° = -109.4 kJ mol ⁻¹	
2. Solar calciner:	CaCO ₃ → CaO + CO ₂	Equation 6
Calcination reaction		(Cannone et al., 2021;
Solid → Gas	ΔH° = +182.1 kJ mol ⁻¹	Shimizu et al., 1999)
3. Slaker:	CaO + H ₂ O → Ca(OH) ₂	Equation 7
Hydrolysis reaction		(Nikulshina et al., 2006)
Gas → Liquid	ΔH° = -310.37 kJ mol ⁻¹	

Fig. 4. Process diagram and equations of CO₂ capture using concentrated solar power. Adapted from (Nikulshina and Steinfeld, 2007).

CO₂ concentrations (500 ppm_v) aiming to simulate CO₂ capture from air at different moisture levels (Nikulshina and Steinfeld, 2007). Results showed that 44% of the CO₂ was removed during the first minute under dry conditions for temperatures between 300 and 450 °C, with a significant drop in CO₂ uptake in the following 20 min (Nikulshina and Steinfeld, 2007). Temperatures above 450 °C thermodynamically favoured the reverse CaCO₃-decomposition reaction to CaO and CO₂ and slowed the forward carbonation reaction, whereas only 2 % of the CO₂ was captured with temperatures below 325 °C.

Interestingly, a positive effect of water vapor (50 %) on carbonation was detected with an 80 % increase in the reaction rate after 100 min, while a 22 times faster rate was observed during the first 20 min. Such improvement was attributed to the CO₂ adsorption on the surface by hydroxyl (OH⁻) groups. Ca(OH)₂ carbonation was also studied at temperatures between 200 and 425 °C, and it proceeded at a faster rate than carbonation with CaO, maintaining a degree of conversion between 10 and 25 % for 100 min. NaOH was proposed as a solid adsorbent to reduce the costs associated with the high temperature required for the endothermic calcination process and the pre-heating of the air entering the carbonation reactor. However, this process was inefficient due to the slow reaction rates and high mass flow rates required (Nikulshina et al., 2008). Several authors agreed that using NaOH as a sorbent is not effective (Goepfert et al., 2012c; Jones, 2011; Murdock et al., 2016). Therefore, research on solid hydroxide sorbent has focused on designing new reactor configurations that reduce energy expenses by using solar power to provide the required energy for the process (Nikulshina et al., 2009; Nikulshina and Steinfeld, 2007). New alkali sorbent materials supported by porous matrices have also been proposed for CO₂-DAC, such as potassium-alumina sorbents, which are competitive materials to amine benchmark sorbents when tested at 1 % CO₂ concentration (Bali et al., 2015; Derevschikov et al., 2014; Murdock et al., 2016; Zhao et al., 2014). A summary of inorganic chemisorbent materials to perform iCO₂-DAC introduced in this section is presented in Table 1.

3. Carbonator: Carbonation reaction Gas → Liquid	Ca(OH) ₂ + CO ₂ → CaCO ₃ + H ₂ O ΔH° = - 109.4 kJ mol ⁻¹	Equation 5 (Nikulshina et al., 2006)
2. Solar calciner: Calcination reaction Solid → Gas	CaCO ₃ → CaO + CO ₂ ΔH° = + 182.1 kJ mol ⁻¹	Equation 6 (Cannone et al., 2021; Shimizu et al., 1999)
3. Slaker: Hydrolysis reaction Gas → Liquid	CaO + H ₂ O → Ca(OH) ₂ ΔH° = - 310.37 kJ mol ⁻¹	Equation 7 (Nikulshina et al., 2006)

3.2. Organoamine sorbents for CO₂-Direct air capture (CO₂-DAC)

One of the most used technological solutions to capture CO₂ from large flue gas streams is chemical absorption using amine-based solutions (Mangalapally and Hasse, 2011a). Although many solvents are available in the market, most power plants still tend to use alkanolamine solutions. The most common alkanolamines used for CO₂ absorption/desorption are primary amines such as MEA, 2-amino-2-methyl-1-propanol (AMP), secondary amines such as diethanolamine (DEA), and diisopropanolamine (DIPA), and tertiary amines such as methyl-diethanolamine (MDEA) (Al-Mamoori et al., 2017; Liu et al., 2016a).

MEA is the most used amine-based solution to capture CO₂ owing to its strong alkalinity and high absorption capacity per mass due to its low molecular weight (Liu et al., 2016a). MEA-based systems have been used to produce food-grade CO₂ from natural gas combustion gases for more than 60 years (Shen et al., 2017). The same process was later adapted to treat flue gases for CO₂ capture (Goepfert et al., 2010) and is also currently used in submarines to purify breathing air (Carey et al., 1983). MEA is an effective absorbent that can react with CO₂ rapidly and form carbamates, and can then be regenerated by heating, although the

Table 1

Summary of inorganic chemisorbent materials to perform iCO₂-DAC.

Category	Characteristics	Example	Reference
Solution-based	Examples	Skim CO ₂ from ambient air	Mikulčić et al. (2019)
	Aqueous solution of sodium hydroxide (NaOH) or calcium hydroxide (Ca(OH) ₂)	Equation 2- Process description Equation 3- Process description	Zeman and Lackner (2004) Murdock et al. (2016)
	Regeneration method	Reactor type for CO ₂ capture	(Mahmoudkhani et al., 2009; Qing et al., 2011)
	High temperatures (over 800 °C) and an oxygen atmosphere	Example of application	(Goepfert et al., 2012b; Spínola et al., 2021; Zeman and Lackner, 2004).
Solid-based	Examples	Carbonation reaction characterization	(Murdock et al., 2016; Stolaroff and Keith, 2008; Zeman, 2007)
	Solid metal oxide (MO) such as CaO-based materials.	Solar-powered regeneration of adsorbent (Ca(OH) ₂)	(Balen, 2005; Dennis and Hayhurst, 1987)
	Regeneration method	CO ₂ capture from the air at different moisture levels	(Nikulshina and Steinfeld (2007)
	Use of NaOH as adsorbent	Use of NaOH as adsorbent	(Goepfert et al., 2012c; Jones, 2011; Murdock et al., 2016)
Solid-based	Heating the metal carbonate beyond the calcination temperature (650–890 °C)	New reactor designs	(Nikulshina et al., 2009; Nikulshina and Steinfeld, 2007)
		New Alkali sorbent supported materials	(Bali et al., 2015; Derevschikov et al., 2014; Murdock et al., 2016; Zhao et al., 2014)

precise reaction mechanism is still controversial (García-Abuín et al., 2013; Lv et al., 2015; Ma et al., 2014). García-Abuín et al. (2013) produced a mixture of carbamate and bicarbonate as the main products during CO₂ absorption using MEA, DEA, and triethanolamine (TEA) aqueous solutions as shown in Fig. 5. In the study from Lv and co-workers (Lv et al., 2015), the end product of the reaction of MEA and CO₂ was dependent on CO₂ loading and on the presence of water, as summarized in Fig. 6. Besides the CO₂ absorption reaction described in Eq. 8, some MEA can also react with bicarbonate or carbonate as described in Eq. 9 and 10, respectively. At high CO₂ loadings, carbamate hydrolysis occurs, where carbamate reacts with protons to form bicarbonate and protonated MEA as described by Eq. 11, while MEA regeneration through the application of heat is described by Eq. 12.

