

Carbon capture for decarbonisation of energy-intensive industries: a comparative review of techno-economic feasibility of solid looping cycles

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Abstract Carbon capture and storage will play a crucial role in industrial decarbonisation. However, the current literature presents a large variability in the techno-economic feasibility of CO₂ capture technologies. Consequently, reliable pathways for carbon capture deployment in energy-intensive industries are still missing. This work provides a comprehensive review of the state-of-the-art CO₂ capture technologies for decarbonisation of the iron and steel, cement, petroleum refining, and pulp and paper industries. Amine scrubbing was shown to be the least feasible option, resulting in the average avoided CO₂ cost of between 62.7 €·t_{CO₂}⁻¹ for the pulp and paper and 104.6 €·t_{CO₂}⁻¹ for the iron and steel industry. Its average equivalent energy requirement varied between 2.7 (iron and steel) and 5.1 MJ_{th}·kg_{CO₂}⁻¹ (cement). Retrofits of emerging calcium looping were shown to improve the overall viability of CO₂ capture for industrial decarbonisation. Calcium looping was shown to result in the average avoided CO₂ cost of between 32.7 (iron and steel) and 42.9 €·t_{CO₂}⁻¹ (cement). Its average equivalent energy requirement varied between 2.0 (iron and steel) and 3.7 MJ_{th}·kg_{CO₂}⁻¹ (pulp and paper). Such performance demonstrated the superiority of calcium looping for industrial decarbonisation. Further work should focus on standardising the techno-economic assessment of technologies for industrial decarbonisation.

Keywords industrial CO₂ emissions, CCS deployment, carbonate looping, net-zero industry, carbon capture benchmarks

1 Introduction

CO₂, whose residence time in the atmosphere is around

100 years [1], is one of the six greenhouse gases (GHG) targeted by the Paris Agreement, signed by 197 countries. These countries are committed to reduce their GHG emissions and keep global warming at least well-below 2 °C. Therefore, to comply with this agreement, the net GHG emissions need to become zero or even negative between 2055 and 2080 [2]. For that reason, several routes for decarbonisation have been identified, including improvement in the material and energy efficiencies, reduction of dependence on fossil fuels and implementation of carbon capture and storage (CCS) [3–5]. Since the major contributors to the global CO₂ emissions are iron and steel, cement, petroleum refining and pulp and paper industries, decarbonisation of these industries must be prioritised to comply with the Paris Agreement. Due to their high energy demand, these industries are classified as energy-intensive industries (EIIs). It needs to be emphasised that these four industries are responsible for around 68% of the industrial direct CO₂ emissions [6] and accounted for around 20% of the global direct CO₂ emissions in 2016 [7].

The decarbonisation of these industries is crucial to reaching net-zero emissions, as evident in the previous review papers published in recent years [5,8–17]. The scope and main conclusions of these reviews are summarised in Table 1. Although some of the reviews include comprehensive appraisals of EIIs, there are significant limitations that need to be addressed. Fennell et al. [10] have conducted an overview of the economic performance of CCS for EIIs. However, their review accounted only for a limited number of studies. Kuramochi et al. [12] have performed a comprehensive review that compared the techno-economic feasibility of CCS technologies in selected industries. However, the development of these technologies was in an early stage at the time this review was published. Markewitz et al. [14] have compared the efficiency, energy consumption and technical viability of the main CCS routes for industrial decarbonisation

without focusing on a specific industry. Napp et al. [15] have also carried out an extensive review of EII's decarbonisation, but did not include a complete detail and analysis of the CO₂ capture technologies due to the lack of available data. The application of CCS to different EII's was covered in detail in the systematic review carried out by Leeson et al. [13]. However, the main goal of that review was to compare the economic performance of CCS technologies, neglecting their thermodynamic performance. Moreover, Yang et al. [9] have done a

comprehensive review about CCS and biomass combined with CCS (BECCS). However, it only focused on the environmental and economic performance, neglecting the thermodynamic performance presented by each CCS technology. Although calcium looping has been recently shown as promising CCS technology by integration on the iron and steel and pulp and paper industries, it was not considered by Yang et al. [9]. Moreover, that review considered only the economic assessments carried out one decade ago.

