

Decarbonization of Power and Industrial Sectors

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Carbon dioxide (CO₂) is the single largest contributor to climate change due to its increased emissions since global industrialization began. Carbon Capture, Storage, and Utilization (CCSU) is regarded as a promising strategy to mitigate climate change, reducing the atmospheric concentration of CO₂ from power and industrial activities. Post-combustion carbon capture (PCC) is necessary to implement CCSU into existing facilities without changing the combustion block. In general, membrane separation is found to be the most competitive technique in conventional absorption as long as the highly-performed membrane materials and the technology itself reach the full commercialization stage.

CO₂ capture

post-combustion

membrane separation

1. Typical Industrial Sectors and Stream Characteristics for CO₂ Capture

As mentioned in the introduction, energy, industry, and transportation are the largest CO₂-emitting sectors, in which the contribution of global greenhouse gas emissions reached almost 90% in 2019 ^[1]. Massive electrification of transportation points has great potential to reduce the CO₂ footprint from this sector in contrast to the additional increase in the demand for power generation ^[2]. As a result, energy and industrial facilities keep their domination as substantial CO₂ emission sources that can be captured and utilized by CCU technologies.

Based on the fuel type, CO₂ emitters in power generation involve coal, oil, and natural gas-fired power stations, and waste-to-energy power plants. Apart from these, the industrial point sources of CO₂ constitute cement plants, crude oil refineries, iron, steel, and petrochemical factories ^[3].

Referring to CO₂ capture from power plants and industrial sectors, the characteristics of the outlet flue gas stream are one of the main factors to be considered regarding the amount of CO₂ in the stream. The pressure of the exhausts is nearly the same for all types of power plants at atmospheric pressure, in contrast to the different levels of temperature depending on the various conditions. For instance, in natural gas-fired combined cycle power plants, the temperature is not lower than 90 °C, in order to avoid condensation and damaging the chimney, and not above 110 °C, in order to utilize as much heat as possible. However, for most CO₂ separation applications, the flue gas is required to be between the temperatures of 40–60 °C. In terms of stream compositions, coal- and oil-fired power stations have similar products of combustion with higher amounts of impurities such as SO_x and NO_x in

comparison with natural gas combustion products, which are relatively clean with trace amounts of NO_x . Since the CO_2 content in the flue gas plays a vital role in the capture cost [3][4][5], coal- and oil-fired plants have advantages, with higher amounts of CO_2 content, at an average of 12–14 mol%, compared to natural gas-fired power plants and combined cycle counterparts, at an average of 8–9 mol% and 3.5–4.5 mol%, respectively. From this perspective, the cost of CO_2 capture for combined cycle power plants is more expensive than others due to the low CO_2 concentration in the stream, although the efficiency of the plant is the highest. Coal-fired power stations have, however, the largest contribution to global CO_2 emissions, at around 10 Gt annually among all other types of power plants.

As for the cement industry, since the CO_2 emissions come from the energy-intensive limestone calcination process [6] and the combustion of fossil fuels, the cement plant flue gases have a high carbon content, at an average of 18–22 mol%. The temperature of the flue gas in cement production can be between 150–350 °C depending on the type of raw material and preheating stages. High-temperature flue gas must also be utilized with appropriate techniques to cool down it prior to flue gas decontamination. The steel production industry is the largest fuel consumer and emits a high CO_2 content in exhaust gas, similar to cement production. Steel and cement industries are responsible for 14–19% of global greenhouse gas emissions (annually, 2.6 and 2.3 Gt CO_2 emissions, respectively). For this reason, methods and techniques of decarbonization in these industries are being developed. When it comes to other CO_2 -emitting industries, they also emit CO_2 resulting from fuel combustion and chemical reactions. However, it can be seen that the annual CO_2 emissions are much lower compared to fossil fuel power plants, cement, and steel industries on a global scale.

2. Membrane-Based CO_2 Capture Technologies

2.1. Membrane Separation

The membrane separation process is a process that uses a special module called a membrane to separate gases in a gas stream by rejecting contaminants (retentate) and passing desired components (permeate) through the membrane module. In this process, pre-treated flue gas containing CO_2 is sent to the high-pressure side of the membrane, and CO_2 is recovered from the low-pressure side.