Despite being a mature technology with about 100% efficiency for CO₂ capture, there are major obstacles to using MEA aqueous systems

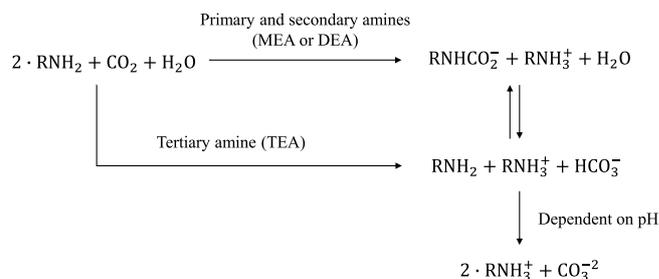
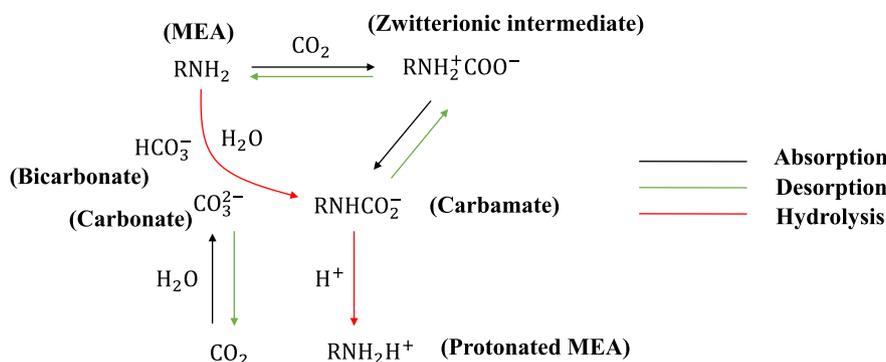


Fig. 5. Reaction mechanism for CO₂ chemical absorption using primary, secondary, or tertiary amines. Adapted from (García-Abuín et al., 2013).



1. MEA-CO₂ absorption	$\text{RNH}_2 + \text{CO}_2 \rightarrow \text{RNHCO}_2^-$	Equation 8 (Lv et al., 2015)
2. MEA-HCO₃⁻ absorption	$\text{RNH}_2 + \text{HCO}_3^- \rightarrow \text{RNHCO}_2^- + \text{H}_2\text{O}$	Equation 9 (Lv et al., 2015)
3. MEA-CO₃²⁻ absorption	$\text{RNH}_2 + \text{H}^+ + \text{CO}_3^{2-} \rightarrow \text{RNHCO}_2^- + \text{H}_2\text{O}$	Equation 10 (Lv et al., 2015)
4. Carbamate hydrolysis	$\text{RNHCO}_2^- + \text{H}^+ + \text{H}_2\text{O} \rightarrow \text{RNH}_2\text{H}^+$	Equation 11 (Lv et al., 2015)
5. Carbamate regeneration	$\text{RNHCO}_2^- + \text{H}^+ \xrightarrow{\text{heating}} \text{RNH}_2 + \text{CO}_2$	Equation 12 (Lv et al., 2015)

Fig. 6. Reaction mechanism for CO₂ capture into MEA. Adapted from (Lv et al., 2015).

for iCO₂-DAC. The major obstacle of MEA-based systems in large scale applications is the high capital and operating costs, mainly associated to endothermic solvent regeneration, due to the high absorption heat capacity of MEA and the high energy consumption related to its regeneration (Liu et al., 2016b; Sakwattanapong et al., 2005). It is estimated that around 65% of the energy required for CO₂ capture in amine scrubbing is associated with heating of the aqueous amine solution to the desorption temperature (Veneman et al., 2012). Another limitation of MEA-based systems is the technical scalability which limits its application to CO₂ capture from common electricity power plants burning coal or natural gas (Shen et al., 2017). Other problems are the corrosive nature of the amines and the fouling of the process equipment (Mandal and Bandyopadhyay, 2006; Q. Wang et al., 2011), in addition to the environmental unsustainability of MEA (Chitsiga et al., 2016; Mangalapally and Hasse, 2011b). The main challenges are summarized

Table 2
Challenges of MEA aqueous systems for iCO₂-DAC implementation.

Challenge	Reference
High capital and operating costs mainly associated with endothermic solvent regeneration due to the high absorption heat of MEA.	Sakwattanapong et al. (2005) Liu et al., 2016
Technical scalability limits its application to CO ₂ capture from common electricity power plants burning coal or natural gas.	Shen et al. (2017)
Corrosive nature of the amines and fouling of the process equipment.	Q. Wang et al. (2011)
Environmental unsustainability of MEA.	Chitsiga et al., 201

in Table 2.

Chemical adsorption using supported amines is one of the most promising pathways to achieve highly selective CO₂-DAC (Goepfert et al., 2010). Replacing water by a solid support greatly reduces the energy required for regeneration, due to the lower heat capacity of solid support materials compared to water. In addition, the evaporation of water is prevented, further reducing the process energy requirement (Veneman et al., 2013). Supported amine adsorbents consist of a high surface-area support with amine functional groups immobilized on, or grafted to, its surface, where a chemical reaction takes place between CO₂ and the amine groups, creating strong bonds and allowing significant uptakes even at low CO₂ partial pressures (Sanz-Pérez et al., 2016b; Veneman et al., 2012). Besides achieving high adsorption rates, the chemically bonded adsorbent will have a higher stability during their adsorption-desorption cycles (Sayari and Belmabkhout, 2010). While physical adsorbents are generally characterized by their textural properties such as surface area, pore diameter and pore volume (Sanz-Pérez et al., 2015; Sevilla et al., 2013), when using mesostructured materials commonly employed to support amine species, the most influential parameters affecting the CO₂ DAC uptake rate are surface area and gas-surface interaction (Sanz-Pérez et al., 2016b). Most of the solid adsorbents used for CO₂ capture from flue gases are physical adsorbents which are not entirely suitable to perform CO₂ DAC, mainly due to the low CO₂ partial pressure. However, chemical adsorption plays a critical role in CO₂ DAC, especially when the CO₂ concentration is below 1% (vol/vol) making the amine loading over the surface area available the most influential parameter. The main advantages of solid support-based

systems for CO₂ capture are summarized in Table 3.

In general CO₂ molecules chemically adsorb on the support material with readily available amine groups on the surface or in the interior of the material structure. Therefore, the higher the concentration of readily available amino groups in the adsorbent material, the higher the CO₂ adsorption capacity (Quang et al., 2015). However, there is a threshold amine loading at which the maximum number of readily available amines is obtained, with maximum CO₂ adsorption capacity. CO₂ adsorption on amino-based mesostructured supported adsorbents is principally limited by diffusion resistance, which increases with higher amino loading and amine molecular weight but decreases with increasing temperature (Sayari et al., 2016). To overcome this limitation supports with larger surface areas, larger pore volumes, and increased pore connectivity can be used (Choi et al., 2009).