Table 1 A summary of the review studies about EII's decarbonisation

Ref.	Review scope	Main conclusions
[12]	<ul style="list-style-type: none"> • Extensive review about the CO₂ capture technologies applied to three industries, iron and steel, cement, petroleum refineries and petrochemicals; • Techno and economic assessment of the technologies, based on standardisation of performance parameters; • Estimation of potential reduction of CO₂ emissions and respective costs for what they categorised as short/medium term and long term technologies. 	<ul style="list-style-type: none"> • No dominant technology for any of the industries analysed; • The costs could be so diverse that for the cement industry, they could vary from 29.2 to 141.5 €·tCO₂⁻¹ avoided when the carbon capture is obtained by calcium looping applied to the pre-calciner or by absorption with monoethanolamine, respectively; • Short-mid-term technologies may have a cost of 43.2–70.2, above 70.2 and 54.0–64.8 €·tCO₂⁻¹ avoided in the iron and steel, cement and refining industries, respectively; • Long-term technologies could be achieved at lower cost, 32.4–59.4, 27.0–59.4 and 32.4 €·tCO₂⁻¹ avoided in the iron and steel, cement and refining industries, respectively; • The economic feasibility of these technologies is strongly dependent to the power market once the excess electricity produced is exported to the grid.
[14]	<ul style="list-style-type: none"> • Overview of CO₂ capture, transport and utilisation; • Comparison of efficiency, energy consumption and technical viability of the main routes available for CCS; • Assessment of environmental impact of CCS technologies; • Summary of the largest CCS projects worldwide. 	<ul style="list-style-type: none"> • At that time, there was no best CCS technology, but gas separation by membranes and solid looping cycles were seen as promising technologies; • The CO₂ use as a raw material could have an effective contribution to a decrease in CO₂ emissions.
[10]	<ul style="list-style-type: none"> • Brief review about the costs of CCS application to five industries, iron and steel, cement, refinery, biomass and high purity sources. 	<ul style="list-style-type: none"> • Unlike the power industry and due to the heterogeneity of the processes involved in petrochemical industries, the costs could reach 150.9 €·tCO₂⁻¹ avoided; • The cost of cement decarbonisation could be as low as 17.2€·tCO₂⁻¹ avoided when calcium looping is employed; • In the cement and iron and steel industries, which involve high-temperature processes, the integration of solid looping cycles seems to be the distinctive solution for the deep decarbonisation of these two industries.
[15]	<ul style="list-style-type: none"> • Comprehensive review about decarbonisation of three industrial industries, iron and steel, cement and refineries; • Review of the energy-efficient technologies in these industries as well as the potential of their implementation; • Assessment of different routes for these industries decarbonisation; • Discussion about the policies that should be adopted as a strategy to mitigate CO₂ emissions. 	<ul style="list-style-type: none"> • An energy/emissions monitorisation system should be implemented and the best available technologies; • Fuel switching, CCS capture, co-location of industries and re-design of the processes were proposed as routes for these industries decarbonisation; • The replacement of fossil fuel by biomass and wastes should be encouraged and CCS should be seen as an option for deep decarbonisation.
[13]	<ul style="list-style-type: none"> • Exhaustive review about different CCS technologies employed in five industries, iron and steel, cement, refining and petrochemical, pulp and paper and high purity sources; • Technical and economic assessment of these technologies; • A mathematical model was proposed to estimate the costs of CCS implementation until 2050; • A sensitivity analysis was also performed. 	<ul style="list-style-type: none"> • The studies about the costs of CCS implementation were scarce and practically non-existent for the pulp and paper industry; • Costs in the other industries could vary from 17.8–106.8 €·tCO₂⁻¹ avoided, which contributes to a high economic uncertainty associated with these technologies and consequently to the delay of its commercialisation; • Delaying CCS implementation will lead to higher costs.
[8]	<ul style="list-style-type: none"> • Review of technologies and policies available to reduce CO₂ emissions in pulp and paper, iron and steel and cement. 	<ul style="list-style-type: none"> • The biomass conversion in heat and power for the pulp and paper plant seems to be the key to decarbonise this industry; • Unlike the previous industry, in the steelmaking process and refining industry, there is not a dominant route although the replacement of fossil fuel by BECCS is mentioned [28]; • In the cement industry, the carbon capture, usage and storage (CCUS) employing calcium looping is the most straightforward route to decrease CO₂ emissions; • Even though some of the technologies are near commercial, policies and incentives to research must be put in practice to reach the Paris agreement targets.
[11]	<ul style="list-style-type: none"> • Evaluation of carbon capture utilisation (CCU) technologies as well as the potential of use pre- and post-combustion capture in the thermal power, EII's and other industries; • Detailed list of commercial projects where carbon capture was already implemented and proved that is a feasible option in the CO₂ emissions abatement. 	<ul style="list-style-type: none"> • The CO₂ utilisation in conjunction with the use of incentives in implementing carbon capture technologies must be seen as a route to follow.