Membrane separation could be a promising technology with operating parameters that go beyond current technologies, as they often feature a small footprint, easy scaling, integration into existing technologies, low operating costs, as well as low energy consumption. Membrane separation in PCC is expected to be a technology that can compete with benchmark absorption. There are mainly three types of membranes based on its material, which are organic (polymeric), inorganic (non-polymeric), and mixed matrix membranes (hybrid organic and inorganic) [7]. Apart from that, membranes can also be used as a membrane contactor, enhancing the solvent-based CO_2 capture processes. Regarding the performance of the membrane, it is highly related to the selectivity and permeability, which is the rate of passive diffusion of molecules through the membrane. In this CO_2 capture method, mainly hollow fiber, a spiral wound, and flat sheet membrane modules are used [8].

Even though there are several commercial applications of membranes in different fields, the number of commercial membranes special for CO₂/N₂ separation is limited. For instance, Kárászová et al. [9] have reviewed various applications of commercial and emerging lab scale membranes which have been tested with flue gas. They also emphasized the existing achievements and barriers of potential membranes and evaluated their conditions of competitiveness with monoethanolamine (MEA) absorption. Apart from that, Chen et al. [10] reviewed three types of polymeric, non-polymeric, and mixed matrix membranes based on pre, oxy-fuel, and post-combustion CO₂ separation, and concluded that more pilot plant tests should be implemented under real flue gas conditions of different fuel combustion products in post-combustion CO₂ capture.

2.1.1. Organic Membranes

In post-combustion CO₂ separation, organic membranes are prepared by polymers such as polyacetylene, polysulfone, polycarbonates, polyetherimides, polyaniline, poly(phenylene oxide), poly(ethylene oxide), and polyvinylamine. Although the polymer-based membranes are used at low temperatures, and plasticization and swelling by water are the main issues, their relatively low cost, diversity, and easy control of processing can greatly outweigh their drawbacks. Additionally, developing the polymer and the combination of the chemical elements during the membrane preparation can be manipulated, which gives an extra advantage for this type [10][11]. In terms of transport mechanism, facilitated transport and solution-diffusion (non-facilitated transport) membranes are reported as the most widely applied and recognized in post-combustion carbon capture [7]. In the solution-diffusion transport mechanism, CO₂ dissolves into the dense membrane followed by its diffusion throughout it. This mechanism is usually divided into rubbery, glassy, and co-polymeric membrane types which have different gas separation performances. For instance, rubbery polymeric membranes have higher permeability with inadequate selectivity, while the glassy type has opposite characteristics [12]. As for the facilitated transport membranes, CO₂ molecules are attached by reactive carriers, forming a temporary product via reversible chemical reaction. Unlike solution-diffusion transport, facilitated transport membranes have relatively higher selectivity and permeability due to the enhancement by both aforementioned transport mechanisms [13]. Facilitated transport membranes are seen as one of the promising technologies for the flue gases from both power and industrial sectors, owing to their ability to separate CO₂ in low partial pressure.

2.1.2. Inorganic Membranes

Non-polymeric membranes are usually based on ceramic, metal, glass, carbon, and zeolite, which can practically provide better chemical and thermal stability than those of polymeric counterparts. For instance, alumina, titania, and zirconia are considered as the best choice for higher temperatures and harsh conditions, in spite of their relatively high cost [14][15]. Regarding the separation of CO₂ from flue gas, mainly N₂, since the dipole moments of both CO₂ and N₂ are zero, the ion transport mechanism is not applicable in this case. It should also be highlighted that their kinetic diameters are quite similar in size, which are 0.333 nm and 0.357 nm, respectively. Inorganic membranes can be mesoporous, microporous, and dense in type with regard to the purpose of use. On the one side, the permeability of microporous membranes is low with higher selectivity, while the characteristics of mesoporous membranes are opposite. Dense membranes, on the other side, have superior performance with their

selectivity compared to mesoporous and microporous, though their permeability is lower [16]. According to Li et al. [17], several types of inorganic membranes, particularly zeolites, have reached the commercially interesting area. However, further cost reduction is needed to deploy them commercially in CO₂ capture processes. Moreover, the processing of inorganic membranes is challenging and they tend to break easily without plastic deformation.