Chemical adsorption of CO₂ has been studied in different ways, 1) using solid amines or polyamines directly as adsorbents, 2) depositing amines or polyamines on a solid support, and 3) chemically bonding amines or polyamines to the surface of a solid (Goepfert et al., 2012b). Solid-supported amines can be classified based on the interaction between the support and active sorbent and the approach used in their preparation as summarized in Fig. 7 (Alessandro et al., 2010; Karademir and Ozmen, 2017; Li et al., 2010; Murdock et al., 2016; Pu and Su, 2018; Sculley and Zhou, 2012; Shen et al., 2017; Veneman et al., 2013). Class 1-supported amines are based on porous supports impregnated with monomeric or polymeric amines, while class 2-supported amines are based on amines covalently linked to the walls of porous materials mainly obtained by binding amines to oxides via silane chemistry or the preparation of polymeric supports with amine-containing side chains. Class 3 supported amines consist of an inorganic support and a chemically grafted polyamine component that is prepared by in-situ polymerization of amine-containing monomers (Goepfert et al., 2012b; Murdock et al., 2016). In this work, special focus is placed on class 1 supported amines describing the main characteristics, the operating principle, main advantages, and limitations, together with representative examples of application. Further information about class 2 and class 3 supported amines adsorbents are available in the literature (Alessandro et al., 2010; Chaikittisilp et al., 2011; Goepfert et al., 2012c; Karademir and Ozmen, 2017; Li et al., 2010; Murdock et al., 2016; Quang et al., 2013; Sculley and Zhou, 2012; Shen et al., 2017; Shi et al., 2020; Veneman et al., 2013).

Class 1-supported amines exhibit weak interactions such as hydrogen bonds, electrostatic interactions, or van der Waal interactions (Goepfert et al., 2012b). They were originally developed by Song and collaborators (Xu et al., 2003) and their main advantage is that large amounts of amines can be added into the pores of supports such as silica (silica gel, precipitated silica, fumed silica) (Chen et al., 2009; Goepfert et al., 2011). In addition to different silica materials, researchers have loaded amines onto mesoporous alumina (Chen and Ahn, 2011), bentonite (Roth et al., 2013), clay (Roth et al., 2013), fly ash (Dindi et al., 2017), activated carbon (Plaza et al., 2007), mesoporous carbon (J. Wang et al., 2013), glass fiber (Li et al., 2008), MOFs (Karademir and Ozmen, 2017), nanofibrillated cellulose (Sehaqui et al., 2015), resin (Chen et al., 2013), propylene fiber (Xu et al., 2015), among others. Polyethyleneimines

(PEIs) are the most common amines, including low and high-molecular-weight varieties and linear and branched forms of the polymer (Choi et al., 2009). A summary of the CO₂ capture performance of various materials functionalized with PEI can be found elsewhere (Shen et al., 2017). PEI has proven to be very efficient, with large adsorption capacities and high amine efficiencies, defined as the moles of CO₂ captured per mole of amine (Chaikittisilp et al., 2011).

Impregnation of polymeric amines such as PEI involves their physical deposition onto the surface of the porous support, which includes the preparation of the amine solution, followed by solvent removal once the wet impregnation is done (Liu et al., 2019; Meng et al., 2018). This technique has been used more frequently than amine grafting, due to its simplicity and the absence of chemical reactions, resulting in lower costs and enabling the use of larger quantities of amines to obtain high adsorption capacities (Azmi and Aziz, 2019; Goepfert et al., 2010). Different amines have been used for preparing supported amines adsorbents, such as MEA, DEA, diisopropanolamine (DIPA), triethanolamine (TEA) (Franchi et al., 2005; Goepfert et al., 2010), polyallylamine (PAA) (Alkhabbaz et al., 2014), and tetraethylenepentamine (TEPA) (Feng et al., 2013). Due to the weak physical interactions that hold amines on the surface, class 1 supported amines gradually lose their adsorption performance due to amine leaching (Goepfert et al., 2012b). Nonetheless, the large number of weaker physisorption interactions may still impart significant stability to these materials (Hicks et al., 2008). Goepfert et al. evaluated the CO₂ adsorption and desorption capacity of various types of amines, including simple amines (MEA, DEA, and TEA) and polymeric amines, such as linear and branched PEI including low molecular weight (LMW, M_w ca. 800) and high molecular weight (HMW, M_w ca. 25000), tetraethylenepentamine (TEP) and pentaethylenhexamine (PEH) under different operating conditions (Goepfert et al., 2010). The results are summarized in Table 4. Branched PEI-HMW showed lower absorption capacity than branched PEI-LMW. However, PEI-HMW did not show any leaching during the test, making it a better choice for CO₂ capture. Notably, PEI-supported materials displayed remarkable behavior when the temperature increased, which is the opposite of what occurs when liquid-phase amines are used. In a subsequent study, Goepfert and collaborators determined that PEI impregnated on fumed silica (FS) and precipitated silica showed the most promising results for CO₂ capture, so they proceeded to apply such materials for CO₂-DAC (Goepfert et al., 2011). The reaction mechanism of CO₂ with PEI is shown in Fig. 8 based on Goepfert et al. (2014). Like its liquid amine counterparts (MEA, DEA, and MDEA), under dry conditions, the main reaction is the formation of carbamate, giving a maximum loading of 0.5 mol CO₂/mole amine. However, under humid conditions, the conversion of carbamate to bicarbonate takes place, therefore, one mol of amine can remove 1 mol of CO₂.

Theoretically, the CO₂ adsorption capacity of PEI is always higher under humid conditions. However, this was not the case for the material FS-PEI-50 and only occurred for FS-PEI-33, corresponding to 50 % (w/w) of FS-50 % (w/w) of PEI and 67 % (w/w) FS-33 % (w/w) of PEI, respectively. This difference in the effect of dry and humid air over the two materials with different amine loading was explained by the authors by a gas diffusion limitation into the adsorbents in the highly loaded material (FS-PEI-50). Several authors have also reported a detrimental effect of water on the adsorbent material (Didas et al., 2014; Goepfert et al., 2011). Another possible explanation is the absence of an appropriate environment on the sorbent for the formation of carbonate and bicarbonate species (Murdock et al., 2016). Nonetheless, the results demonstrated that PEI-impregnated materials can be used to capture CO₂ from diluted sources, especially from the atmosphere, having the capacity to operate under both dry and humid conditions. Other studies on amines or support material reported in the literature are summarized in Table 5. A summary of organoamine adsorbent materials to perform

Table 3
Advantage of solid support-based systems ahead of iCO₂-DAC implementation.

Advantage	Reference
Reduced energy requirement for regeneration, due to the lower heat capacity of solid supports compared to water.	Goepfert et al. (2010)
Water evaporation is prevented, reducing the process energy requirement.	Veneman et al. (2013)
High surface-area support with strong bonds allows significant uptakes even at low CO ₂ partial pressures.	Sanz-Pérez et al., 2016 Veneman et al. (2012)
Higher adsorption rates, with higher stability during their adsorption-desorption cycles.	Sayari and Belmabkhout (2010)