(Continued)

Ref.	Review scope	Main conclusions
[5]	<ul style="list-style-type: none"> • Comprehensive review of decarbonisation of seven industries, iron and steel, cement, petrochemical, pulp and paper, ceramics, glass and food; • Roadmap for the deep decarbonisation of these industries and their potential to reach the imposed targets until 2050. 	<ul style="list-style-type: none"> • CCS, biomass and bio-based waste, process heat provision, alternative feedstock, electrolysis, combined heat & power (CHP), industrial ovens and membrane process were the main routes identified; • CCS was the only trans-sectional option that had the potential to mitigate the CO₂ emissions with a potential between 25% and 55% for total decarbonisation; • All the work done so far is not enough for deep decarbonisation and there is no yet a dominant technology being necessary to develop new technologies.
[16]	<ul style="list-style-type: none"> • Review of technologies and policies to reach net GHG emissions by 2050–2070 in the cement, iron and steel, chemical, and plastics industries. 	<ul style="list-style-type: none"> • Use of mineral and chemical admixtures, re-design building techniques to decrease the demand for concrete, improvement of the thermal efficiency of processes during cement production, fuel switching, electrification of cement kilns and CCS were the main options to full decarbonisation of cement industry; • In the iron and steel industry, the implementation of CCS and the replacement of fossil fuels by hydrogen or direct electrolysis were the main paths for reducing the CO₂ emissions of this industry; • Development of clean processes, by avoiding the use of fossil fuels, the use of biomass feedstocks and recycled chemicals, and the use of CO₂ as feedstock, the improvement of separation technologies and CCS were identified as the main routes to decarbonise the chemical and plastics industry; • Although the low carbon technologies will become cheaper, they are not enough for deep decarbonisation across the studied industries. Certain policies such as carbon pricing, government incentives for research, development and deployment, and energy efficiency or emissions standards should be adopted.
[17]	<ul style="list-style-type: none"> • Review of technical options, policies and barriers to decarbonise the iron and steel, mining, cement and refinery industries, taking the Swedish case as reference. 	<ul style="list-style-type: none"> • Electrification, fuel switching to low carbon fuels, CCS and when possible, a fossil free production are necessary deep decarbonisation of EIIs; • The use of less raw materials, improvement of material efficiency and implementation of circular economy were also identified as decarbonisation pathways; • There is necessary keep going development of decarbonisation technologies and its test at large scale in order to reach the commercial viability; • The incentive of low carbon technologies should be a priority, which can be achieved by implementation of new policies and incentives.
[9]	<ul style="list-style-type: none"> • Comprehensive review of decarbonisation of five industries, iron and steel, cement, petrochemical, pulp and paper and hydrogen; • Techno and economic assessment of the technologies CCS or BECCS based on a standardisation of performance parameters; • Estimation of CO₂ mitigation potential and respective costs. 	<ul style="list-style-type: none"> • CCS only had the CO₂ mitigation potential up to 74% however, this figure could reach the 2548% for BECCS implementation in pulp and paper industry; • The iron and steel, pulp and paper and hydrogen could become carbon negative industries; • These results could be achieved with a CO₂ avoided cost lower than 100 €·t_{CO₂}⁻¹; • There were some discrepancies in literature regarding the potential and the economic assessment of the reviewed technologies.

Other comprehensive reviews of EIIs decarbonisation have also been published in the last five years [5,8,16,17]. Nevertheless, these focused on appraising all the available routes and policies for deep decarbonisation of EIIs. As a result, the CCS option was not evaluated in detail. Furthermore, Nurdiawati and Urban [17] have based their work on the Swedish industrial scenario and the considered technology options were selected specifically for the iron and steel and cement industries. Koytsoumpa et al. [11] have only analysed the high-level CCS options (i.e., pre- and post-combustion capture) and have not focused on a particular technology. Furthermore, despite being the fourth most polluting EII, the pulp and paper industry was considered in 50% of the reviews in the current literature [5,8,9,13,17]. Although carbon capture is forecasted to play a critical role in industrial decarbonisation, the recent progress in this field has not been thoroughly reviewed, analysed and discussed. Significant progress has been made in reducing the energy intensity, and improving the economics, of CCS, especially in the past 5 years. However, the majority of the reviews in this area were published between 2012–2017. As a result, the data and analysis presented in these reviews focus solely on the early-stage development of the CCS technologies. Solid looping cycles were recently shown to have a high potential to decarbonise energy and industrial systems,