2.1.3. Mixed Matrix Membranes

Many efforts to reach better performance of selectivity/permeability trade-off relationship on Robeson upper bounds have led to the fabrication of new a membrane technology by hybrid organic and inorganic (mixed matrix) membranes, further improving the polymeric membranes. Mixed matrix membranes are prepared from a polymer matrix filled by inorganic fillers, such as carbon nanotubes [18], metal organic frameworks [19], and zeolites [20], enabling them to take an advantage of both organic and inorganic membrane properties. Several review papers have analyzed the mixed matrix membranes and compared them to the traditional polymeric counterparts. For example, Kamble et al. [21] thoroughly reviewed and analyzed the recent works in the field of mixed matrix membranes and their inorganic fillers, emphasizing advances and the current problems of inorganic fillers materials.

2.2. Hybrid Membrane Systems

2.2.1. Membrane Contactors

Another application of membranes in CO₂ capture processes is membrane contactors, which are a combination of membranes with solvent sorption. Membranes can be used at the gas–liquid interface, separating two phases by allowing only CO₂ molecules pass through the membrane (dense or microporous) to the solvent side [22]. Generally, the hollow fiber and flat-sheet membrane contactors are two of the most researched technologies. In practice, the hollow fiber module is commonly used. In terms of the type of membrane material, polymeric or inorganic membranes could be chosen in response to the conditions applied. Since the process is based on the combination of membrane and solvent absorption, there are some requirements for the selection of membrane material and appropriate absorbent, including the limitations of both technologies. For instance, the selected material should provide the features of high hydrophobicity to minimize the wetting effects, thermal and chemical stability to maximize the durability, and high porosity to minimize the mass transfer resistance [23]. Absorbent characteristics also play an important role, as they have a direct influence on the process efficiency and economic aspects [24]. The commonly used solvents in this process are alkanolamines [25], amino acid salts [26], inorganic solvents [27], ammonia [28], and ionic liquids [29][30].

Membrane contactors are considered a promising technique since they allow for the avoidance of several issues such as channeling, flooding, and foaming in the conventional packed and tray columns. Apart from that, the equipment size of the column can significantly be decreased, up to 70% in size and 66% in weight, generating 4–15 times higher mass transfer area per unit volume over the traditional technique [22][31]. However, in addition to wetting and fouling of the membrane, one of the main problems of this method is extra resistance in mass transfer due to the availability of the membrane between these two phases.

2.2.2. Hybrid Membrane-Absorption

Another possible application of membranes is to use them as an additional unit in the absorption process in order to improve the driving force of the mass transfer, further concentrating CO₂ in the flue gas. This hybrid technique was initially studied by the University of Texas at Austin in collaboration with Membrane Technology and Research [32]. They integrated the selective membrane recycle unit into the absorption process in series and parallel methods. According to the results, through the best parallel configuration, the size of the absorber and flue gas flowrate can be reduced by nearly half, increasing the CO₂ content in the flue gas from 13% to above 23%. Apart from that, several other studies have been undertaken in this field with respect to design and operational variations [33][34], economic cost evaluation [35], and possible applications in natural gas combined cycle (NGCC) power plants in selective exhaust gas recirculation (SEGR) [36][37]. Overall, this technique can be a feasible option for low CO₂ partial pressure flue gases, particularly coal-fired and NGCC power plants (around 4% and 12–14% by volume, respectively), as long as more pilot plant tests are implemented under the real flue gas conditions.

3. Comparison of Membrane and Other Technologies for Post-Combustion CO₂ Capture

3.1. Technology Readiness Level and Scalability

Technology readiness levels (TRL) are the technological maturity assessment of the technology developed by The National Aeronautics and Space Administration (NASA) that dates back to its origin in the 1970s. TRLs are divided into nine levels, starting from initial observations and concepts at TRL 1 and at TRL 9, at which the technology is the most mature [38].