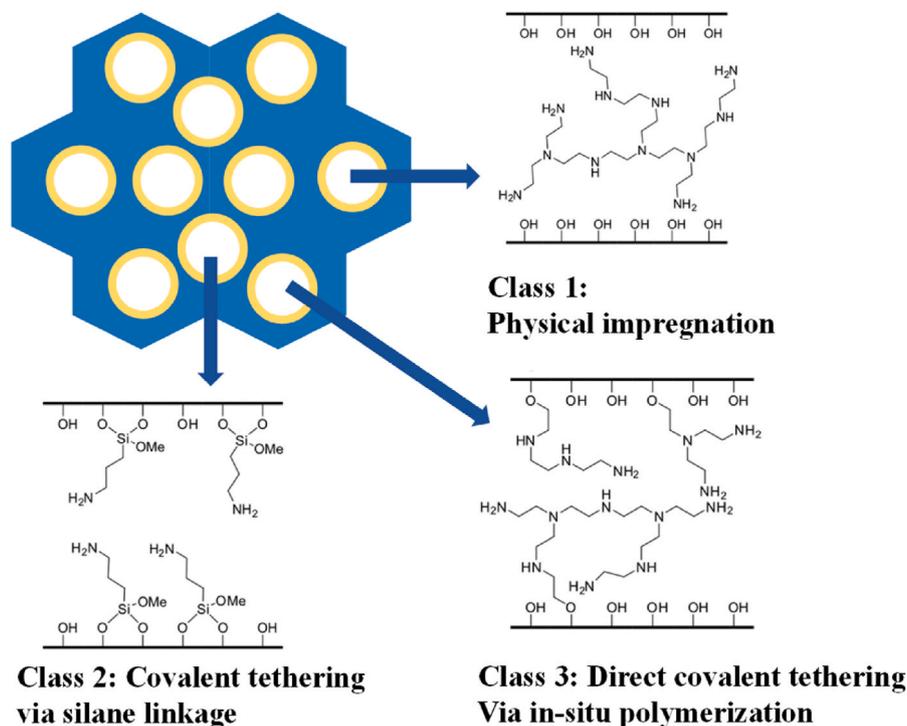


Fig. 7. Three classes of amine-modified adsorbents. Class 1 adsorbent:PEI) impregnated in porous materials. Class 2: amines covalently tethered through silane linkages. Class 3: in situ aziridine polymerization on a solid material. Adapted from (Li et al., 2010; Murdock et al., 2016; Shi et al., 2020).

Table 4

Absorption of CO₂ on various amine/precipitated silica (1:1) adsorbents at 70 °C. Adapted from (Goepfert et al., 2010).

Amine	Adsorption capacity (mg CO ₂ per g adsorbent)	Observation
Simple amines (MEA, DEA, and TEA)	Between 3 and 113	Not suitable due to leaching problems and/or poor CO ₂ absorption
TEP	200	Some amine leaching and slow desorption compared to PEI (linear)
PEH	192	Some amine leaching and slow desorption compared to PEI (linear)
PEI linear	173	Suffered important leaching problems at the highest temperatures tested (100 °C)
Branched PEI (HMW)	130	No Amine leaching
Branched PEI (LMW)	147	Minimal amine leaching

iCO₂-DAC introduced in this section is presented in Table 6.

1. MEA-CO ₂ absorption	$\text{RNH}_2 + \text{CO}_2 \rightarrow \text{RNHCO}_2^-$	Equation 8 (Lv et al., 2015)
2. MEA-HCO ₃ ⁻ absorption	$\text{RNH}_2 + \text{HCO}_3^- \rightarrow \text{RNHCO}_2^- + \text{H}_2\text{O}$	Equation 9 (Lv et al., 2015)
3. MEA-CO ₃ ²⁻ absorption	$\text{RNH}_2 + \text{H}^+ + \text{CO}_3^{2-} \rightarrow \text{RNHCO}_2^- + \text{H}_2\text{O}$	Equation 10 (Lv et al., 2015)
4. Carbamate hydrolysis	$\text{RNHCO}_2^- + \text{H}^+ + \text{H}_2\text{O} \rightarrow \text{RNH}_2\text{H}^+$	Equation 11 (Lv et al., 2015)
5. Carbamate regeneration	$\text{RNHCO}_2^- + \text{H}^+ \xrightarrow{\text{heating}} \text{RNH}_2 + \text{CO}_2$	Equation 12 (Lv et al., 2015)

3.3. Strategies for iCO₂-DAC adsorption material regeneration

Adsorption material regeneration is the most critical step for iCO₂-DAC. This involves the desorption method applied, desorption rate, and material stability. The regeneration of amine-based support materials can be performed through different methods such as temperature swing adsorption (TSA), pressure swing adsorption (PSA), or vacuum swing adsorption (VSA). In TSA, the adsorbent material is heated by a hot gas

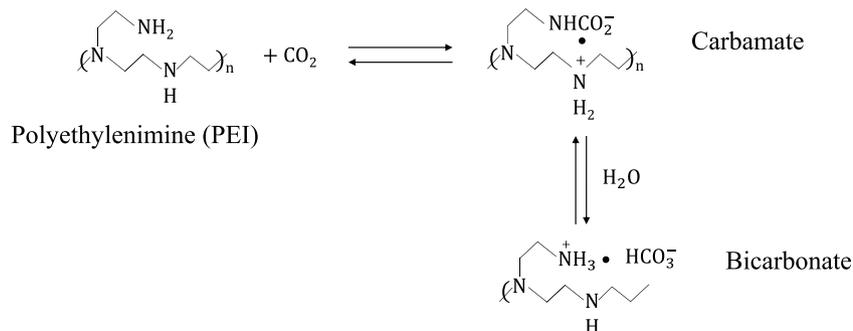


Fig. 8. Simplified mechanistic reaction model of CO₂ with PEI. Adapted from (Goepfert et al., 2014).

Table 5Adsorption of CO₂ on various support materials.

Amine	Adsorption capacity (Mmol CO ₂ per g adsorbent)	Reference
Hyperbranched aminosilica with an organic content of 42.5%	1.44	Choi et al. (2011a), 2011b
Triamine-grafted pore-expanded mesoporous silica MCM-41	0.98	Belmabkhout et al. (2010)
PEI-LMW impregnated onto an SBA-15 mesoporous silica support containing isolated Zirconium (Zr). ZrX-SBA-15, where X is the Zr content from 0 to 14 mol %	0.26–0.85	Kuwahara et al. (2012)
PEI-SBA-15 (0 % Zr)	0.19	Kuwahara et al. (2012)
Commercial porous silicate (CARiACT G10 HPV) impregnated with 45 % PEI-LMW	2.36	Choi et al. (2011b)
Silica monocellular foam (MSCA) material impregnated with a 46 % branched PEI-LMW	4.74	Chaikittisilp et al. (2011)

for desorption, while in PSA the CO₂ is adsorbed at high pressures and then desorbed, concentrated, and collected at lower pressures (Elfving et al., 2017; Reynolds et al., 2005). In VSA, the adsorption step is performed at atmospheric pressure and desorption is obtained under low pressure (Lijuan et al., 2021; Tlili and Gre, 2009). Using nitrogen as the sweep gas during CO₂ desorption may be impractical if a high-purity CO₂ is required for downstream applications (Zhang et al., 2016). Using pure CO₂ as sweep gas for CO₂ desorption can yield high-purity CO₂, but may result in thermal degradation and lower working capacities (Hoffman et al., 2014; Veneman et al., 2012).

Researchers have studied the effect of CO₂ on several amine-containing adsorbents under different TSA conditions and observed that the deactivation of the adsorbent material was due to the formation of stable urea groups at the expense of amine. However, none of the materials deactivated when humidified gases were used instead of dry gases (Sayari and Belmabkhout, 2012). Alternatively, low-quality or pure steam has been proposed to purge CO₂ in TSA desorption to alleviate the thermal degradation of amines. However, this approach faces many challenges associated with the lack of stability of the material's pore structure, the higher thermal energy penalty, and the need for additional devices to condense water from the desorbed CO₂ stream (Gray et al., 2009; Li et al., 2010). The combination of PSA and VSA processes, PVSA, was investigated at 5.5 bar pressure and 50 mbar vacuum (Chou and Chen, 2004; Zhao et al., 2017). Other works in the literature report the combinations of temperature vacuum swing adsorption (TVSA) (Jiang et al., 2020; Su and Lu, 2012) and pressure swing adsorption (PTSA) (Mulgundmath and Tezel, 2010; Song et al., 2016) to reduce the operational cost of the process. Lackner and collaborators proposed a novel process, moisture swing adsorption, which offered a low-energy method for adsorbent regeneration during CO₂-DAC (T. Wang et al., 2011, 2013).