showing a techno-economic superiority compared with other CCS technologies. In the energy sector, the retrofits of calcium looping to power plants were shown to reduce the efficiency penalty associated with CCS as a result of availability of high grade heat [18]. It is important to emphasise that limestone, the most commonly considered sorbent for calcium looping, is also used in the iron and steel [19], the cement [20], and the pulp and paper [21] industries. Therefore, these industries can take advantage of their inherent decarbonisation potential at lower energy requirements and CO₂ avoided costs when calcium looping is implemented. For example, the retrofit of calcium looping in the cement industry was shown to result in the CO₂ avoided cost of 58.6 €₂₀₁₄·t_{CO₂}⁻¹, which was around 25% lower than that of amine scrubbing retrofit (80.2 €₂₀₁₄·t_{CO₂}⁻¹). Such a decrease in the cost of CO₂ avoided due to calcium looping retrofit can reach up to 70% in the iron and steel industry (12.5–15.8 €₂₀₁₀·t_{CO₂}⁻¹, calcium looping against 45–60 €₂₀₁₀·t_{CO₂}⁻¹, amine scrubbing).

This work aims to present a comprehensive review of CCS technologies for the decarbonisation of the EIIs, with the main focus on solid looping cycles. Although the concept of solid looping cycles has been known for 20 years, the development of this technology has only intensified in the last decade. For this reason, this review

does not include studies published before 2009. This work aims to critically appraise the viability of solid looping cycles, particularly calcium looping, as a route for decarbonisation of EIIIs. For this reason, a techno-economic comparative assessment of CCS for the four EIIIs was performed. As shown later in this review, the current literature presents large discrepancies in the key performance indicators used to assess the CCS retrofits to EIIIs. Therefore, to achieve as fair comparison as possible, this work considered the cost of CO₂ avoided as the key economic performance indicator and the equivalent energy consumption as the thermodynamic performance indicator. Finally, because this field is of commercial interest, this work also provides an overview of recent developments in calcium looping and compares its performance with other, more mature CCS technologies. It should be noted that, although membrane-based technology has been shown recently promising results in the iron and steel [22–24] and cement [24–27] it was not included in this review due to fewer research studies published across the four EIIIs.

2 Overview of CCS

CCS is a chain of processes from the CO₂ capture to its transport and long-lived storage, with the CO₂ capture being the most expensive and energy-intensive step [29,30]. Across the main industries, three routes have been identified as CO₂ capture strategies: pre-combustion capture, post-combustion and oxy-fuel [14,31,32]. The principle of each CO₂ capture approach is presented in Fig. 1. In pre-combustion capture, CO present in the syngas produced by reforming or partial oxidation is converted to CO₂ and then separated from H₂ at high pressure. The post-combustion capture involves the removal of CO₂ from the flue gases at low CO₂ partial pressure. It is considered an end-of-pipe solution that can be retrofitted to existing systems without major modifications to the plant layout. Yet, due to the heterogeneity of industries, some processes have more than one CO₂ emissions point. As a result, the flue gases from different parts of the process may need to be combined before CO₂ capture [13], reducing the technical viability of the post-combustion capture. The oxy-fuel combustion process assumes that the fuel combustion takes place in pure O₂ rather than air. This leads to a higher CO₂ purity due to the absence of N₂. Another advantage of the absence of N₂ is the reduction of NO_x emissions.

As shown in Fig. 1, the pre- and post-combustion capture require CO₂ separation from the syngas or the flue gas, respectively. Depending on the gas streams characteristics, the following physical-chemical separation processes have been employed in the CO₂ abatement [33]: 1) gas-liquid separation that involves physical or chemical absorption with solvents, called solvent scrub-

bing; 2) gas-solid separation that is based on adsorption by solid adsorbents; 3) membrane-based separation; 4) cryogenic, where the gases are cooled to a very low temperature and the CO₂ is separated.

Among the abovementioned separation processes, amine scrubbing is the most mature technology [25]. Initially proposed by Bottoms [34] to separate the acidic gases present in a gaseous mixture, amine scrubbing has been used as CO₂ separation technology for several decades [29]. This CCS technology use monoethanolamine or methyl diethanolamine as the most common solvents. However, diethanolamine, dominoethoxyethanol, diisopropanolamine, triethanolamine and Ucarsol[®] are also among the chemical solvents considered for chemical solvent scrubbing [11]. In physical scrubbing, the nature of the solvents is more diverse and solvents such as dimethyl ether of polyethylene glycol, methanol, *N*-methyl-2-pyrrolidone and propylene carbonate can be used [11]. Nevertheless, some issues have been identified with this technology in the power industry. These issues include: 1) the solvent degradation at higher temperatures; 2) adverse reactions with some components in the flue gas, such as SO₂ and O₂ [29]; 3) the expensive solvent production [35]; 4) the solvent concentration limited to 30 wt% (monoethanolamine) due to corrosion issues [36]; 5) the efficiency penalty of 9.5% to 12.5% [37]; 6) the generation of high volumes of hazardous wastes, mainly heat-stable salts and carbamate polymers, as a consequence of the thermal and oxidative degradation of the solvent [29,38].