Although carbon capture and storage technology (CCS) is one of the solutions to the climate change problem, there are several challenges from an economic point of view involving the interconnection between the emitting points and CCS technology. According to Global CCS Institute [39], there are 30 commercialized carbon capture, storage, and utilization facilities with a global CO₂ removal potential of around 43 million metric tons of CO₂/year. In addition to this, 164 other CCS projects are in the stage of construction and development. If all announced CCS facilities are launched, the potential of CO₂ removal will increase multiple times. However, several factors, such as the current global economic crisis, the relatively low cost of CO₂, project installation, and operating costs, may lead to the suspension of the efforts to launch these facilities. For example, the Petra Nova CCS project, capable of 1.4 million metric tons of CO₂ removal annually, was shut down on 1 May 2020 due to the significant decrease in the price of oil, although around 1 billion USD was already spent on this project [40].

Post-combustion carbon capture based on an amine absorption technology is fully commercialized (TRL 9) for large-scale applications and is used as a benchmark rather than other liquid solvents, piperazine (PZ), chilled ammonia, ionic liquids, alkaline solutions, and blended solvents [41], and other capture technologies. Amine, mostly MEA, absorption needs a significant quantity of heat for rich solvent recovery and power for CO₂ compression, as well as for electrical equipment. The CO₂ capture cost for the absorption process ranges between 50 USD CO₂/ton

and 100 USD CO₂/ton depending on industry and solvent type [42][43][44]. The second-generation post-combustion absorption technologies involve the PZ solvent-based absorption (TRL 7-8) and the chilled ammonia processes (CAP) (TRL 7). The PZ chemical absorption has been tested at the NGCC power plant and is ready to capture CO₂ in a large-scale application. The CAP is moving toward the commercialization stage after testing in different flue gas streams and is suitable for large-scale applications. An ionic liquid (IL) absorption technology is still in the research and development stage (TRL 2-3). ILs should be developed to overcome challenges such as toxicity, solvent cost, viscosity, low absorption capacity, corrosive nature, and hygroscopicity. Novel phase-change solvents (TRL 5-6) are currently being developed at a low-rate pilot scale in a relevant environment [45] and are expected to be available for commercialization in the next years.

While cryogenic separation for post-combustion carbon capture is the best technology to obtain pure CO₂ in liquid or solid form, it is not yet scaling up (TRL 3-4) in post-combustion carbon capture due to the high demand for energy in the low CO₂ composition [46][47]. Apart from that, flue gas impurities, particularly water, need to be removed in order to avoid blockage issues caused by solid ice formation of water at low temperatures, which further increases the cost of separation. Therefore, it should be noted that this technology might only be feasible when the cold energy source, such as liquefied natural gas vaporization process, is available at near locations [48].

The calcium looping process can capture CO₂ from a large-scale power plant and other industrial flue gases [49]. Due to the high temperature of the processes in the carbonator/calcliner fluidized bed reactors and additional requirements for oxygen, there are difficulties in implementing the calcium looping process from an economic point of view. For instance, in carbon capture from natural gas combined cycle power plant flue gases, the CO₂ capture cost of calcium looping is between 90 USD CO₂/ton and 100 USD CO₂/ton, which is significantly more than the benchmark amine (MEA) capture process [50]. However, this technology seems more attractive because of the inexpensive natural limestone, the possibility of diverting used CaO to cement production, power/steam generation from waste heat, and its being much less hazardous to the environment compared to solvents.

Regarding CO₂ bio-fixation, CO₂ can play a crucial role in boosting algae and crop cultivation. Microalgae photosynthesis, from the scalability context, is possibly assessed as a medium or even higher scale technology, considering its significant limitations including large space requirement, wastewater availability, algae sensitivity to the impurities, and high cost of control. Unavailability of sunlight at nighttime also affects the efficiency of CO₂ removal. However, it is considered as the best-fitting technique for flue gas streams with relatively low CO₂ content, such as the flue gas from NGCC power plants, without affecting the efficiency of the plant [51]. CO₂ consumption in greenhouses is becoming another trend for yield boosting in many countries. For instance, the Netherlands stands out as a country in which CO₂ is used in greenhouses up to 6.3 Mt per year [52]. Although carbon bio-fixation is generally at low TRL levels [53][54], its estimation for the end of this decade is relatively large, as the demand for biofuels and bio-based feed products rises [55].