Moisture swing adsorption trades the input of heat in the thermal swing or mechanical energy in the pressure-based swing against the consumption of water, whose evaporation provides the free energy that drives the cycle. Therefore, moisture swing adsorption can simplify the adsorbent regeneration process, especially in places where water is not a limiting factor. However, this process might not be suitable for CO₂-DAC adsorbents that are not tolerant to high moisture content or whose CO₂ adsorption capacity decreases with high moisture content. The right conditions for the regeneration step must be selected to avoid problems such as amine thermal degradation through the formation of open chain and/or cyclic urea (Sayari et al., 2012). TSA has been the most commonly applied methodology to regenerate amine-based supported materials. Due to the relatively low volatility of amine species (Murdock et al., 2016), the adsorbent material regeneration step can be performed

Table 6Summary of organoamine adsorbent materials to perform iCO₂-DAC.

Category	Characteristics	Example	Reference
Amine-based solutions	Examples Alkanolamine solutions made of primary amine (MEA, AMP), secondary (DEA), (DIPA) and tertiary amines (MDEA). Regeneration method Temperature (over 800 °C) and vacuum swing	Type of amines MEA characterization MEA, DEA and TEA reaction mechanisms MEA regeneration costs MEA-based systems challenges	(Al-Mamoori et al., 2017; Liu et al., 2016a; Mangalappally and Hasse, 2011a) (Liu et al., 2016a; Shen et al., 2017) (García-Abuín et al., 2013; Lv et al., 2015; Ma et al., 2014). (Liu et al., 2016b; Sakwattanapong et al., 2005). (Mandal and Bandyopadhyay, 2006; Veneman et al., 2012; Q. Wang et al., 2011)
Supported amines	Example Low molecular weight and high molecular weight polyethyleneimine (PEI) impregnated in porous materials (silica, carbon fibers, polymers). Regeneration method Temperature (below 110 °C) and/or vacuum swing	Supported amines characterization CO ₂ adsorption on amino-based mesostructured supported adsorbents characterization Limitations of amino-based supported adsorbents Adsorption mechanisms Supported amines adsorbents classification	(Bos et al., 2019; Elfving et al., 2021; Sanz-Pérez et al., 2016a; Schellevis et al., 2021) Sayari et al. (2016) Choi et al. (2009) Goepfert et al. (2012b) (Alessandro et al., 2010; Karademir and Ozmen, 2017; Li et al., 2010; Murdock et al., 2016; Pu and Su, 2018; Sculley and Zhou, 2012; Shen et al., 2017; Veneman et al., 2013)

under relatively mild conditions with low energy consumption (Azmi and Aziz, 2019). The thermal energy required for the regeneration of CO₂ adsorbents is regarded as the most important criterion to evaluate the applicability of different materials and processes (Zhang et al., 2016).

The relationship between adsorption and desorption characteristics of silica-organic hybrid adsorbents as a function of parameters such as adsorption and desorption temperature, flow rate, and PEI characteristics (loading and MW) has been investigated (Goepfert et al., 2014). The effect of temperature on the CO₂ adsorption capacity of FS-PEI-33 and FS-PEI-50 materials showed a decreasing capacity from 50 to 70 mg CO₂ per gram, respectively, to almost zero when the temperature was increased from room temperature (25 °C) to 85 °C. Since the temperature difference typically required for adsorption and desorption is near 50–60 °C, this type of material could be used under low-temperature conditions, significantly decreasing the amount of energy required for solvent regeneration at mild temperatures (80–85 °C). However, other authors found a counterintuitive trend, where higher CO₂ uptakes occurred at elevated temperatures, which was explained by the

competition of thermodynamic factors (increased adsorption at low temperatures) and kinetic factors (better diffusion of CO₂ through polymeric PEI layers at high temperatures and higher reaction rates) (Xu et al., 2002, 2003). Regarding the effect of sweeping gas flow rate and desorption temperature, higher nitrogen flow rates decrease the time needed for CO₂ desorption while high desorption temperatures (100 °C) provide faster CO₂ desorption rates than lower desorption temperatures (70 °C). Goepfert et al. proved PEI-based material (FS-PEI-50) was resilient after performing 4 and 5 cycles of adsorption/desorption cycles to test the resilience of the materials (Goepfert et al., 2010, 2011).

The regenerability of a solid molecular basket adsorbent prepared from PEI (50%) applied on SBA-15 to capture CO₂ from the air was tested in 20 cycles of absorption/desorption. Up to 750 adsorption/desorption cycles were performed to study the effect of water vapor on amine-containing CO₂ adsorbents (Sayari and Belmabkhout, 2010). However, studies with more adsorption/desorption cycles are needed to evaluate the long-term stability and repeatability of the results over adsorption/desorption cycles. Overall, class 1 amine-based adsorbents, specifically PEI impregnated on based fumed/precipitated silica have shown favourable characteristics for iCO₂-DAC, due to relatively low cost and straightforward preparation of the adsorbent material. Still, the energy cost for regeneration needs to be reduced. This can be achieved by modifying the bonding between CO₂ and the capture medium to be weaker or using a less-intensive regeneration methodology to release CO₂ other than heat or vacuum (Beaumont, 2022). For example, humidity-swing (Shi et al., 2019; Van Der Giesen et al., 2017; T. Wang et al., 2011) or electro-swing CO₂ adsorption-desorption methodology (Voskian and Hatton, 2019). It is critical to select a sorbent that resists thermal, chemical, and mechanical stress, as sorbents are constantly exposed high temperatures, chemically reactive molecules such as ozone and continuous changes in pressure, effects that can significantly reduce the lifetime of the sorbents. Modifying the surface of solid amine adsorbents by mean of the addition of surfactants has been suggested as a strategy to reduce the energy involved in amine regeneration (Chen et al., 2023).

3.4. Operating conditions for an efficient CO₂ adsorption-desorption process: mass transfer efficiency optimization through gas contactors designs

The goal of CO₂-DAC is to remove CO₂ efficiently, but not necessarily as much CO₂ as possible as is the case for CCS from flue gas. Since flue gas scrubbing is the *de facto* standard in CO₂ capture, it is helpful to compare CO₂-DAC with flue gas scrubbing (Lackner, 2013). In the case of CO₂-laden flue gas streams, different reactor configurations have been proposed for contacting gas phase within solid adsorbents including fixed-bed (Lara and Romeo, 2017; Li et al., 2013), fluidized beds (K. Kim et al., 2020; Veneman et al., 2012; Zhang et al., 2014b) and moving bed (Jung et al., 2017). For amine-based solid adsorbents, a fluidized bed is one of the most commonly applied technologies for CO₂ capture (Jung et al., 2017; K. Kim et al., 2020; Veneman et al., 2012). Veneman and co-workers (Veneman et al., 2012) utilized a circulating fluidized bed (CFB) reactor with a supported amine adsorbent, specifically silica and polymethylmethacrylate (PMMA) support impregnated with tetraethylenepentamine (TEPA) solution, for post-combustion CO₂ capture. Experimental studies on CO₂-DAC are principally focused on feasibility analysis and characterization of adsorbent materials. Therefore they are mostly limited to small-scale adsorbent testing using thermal gravimetric analysis (TGA) (Goepfert et al., 2014; Zhang et al., 2018) or small-volume U-shaped quartz reactors (Chen et al., 2013; Goepfert et al., 2014). This approach has been mostly applied at an early stage of development since, in addition to minimizing the material and energy inputs needed for each experiment, it provides a controlled environment eliminating possible perturbations during the adsorption and desorption processes.