Amine scrubbing has been extensively studied across EIIIs [3,32,39–44], but due to high costs and the abovementioned issues, the use of hot potassium and ammonia-based solvents has also been studied as an alternative to amine-based solvents [11,25]. NH₃ is less corrosive and subject to lower rates of degradation compared to amine-based solvents. It also presents a higher CO₂ absorption capacity and requires less energy once the regeneration is carried out at lower temperatures. However, due to its high volatility and slower kinetics, ammonia scrubbing was deemed not to be a feasible option for the decarbonisation of the EIIIs [45].

Oxy-fuel combustion is another technology at a relatively high technology readiness level [40–42,46] and close to its commercialisation [18]. Since a high CO₂ purity stream is produced during fuel combustion in O₂/CO₂ environment, this technology does not require a CO₂ separation process that constitutes its main advantage. However, to keep the desired purity of CO₂, potential air infiltration into the boiler should be minimised, implying high safety procedures [47]. Moreover, the O₂ production by a cryogenic air separation unit (ASU) increases the costs associated with this technology [48]. Despite the advantage of oxy-fuel compared with amine scrubbing, it is not commercialised due to the issues mentioned and the lack of consistent economic data [13].

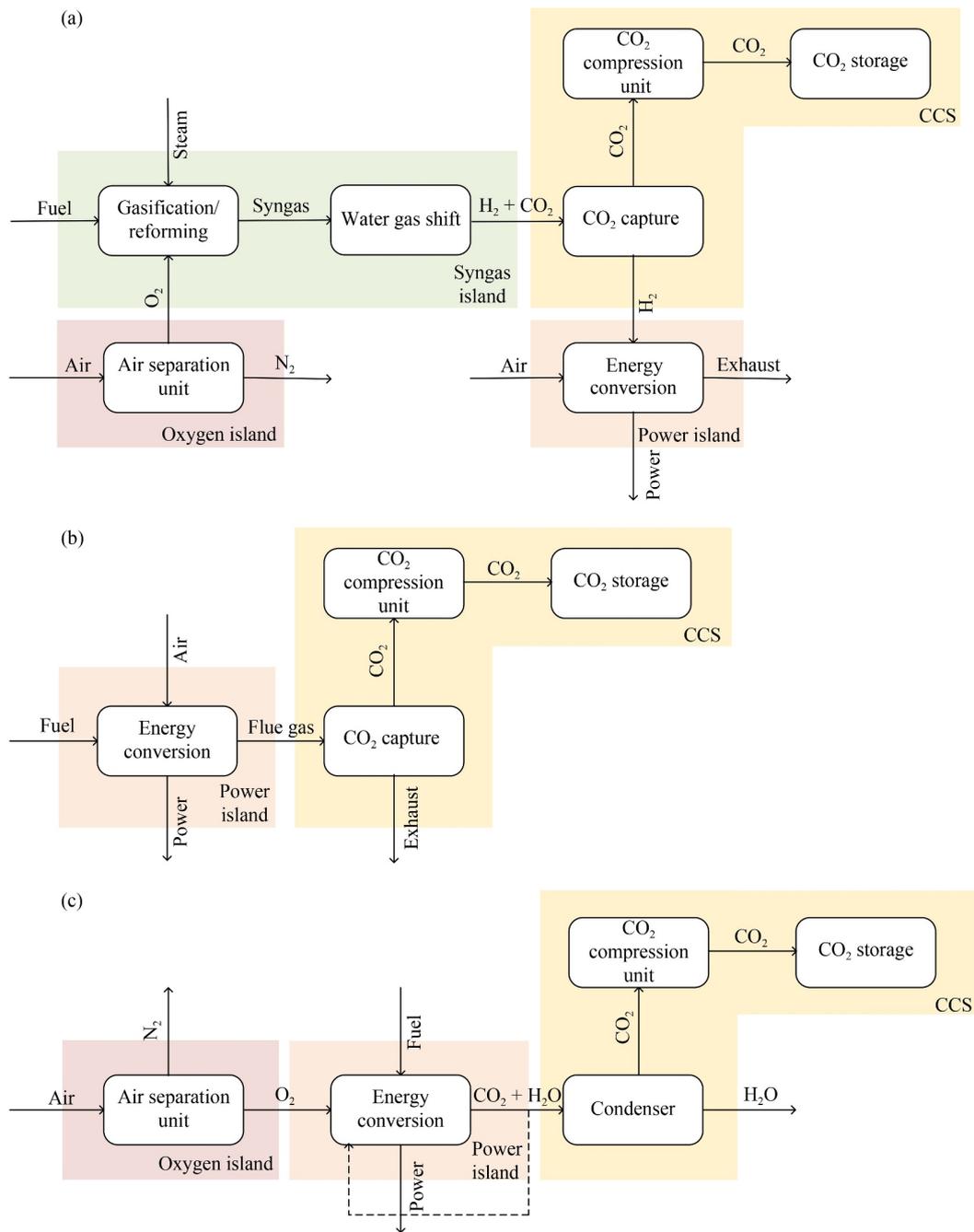


Fig. 1 Block flow diagrams for (a) pre-combustion capture, (b) post-combustion capture, and (c) oxy-fuel combustion CO₂ capture.

To overcome the drawbacks of mature CCS technologies, emerging technologies have been developed. Recently, solid looping cycles, such as chemical looping and calcium looping, have been shown to be a promising technology for the decarbonisation of EIIs [19,21,25,49]. Furthermore, the extensive review performed by Adánez et al. [50] on chemical looping combustion (CLC) of solid fuels has concluded that EIIs, such as the pulp and paper and cement industries, are potential industries to apply CLC. As it was shown by Vilches et al. [51], a circulating fluidised bed gasifier can be coupled to a bubbling bed boiler and operate as a CLC unit. In such a