The adsorption process is assessed as a viable method for gas purification. When it is implemented in PCC, challenges occur related to flue gas characteristics. In terms of pressure-based sorbent processes, the suitability of these processes to CCS highly depends on CO₂ content from an economic point of view. An energy penalty will be

significantly higher at low CO₂ partial pressure sources than other high CO₂ partial pressure gas streams (hydrogen production, steel, and other industrial sectors) for PSA/VSA adsorption. Although the CCS facility based on a VSA (VPSA) was demonstrated (TRL 8–9) by Air Products at the steam-methane reformers (CO₂ from syngas), this technology can be a feasible option to commercialize for post-combustion CO₂ capture with higher CO₂ concentration flue gases [46]. The temperature swing adsorption (TSA), particularly the Kawasaki Carbon Capture System [56] and Svante VeloxoTherm™ Rapid Cycle Temperature Swing Adsorption [57], reached a demonstration-scale with innovative sorbents and adsorption reactors. In other emerging CO₂ capture technologies by adsorption (TRL < 5), scientific research and development are being conducted in order to solve problems depending on the limited scale of sorbent capacity, selectivity under realistic pressure conditions, moisture sensitivity, and slow kinetics. In terms of adsorbent materials, zeolites and carbon-based solid sorbents are more mature than other adsorbents and are widely used in large-scale applications [58].

Membrane gas separation is becoming one of the promising options in CO₂ capture from fuel combustion flue gases. Although polymeric membranes have already been commercialized in natural gas processing [57], in post-combustion CO₂ separation, there are only three polymeric membranes so far that have been demonstrated on a pilot scale reaching the level of TRL 5–6. These are Polyactive® membrane by Helmholtz-Zentrum Geesthacht in Germany, Polaris® membrane by Membrane Technology and Research Institute in the USA, and fixed-site-carrier membrane by the Norwegian University of Science and Technology in Norway [59]. Nevertheless, there are still some major issues remaining when tested under real flue gas conditions. These problems include humidity-based membrane resistance, thermal influences on transport properties (permeability and selectivity), stability of the membrane layer for a long period, and the impurities of the flue gas [9]. However, in the last decade, significant progress has been made in CO₂ capture using polymer matrix membrane processes. According to Janakiram et al. [60][61][62], water content in the flue gas is no longer an impurity, but it is a promoter. Humidity in the module can increase membrane permeability. They tested hollow fiber modules of hybrid facilitated transport membranes for flue gas coming from the cement industry. Membrane performance improved when there was more water in the flue gas stream. As for the inorganic membranes, they are still in the lab scale development, with TRL 3–4 [48], due to several problems related to the permeability of dense membranes, difficult processing, and high cost, as mentioned in the previous section. According to Jusoh et al. [63], the fabrication cost of membrane modules for zeolite is 150 times higher than polymeric hollow fiber membrane modules, at 3000 USD and only 20 USD per square meter, which leads to its limitations in industrial applications. In terms of hybrid membrane-absorption techniques, on the one hand, there are several field trial and pilot scale studies of membrane contactors in CO₂ capture. However, membrane wetting and its compatibility with solvent still remain as the major issues. For instance, the latest pilot study of membrane contactors, reported by Scholes et al. [64], revealed that membrane contactors can be a feasible option for industrial applications, though the energy consumption in the pilot study is higher than conventional capture technique (less than 4.2 MJ/kgCO₂) due to thermal losses in membrane modules and energy integration issues. Membrane separation integrated absorption technology, on the other hand, was assessed at a TRL of 4 as a conceptual study by the Global CCS Institute in 2021 [57]. Freeman et al. [65] conducted a bench scale study of hybrid membrane-absorption CO₂ capture from coal-fired flue gas.