The configurations used are not entirely limited to TGA or U-shaped

quartz tubes, but contactors tested also include monolith (Kulkarni and Sholl, 2012), fluidized beds (Zhang et al., 2014a) and fixed bed contactors (Bajamundi et al., 2019; Elfving et al., 2017; Schellevis et al., 2021; Yu and Brilman, 2017), being the latter the most applied, especially in research studies once the amine-based support material has been already developed. Hence, fixed bed adsorption is applied when research is focused on the optimization of operational conditions (Schellevis et al., 2021), adsorbent material characterization (Wang et al., 2012), experimental testing (Wurzbacher et al., 2011, 2012, 2016, 2012, 2016), and modelling (TSA and TVSA) of regeneration methods (Ben-Mansour et al., 2016; Elfving et al., 2017), reactor design and control strategy (Yu and Brilman, 2017), production of high purity CO₂ streams (Bos et al., 2019) and process control improvement (Bajamundi et al., 2019). Samanta et al. (2012) summarized the advantages of fluidized-bed contactors in comparison to fixed bed as 1) excellent gas-solid contact due to vigorous agitation of adsorbent particles, 2) minimum diffusional resistance, 3) uniformity of temperature, 4) faster overall kinetics. However, fixed-bed contactors are generally preferred over fluidized beds because they minimize the height of the bed, resulting in a low-pressure drop and allowing a more compact design, which reduces capital expenditures (CapEx). Fluidized beds require a certain bed height to maintain a uniform gas distribution since non-uniform gas distribution can lead to local bypassing of the adsorbent bed, reducing gas removal efficiency (Yu and Brilman, 2017). In addition, fluidized beds require a gas distributor for an even gas profile along the bed, and a freeboard zone for gas-solid separation, increasing the pressure drop. Zhang et al. (2014a) indirectly demonstrated this by evaluating the CO₂-DAC adsorption capacity of a circulating fluidized bed (CFB) adsorber and a bubbling fluidized bed (BFB) desorber using PEI-silica adsorbent as the circulating bed materials. While the CFB-BFB system captured nearly 100% of CO₂ from ambient air, it had high electricity consumption due to the high-pressure drop.

Recent works have suggested the use of radial flow reactors (RFR) to reduce the energy penalty related to the pressure drop during CO₂ adsorption in fixed-bed contactors (Yu and Brilman, 2017, 2020). Counter-current moving bed contactors have also been proposed to overcome the rapid decrease in the solid adsorbent activity during CO₂-DAC, but the fixed-bed configuration still offered 15–25 % higher CO₂ adsorption capacities compared to the counter-current moving bed configuration (Schellevis et al., 2020). Therefore, for large-scale CO₂-DAC applications, shallow RFR is envisioned as a suitable technological solution.

4. Advancing the future: Integrating microbial electrosynthesis with iCO₂-DAC

In addition to enhancing air quality through capturing CO₂ from closed environments, there are further environmental and economic benefits that can be achieved by converting it into valuable green chemicals and fuels for on-site use or introduction to the market. This section examines the progress of a pioneering, environmentally friendly modular prototype to produce carbon-neutral commodity chemicals, while effectively removing CO₂ from indoor environments, thereby improving IAQ. Fig. 9 presents a process diagram illustrating the envisioned technology. An airstream from an indoor environment flows into an iCO₂-DAC prototype named CO₂ micro-concentrator module (CO₂-MCM) to adsorb the CO₂ while releasing a stream of free of CO₂ or with significantly lowered indoor CO₂ will be recirculated back into the indoor environment. The key feature of the envisioned CO₂-MCM module is the original design and fabrication methodology based on additive manufacturing (3D printing), maximizing the contact between the gas and solid adsorbent, and as well in the miniaturization of the device, which helps to optimize the energy utilization during the desorption step and as well increases the tightness of the device, avoiding heat leaks into the surroundings. Once the CO₂-MCM module is saturated, CO₂ is then desorbed and delivered into the second module, named microbial

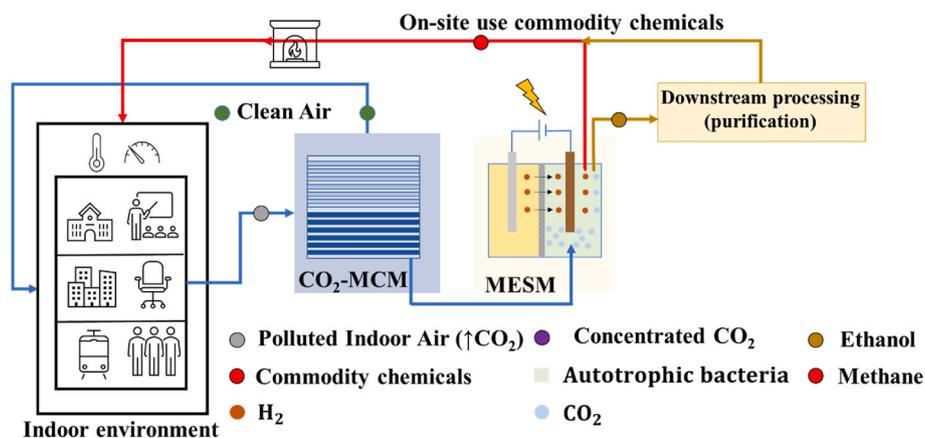


Fig. 9. The envisioned solution to produce valuable commodity chemicals based on CO₂ capture and conversion from indoor environments is based on the MICRO-BIO process operating principle diagram (López et al., 2022). Legend: CO₂-MCM: CO₂ Microconcentrator Module, MESM: Microbial Electrosynthesis Module. The goal is to capture CO₂ from indoor environments and convert it into sustainable fuels in microbial electrosynthesis reactors.

electrosynthesis module (MESM), where electrochemically produced hydrogen (H₂) is mixed with the CO₂ desorbed from the CO₂-MCM and utilized by specialized microorganisms to produce short-chain chemical commodities, such as methane and ethanol. Although process automation and integration of the different modules is not discussed in this manuscript, both are key elements to achieve a mature, versatile, and robust operation of the envisioned technology, as is envisioned as a fully automated device to be integrated within indoor facilities.

This section provides a short overview of the fundamentals of microbial electrosynthesis, reactor design, and operating conditions. Additionally, a case study assessing the use of microbial electrosynthesis to recycle the captured CO₂ from three indoor environments (schools, offices, and underground metro carriage) is included.

4.1. Fundamentals of microbial electrosynthesis

Among the available CO₂ conversion technologies (bio)electrochemical cells can be easily connected to the electric grid, making them the most suitable option for recycling CO₂ inside buildings (Grim et al., 2020). Microbial electrosynthesis (MES) in bioelectrochemical cells is one of the potential carbon capture and utilization technologies under development and is capable of producing renewable fuels such as methane and ethanol (Dessi et al., 2021; Nevin et al., 2010).