case, the former assumes the function of a fuel reactor and the latter works as air reactor, while generating heat. Since the energy demand of these two industries is met by using boilers, the implementation of CLC would be suitable with a minor need for retrofitting.

2.1 Solid looping cycles

The most common layout of the solid looping cycles consists of two interconnected fluidised beds. A metal oxide is used as an O₂ carrier (i.e., Cu, Ni) or CO₂ carrier (i.e., CaO, MgO) in chemical or carbonate looping,

respectively. The carrier circulates between the two reactors in alternate cycles of reduction-oxidation or carbonation-calcination, which is the most commonly studied configuration. Nevertheless, recently in the iron and steel industry, Fernández et al. [52] have proposed using packed-bed reactors. In such a configuration, the gas, instead of sorbent, circulates between the reactors. Generally, the oxidation and carbonation reactions are exothermic, whereas the reduction and calcination reactions are endothermic, except in the CuO/Cu cycle [53]. The regenerator reactor can be fed with solid, liquid or gas fuels of renewable (i.e., biomass) or non-renewable (i.e., natural gas) origin.

A combination of heat, electricity, chemicals and fuels can be generated by solid looping processes [54–58]. Depending on the target output, solid looping processes can be classified as the following [55]: 1) CLC or calcium looping if the generation of heat or electricity is the main purpose; 2) chemical looping gasification or calcium looping gasification, with the note that gasification can also be referred to as a reforming process when steam or CO₂ is added to the reactor to enhance their reforming reactions [55]. Energy vectors (i.e., hydrogen) and fuels (i.e., syngas) are the main products in such processes.

It should be noted that the difference between gasification and combustion processes is the extent of carbonaceous fuel oxidation. In the gasification process of solid fuels, carbon is first partially oxidised to CO and then further water-gas shift (WGS) reaction takes place to promote the formation of H₂ and CO₂. In the reforming process of gaseous fuels, light hydrocarbons (i.e., CH₄) are first broken down to syngas comprising a mixture of H₂/CO/CO₂, followed by the WGS reaction. Conversely, the combustion process involves the complete oxidation of the fuel to CO₂ while the H₂ on it is oxidised to water vapour.