3.2. Overall Technology Comparison for CO₂ Capture

Here is a general discussion about five different CO₂ capture technologies and their comparison based on CO₂ purity and recovery, scalability, TRL, capital expenditures (CAPEX), operational expenditures (OPEX), and environmental benefit without certain metrics. In order to carry out the best realistic comparison among these technologies, there are many factors and conditions to consider for each specific case and technology that makes the work more challenging. Therefore, the following discussion and comparison are conducted to make a general overview of the key highlights of each technology.

Regarding the captured CO₂ recovery and purity, the cryogenic method is evaluated as the most suitable technology, capable of 99.99% product purity, since this process is based on obtaining CO₂ in liquid or solid form at a very cold temperature. This technology has been applied in mainly air separation units, blue hydrogen production, natural gas processing plants, and biogas processing so far. Nevertheless, the challenges that the process is energy intensive and requires a high concentration of CO₂ in the flue gas stream are hindering this technology's commercial deployment and scalability assessment in the post-combustion field. Absorption, calcium looping, and adsorption (TSA) also have great potential to obtain a relatively pure CO₂ product, but the equipment cost and operating cost also increase. Membrane separation and adsorption (VPSA) processes are not favorable in this context due to the necessity for multiple stage installation, which leads to additional CAPEX and OPEX. CO₂ bio-fixation can be assessed for only CO₂ recovery rate, which is quite a bit lower than other techniques, since the process directly utilizes the captured CO₂.

From the scalability point of view, absorption by amines can be evaluated as the most reliable technology and capable of capturing CO₂ at a large scale, followed by the calcium looping process, which is not fully mature yet. For example, Zanco et al. [66] performed a comparative study of absorption, adsorption, and membrane technologies, selecting the most mature and effective techniques under the same condition for all. According to the results, adsorption and membrane technologies are more cost competitive than the absorption process in terms of small-scale plants. However, at large scale plants and higher CO₂ recovery rates, absorption is found to be the most cost-effective. Carbon bio-fixation can also be applied for large scale power plants without a need for flue gas pre-processing.

When the TRL levels are considered, as discussed previously, the number of techniques that have reached maturity is higher in the absorption process than others, followed by adsorption techniques. However, membrane separation (polymeric and mixed) is assessed as the fastest developing technology due to its wide range of characteristics that have not yet been studied well compared to the cryogenic separation method, which attracts the least attention in terms of commercial deployment in PCC.

Regarding the overall CAPEX and OPEX, facilitated transport membranes (polymer mixed) can be more potent than other techniques due to their acceptance of water, which makes them unique among other membrane types [62], although this membrane is not commercially available and not fully mature yet. As membrane science has been developing rapidly in recent years, its advantages, such as easy scale-up, small footprint, and, particularly, lower energy consumption, can overcome the conventional benchmark absorption technology. In terms of carbon bio-fixation, this technique requires very high capital investment due to the large number of photobioreactors'

installation and large area requirements. In contrast, OPEX is relatively low, as the process goes at ambient conditions. Ca-looping also seems acceptable from the OPEX, since the sorbent (CaO) is relatively cheap, but the additional air separation unit leads to an increase in the CAPEX. However, in the case of amine scrubbing and solid sorption, the CAPEX and OPEX highly depend on which type of solvent or sorbent and techniques are used, their scalability, availability at low cost, site conditions, maturity level, and many other factors. For instance, in the MEA absorption process, the majority of the OPEX is connected to the solvent regeneration energy and its heat integration to the point source, followed by solvent loss and its degradation, while CAPEX varies in response mainly to the dimensions of the columns, packing or tray type, and heat exchangers based on the flowrate, CO₂ partial pressure in the flue gas, purity requirement of CO₂, and capture rate.

As for the environmental concerns, it is very difficult to evaluate without a rigorous lifecycle analysis of each technology. However, in general, carbon footprint of absorption with amines can be higher than other techniques since there is an extra emission of solvent, which has an even higher impact on the environment than CO₂. Solid sorption, calcium looping, and cryogenic separations also possibly have more environmental stress (depending on the energy intensity and its source) than membrane separation and carbon bio-fixation, which are both considered as the most energy-efficient, with less of a carbon footprint.

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