MES reactors rely on chemolithoautotrophic microorganisms that reduce CO₂ by harvesting electrons from a cathode electrode through three pathways (Logan et al., 2019; Tremblay et al., 2017): (i) direct contact *via* membrane-bound, electron-transfer proteins such as c-type cytochromes; (ii) self-generated or added soluble electron shuttles (e.g. proteins such as flavins); (iii) through electrochemically or bioelectrochemically produced mediators such as H₂ and formate. When the cathode electrode is poised at a potential low enough to enable abiotic H₂ evolution (−0.414 V vs SHE in theory under standard conditions, but often below −0.6 V due to overpotential), the H₂-mediated pathway often prevails (Tremblay et al., 2017). Acetate, ethanol, and methane are the most common products obtained in MES reactors.

Acetate is commonly synthesized by acetogenic microorganisms belonging to the order of Firmicutes *via* the Wood-Ljungdahl pathway, where H₂ acts as an energy source for the reduction of CO₂ to a C₁ carrier (i.e., formate), which is then attached to a second CO₂ molecule to obtain a C₂ compound (Liu et al., 2020). Similarly, methane is produced from CO₂ by Euryarcheota such as *Methanobacterium* and *Methanococcus* sp mainly by the H₂-mediated archaeal-type Wood-Ljungdahl pathway (Borrel et al., 2016), although methane production through direct electron transfer pathways has been also postulated (Lohner et al., 2014). Ethanol production has been reported in MES cells at low pH

(<5.5) and after acetate accumulation (Blasco-Gómez et al., 2019; Srikanth et al., 2018). The findings above suggest that microorganisms produce ethanol *via* the acetaldehyde: ferredoxin oxidoreductase enzyme in response to the diffusion of undissociated acetic acid inside the cell, to avoid an imbalance of the proton motive force and maintain ATP homeostasis (Valgepea et al., 2017). The simultaneous presence of acetate and ethanol can trigger chain elongation through the reverse β oxidation pathway, resulting in the production of C₄ and C₆ carboxylic acids and alcohols (Jourdin et al., 2019; Vassilev et al., 2018). However, to date, the selective and high-rate production of such compounds remains a major challenge in the MES research field.

4.2. Reactor design and operation conditions

The product spectrum and production rates in MES reactors are widely variable and dependent on several factors, including the reactor configuration, electrode materials, inoculum source, and operation parameters such as applied potential, pH, temperature, inorganic carbon source, and purity (for a review, see (Dessi et al., 2021)). Both acetate and methane have been produced with high selectivity and Coulombic efficiency (>90%) by mixed cultures at production rates generally higher than those obtained with pure cultures (Jiang et al., 2019). Product selectivity towards acetate can approach 100% when operating MES reactors under thermophilic conditions (Rovira-Alsina et al., 2020). Selectivity towards ethanol can be enhanced by setting low pH (<4.7), hydrogen partial pressures above 1 atm, and limiting the availability of inorganic carbon to < 100 mg CO₂ dissolved/L (Blasco-Gómez et al., 2019). However, ethanol selectivity by mixed cultures in MES reactors is so far limited to 45% (Srikanth et al., 2018).

The reactor geometry, and in particular the electrode design, is a key factor for achieving high production rates in MES reactors. Optimal electrode materials should be economical, scalable, biocompatible, and conductive, with high surface area, and chemical and mechanical strength (Santoro et al., 2017). While flat, carbon-based electrodes are cheap and easily scalable, they have a low surface for microbial adhesion. In fact, both the highest acetate and methane production rates so far have been achieved by using 3-D structured electrodes, which are promising candidates for scaling-up. Jourdin et al. (2016) deposited multiwalled carbon nanotubes on reticulated vitreous carbon to support the development of a relatively thin (7.5 μm) biofilm that sustained current densities up to 200 A/m² and achieved a remarkable acetate production rate of 790 g/(m²·d). However, the production rate per volume of catholyte was relatively low (0.37 g/(L·d)), suggesting that further improvement is necessary to make such a design scalable.

Romans-Casas et al. (2022) achieved the highest ethanol production

rate from CO₂ of 10.95 g/(m²·d) (0.36 g/(L·d)) with a mixed culture dominated by *C. autoethanogenum* or *C. ljungdahlii*, jointly optimizing key operating factors (pH < 4.5, pCO₂ < 0.3 atm, pH₂ > 3 atm, and acetic acid concentration >6 g/L). Notably, this was achieved in H-type cells equipped with a flat carbon electrode, and the productivity can be further improved by optimizing the cell design. So far, the highest methane production rate per electrode unit of 202 L/(m²·d) was obtained by Zhou et al. (2021) in an electromethanation reactor. The cathodic chamber was filled with anaerobic granular sludge in contact with a Pt–Ti mesh current collector, which acted as a low-cost and easily scalable catalyst for high-rate methane production and demonstrated high tolerance to pH and oxygen disturbances. Tailored, 3-D printed biocathodes, based on conductive carbon hydrogen coated with NiMo-alloy, have been recently demonstrated to facilitate electro-methanogenesis, reaching an unprecedented specific volumetric production of 2.2 L/(L·d) with 99% CE, showing great promise for scale-up (Kracke et al., 2021).

4.3. Potential of CO₂ recycling from closed environments (iCO₂-DAC)

MES is a potential technology to decarbonize flue gas from carbon-intensive industries, but its adoption at a large scale is hindered by its high capital and operation costs, mainly attributed to the anode electrode fabrication and the electricity cost, respectively (Jourdin et al., 2020). Despite the increasing costs associated with carbon emissions, MES is unlikely to become an economically competitive decarbonization technology for large industries in the short term (Wood et al., 2021). However, niche applications with lower capital costs and not requiring high current densities (>10 mA/cm²) may be feasible. Direct capture and conversion of atmospheric CO₂ and its electrochemical conversion to useful products such as ethanol is a negative-emission technology with a potential scale of >33 GT CO₂/y. However, its adoption at scale is still limited by the high energy requirements (1500–4000 kWh/ton CO₂) for capturing CO₂ from the ambient concentration of about 420 ppm (Pace and Sheehan, 2021).

Carbon conversion technologies such as MES can be applied downstream of the carbon capture units to recycle CO₂ from closed, busy environments such as trains, offices, and schools. In such environments, CO₂ can accumulate to concentrations above 2000 ppm (Becerra et al., 2020; Park and Ha, 2008), decreasing the energy demand for its capture in comparison to atmospheric CO₂. The captured CO₂ can be then converted to products such as methane or ethanol, which can be used on-site as a renewable fuel for heating. The following sections explore the hypothetical installation of carbon capture and conversion technologies in three different scenarios: (i) a high school, (ii) an office, and (iii) a metro train cabin, as described in Table 7. For each scenario, the cell design parameters are calculated based on the state-of-art MES cell performance reported in the literature for the production of methane and ethanol to be used as alternative fuels to heat the environments, assuming an external temperature of 10 °C and a target internal temperature of 20 °C. Therefore, the design goal is to determine the fuel requirements for each scenario to increase the room temperature from 10 °C to 20 °C and maintain it constant. A preliminary economic analysis was carried out based on the current market price of electric energy to assess the feasibility of this visionary technology. The parameters and assumptions used for the calculations are summarized in Tables S1 and S2 in the supplementary material section. Further information about the calculations can be found in the supplementary material section. For the CO₂ capture unit, only the electric power required for CO₂ desorption at 80 °C has been included in the calculation, as reports in the literature confirms that the majority of the energy used in the process, specifically a 80% of the energy goes to thermal heat applied to the capture medium, while the remaining 20% to electricity to power the fans, vacuum pumps, compressors as well as other equipment (Beaumont, 2022; McQueen et al., 2021).