2.1.1 Chemical looping

Chemical looping employs a metal oxide as an oxygen carrier that transfers oxygen between the two reactors. Nickel, copper, iron, manganese and cobalt-based oxygen carriers have been studied as oxygen carriers [54,59]. The following characteristics must be presented by them [60]: 1) be reactive in reduction and oxidation steps; 2) should be able to fulfil stoichiometric combustion; 3) be stable at high temperatures, even after a large number of cycles; 4) its structure should not be affected by the friction resultant from the fluidisation; 5) be fluidisable and should avoid the formation of clusters; 6) be eco-friendly and no hazardous to operators; 7) be viable economically. Figure 2 presents a basic scheme of this technology. In the first reactor, called the air reactor, the reduced metal oxide is oxidised and the regenerated oxygen carrier returns to the fuel reactor. This step is represented by Eq. (1):

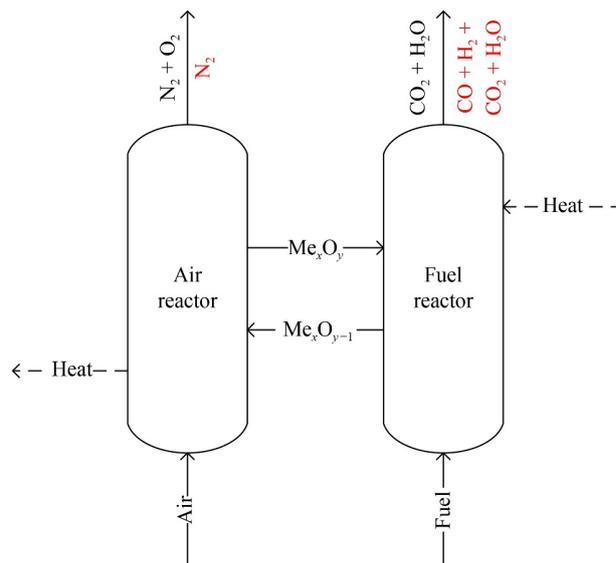
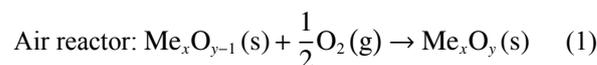
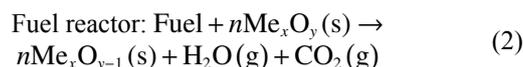


Fig. 2 Simplified scheme of chemical looping process (black text: products from CLC; red text: products from chemical looping gasification).



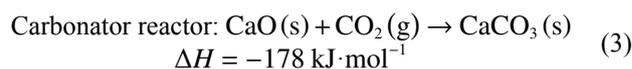
Then, the oxidised oxygen carrier is fed to the fuel reactor, where it is reduced on contact with the fuel to produce a stream of CO₂ and water vapour, according to Eq. (2),



The water vapour is then condensed and a high-purity CO₂ stream is sent for compression, avoiding the need for additional separation processes. The cost of these separation processes can be varied depending on the partial pressure of CO₂ and the technology used [10].

2.1.2 Calcium looping

Calcium looping was initially proposed by Shimizu et al. [61] as a CCS option for the decarbonisation of the power industry. Since CaCO₃ is formed during the carbonation reaction, the calcium looping is also known as carbonate looping [62,63]. The concept of this technology is shown in Fig. 3. The CO₂ from the flue gas or syngas is captured by the sorbent (i.e., CaO), which circulates between the two reactors, leading to the formation of CaCO₃ in the carbonator. The decomposition of CaCO₃ to CaO and CO₂ takes place in the calciner, which requires high-grade heat provided by the oxy-fuel combustion. As shown in Eqs. (3) and (4), this process is based on the reversible reaction between CaO and CO₂:



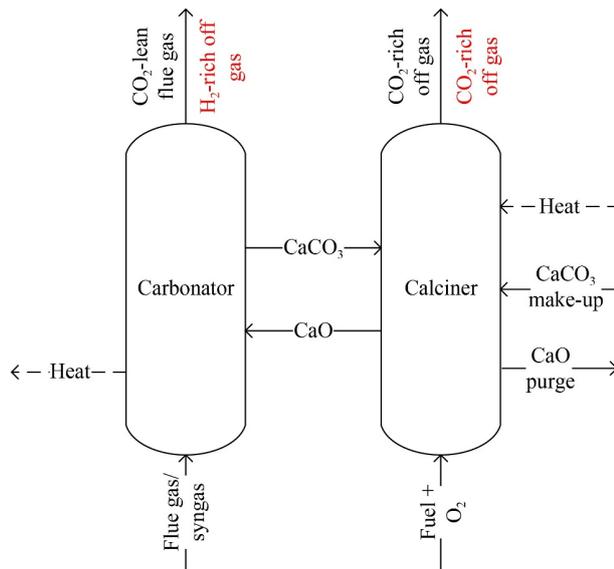
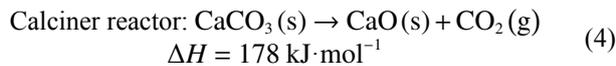


Fig. 3 Simplified scheme of calcium looping technology (black: products from calcium looping; red: products from calcium looping gasification).