For the conversion unit, a carbon capture of 100% was assumed as it

Table 7

Description of the three scenarios with technical and environmental parameters.

Parameter	Scenario		
	High school	Office	Metro cabin
Room height	3	3.5	3
Room length	8	8	20
Room width	8	8	3
Window size (m ²)	2	2	1.2
External walls ^a	2	2	6
Windows	2	1	6
External temperature (°C)	10	10	10
Target indoor temperature (°C)	20	20	20
Heating loss through walls (J/(s K·m ²))	1 ^b	0.6 ^b	1.45 ^c
Heating loss through windows (J/(s K·m ²))	2.5 ^b	2 ^b	5.8 ^d
Average CO ₂ concentration (ppm)	1750 ^e	945 ^f	2265 ^g

^a Walls exposed to outside.

^b Reference: <https://www.omnicalculator.com/construction/heat-loss>.

^c Reference: Zhao et al. (2016); Reference.

^d https://www.engineeringtoolbox.com/heat-loss-transmission-d_748.html.

^e Reference: Becerra et al. (2020).

^f Reference: Allen et al. (2016).

^g Reference: Park and Ha (2008).

is achievable in well-designed, recirculated electrolyzers, and the operating cell voltage was optimistically estimated as 1.8 V (Jourdin et al., 2020). Only the electricity necessary for the actual conversion was included in the calculation, whereas the costs relative to the balance of plant (BoP) were considered negligible.

A comparison between MES-synthesized methane and ethanol to be used for heating purposes is presented in Table 8. Based on the highest production rates and efficiencies obtained so far in the literature, methane appears as the most viable solution, resulting in substantially lower cell volume and power consumption than those calculated for the ethanol scenario. This suggests that further research efforts are required to improve the ethanol productivity in MES cells beyond the current record of 10.95 g/(m²·d) and 49% CE (Srikanth et al., 2018). Another challenge associated with ethanol production is the requirement of a downstream separation and purification step, which will further increase the footprint and costs of the CO₂ conversion plant. Ethanol purification is typically achieved by the energy-intensive distillation process, which can account for at least 40% of the total ethanol production costs (Muhammad and Rosentrater, 2020), or even more with the low product titers (typically <10 g/L) expected in MES reactors.

Conversely, relatively small methane-producing cells (1–2 m³) can

Table 8

Calculated fuel requirements, reactor design parameters, and electricity consumption to convert the captured CO₂ to methane or ethanol and use this fuel to maintain the room temperature at 20 °C.

Parameter	Scenario methane			Scenario ethanol		
	High school	Office	Metro cabin	High school	Office	Metro cabin
Fuel required for temperature control (kg/h)	0.05	0.03	0.57	0.10	0.07	1.06
CO ₂ converted (kg/h)	0.15	0.10	1.61	0.41	0.28	4.52
Cathode electrode required (m ²)	9.4	6.3	102.6	212.8	143.4	2331.1
Cell volume required (m ³)	1.7	1.2	18.9	12.9	8.7	141.8
Power consumed, CO ₂ desorption (kWh/d)	3.8	2.5	41.4	10.6	7.2	116.5
Power consumed, CO ₂ conversion (kWh/d)	30.2	20.4	331.3	59.8	40.3	654.6
Total electricity cost (£/d)	6.1	4.1	67.1	12.7	8.5	138.8

be installed in dedicated utility rooms near schools/offices to generate a pure methane stream that can be directly used as renewable fuel without further treatment (Noori et al., 2020). The CO₂ collected from each room with the portable CO₂ concentrators can be transported to centralized cell stacks, where the CO₂ is desorbed and converted to methane to produce heat for the building. A similar approach applies to the metro cabin scenario, where the MES cells can be installed in the train station.

Based on the calculations of this example, the methane produced can be used for heating the school/office rooms from 10 to 20 °C at an electricity cost of 4.1–6.1 €/d per room, when considering the non-household EU average electricity cost of 180 €/MWh (December 2022). Currently, such cost is higher than the estimated costs for heating using traditional fuels such as gasoline (1.5–2.3 €/d per room) and natural gas (1.0–1.5 €/d per room). Nevertheless, with the rapid technological advancements in MES and the growing concern regarding carbon emissions, the concept of producing sustainable fuels in MES using low-cost, renewable energy sources (e.g., photovoltaic panels on the roof of the building) can soon become a reality.

Regarding the automation needs of the MES studied within this section, automated operation of MES is required for an improved process control, as well for the proper maintenance of MES reactors in the envisioned technology, for example for controlling a minimizing the liquid waste generated from nutrients supply to the MES reactor. This topic is not studied in detail in this work, although is an important implication of the application of MES reactors within indoor facilities, as this will greatly impact on the maintenance needs of the MES reactor.

5. Conclusions

CO₂ capture and recycling technologies in indoor environments are a promising approach to mitigate the detrimental effects of high CO₂ levels on human health. Ongoing research on CO₂ capture materials, such as zeolites, metal-organic frameworks, and amine-based materials, has yielded encouraging results. The utilization of captured CO₂ as a feedstock for MES presents an opportunity to generate valuable products, such as fuels, chemicals, and materials, decreasing the carbon footprint of buildings. As these technologies continue to progress, they are likely to become more efficient, cost-effective, and scalable, enabling broader adoption in various settings. The conversion of CO₂ into useful products provides an incentive for capturing CO₂ from indoor air, thereby addressing both indoor air quality and the carbon footprint of buildings. The integration of CO₂ capture and recycling technologies into heating, ventilation, and air conditioning (HVAC) systems and building materials holds great potential as a sustainable solution for mitigating indoor CO₂ accumulation. For instance, CO₂ capture materials can be incorporated into air filters or building materials, while CO₂ recycling technologies can be integrated into HVAC systems to produce energy. To make these technologies widely applicable in the indoor environment, the development of low-cost, scalable, and energy-efficient CO₂ capture and recycling technologies is necessary. The collaboration between researchers, industry, and policymakers is essential to expedite their development and adoption.

CRediT authorship contribution statement

L.R. López: Validation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Supervision, Visualization, Writing – original draft, Writing – review & editing, Conceptualization, Data curation. **P. Dessì:** Formal analysis, Data curation, Methodology, Visualization, Writing – original draft, Writing – review & editing, Conceptualization. **A. Cabrera-Codony:** Validation, Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Supervision, Visualization, Writing – original draft, Writing – review & editing. **L. Rocha-Melgno:** Methodology, Conceptualization, Writing – review & editing, Validation, Visualization, Writing – original draft. **N.J.R. Krakman:** Conceptualization,

Methodology, Validation, Visualization, Writing – original draft, Writing – review & editing. **M.D. Balaguer:** Writing – original draft, Writing – review & editing, Conceptualization, Data curation, Investigation, Methodology, Validation, Visualization. **S. Puig:** Visualization, Validation, Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Supervision, Writing – original draft, Writing – review & editing.

Declaration of competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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Appendix A. Supplementary data

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