In the first reactor, the so-called carbonator, the CO_2 is removed from the flue gas or syngas at 600–750 °C. In the second reactor, the so-called calciner, the decomposition of the formed carbonate takes place at 870–950 °C and a CO_2 -rich gas is produced [64]. Calcium looping, involves oxy-combustion in the calciner to provide heat for sorbent regeneration to ensure the high-purity of the CO_2 stream. This technology also presents the following advantages over mature amine scrubbing [29]: 1) use of fluidised beds, a well-known technology at large scale [65]; 2) the thermal energy released in the carbonator can be recovered to produce steam, which is one of the key benefits of this technology [66]; 3) the low cost and high availability of the natural sorbents, limestone; 4) the sorption capacity of limestone-based sorbents after a large number of cycles is higher than that for many synthetic sorbents [47]; 5) the sorbent is SO_2 selective which contributes to a partial desulphurisation of the stream; 6) the purged CaO can be used as raw material in the cement and iron and steel industries [19].

However, the reactivity decay of the sorbent over the cycles is the main challenge of calcium looping [67,68]. The loss in the adsorption capacity is attributed to the sorbent sintering during the calcination [69,70] that is exacerbated by high temperatures, long cycles, high CO_2 and steam partial pressures and by contaminations [69]. Moreover, due to abrasion and fragmentation, a small fraction of sorbent particles can leave the calciner. Therefore, the fresh sorbent is continuously fed to it [71]. As the CaCO_3 decomposition to CaO and CO_2 at high pressure requires temperatures higher than 900 °C [72], another challenge of this technology is to find a less energy-

intensive process for sorbent regeneration. To overcome this limitation, a new configuration was developed, first proposed by Lyon [73]. He has suggested that the coupling of a solid looping cycle, Fe/FeO , can deliver the heat required in the calcination step. This was provided by the exothermic oxidation of the Fe to FeO with air, which took place in the same fixed bed reactor as the calcination. Although this new loop met the calcination heat requirement, the CO_2 emissions problem was still unsolved. This was because the CO_2 formed during the calcination was diluted by the N_2 present in the air and thus released to the atmosphere. Then Abanades et al. [74] proposed the replacement of Fe/FeO by CuO/Cu , known as Ca-Cu chemical looping. In this case, the heat requirement for the CaCO_3 decomposition was delivered via oxidation of fuel in the reaction with CuO that was produced in another reactor through the oxidation of Cu to CuO with air. Consequently, a CO_2 -rich stream is produced in the integrated calciner and fuel reactor, since there is no dilution by N_2 .

To date, solid looping cycles have been extensively researched in the power industry, and some calcium looping and CLC pilot scale units were already built and assessed: the 1.7 MW_{th} pilot in La Pereda [66], the 0.2 MW_{th} facility in Stuttgart University [75], the 1 MW_{th} unit in Darmstadt [76], the 1.9 MW_{th} pilot in Taiwan of China [77], the 120 kW_{th} CLC unit in Wien University [78], the 100 kW_{th} CLC unit in Chalmers University [79], the 3 MW_{th} CLC unit at ALSTOM labs [80] and more recently, the 3 MW_{th} CLC unit in China (CHEERS project) [81]. However, the effectiveness of this technology does not limit just to the power industry. Regarding the EIIs, calcium looping has been intensively explored in the cement industry by CEMEX in Mexico [10] and in the past few years, the feasibility of its integration in the steelmaking process has also been studied by Fernández et al. [52] and Tian et al. [19]. Regarding the petrochemical and pulp and paper industries, calcium looping has not received much interest and only a few papers were found in the literature.

3 Carbon capture for decarbonisation of EIIs

3.1 Considerations

In this review, the CO_2 capture technologies for decarbonisation of the EIIs were compared with respect to their techno-economic performance. The thermodynamic performance of the entire system was evaluated using an equivalent energy consumption (EE , $\text{MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$), defined in Eq. (5). This is because the considered CCS technologies require energy of different quality: thermal energy of fuel (Q_{LHV}), thermal energy of steam (Q_{th}) and electrical

energy (P_e). To ensure a fair comparison, the electric energy was converted to thermal energy using an electric efficiency of 45.9% [20]. To convert the steam energy to the same basis of calculation, it was assumed that steam was produced in a boiler with an efficiency of 90% [52]. The \dot{m}_{CO_2} is the flowrate of CO_2 captured ($\text{kg}\cdot\text{s}^{-1}$).

$$EE = \frac{\left(Q_{\text{LHV}} \pm \frac{Q_{\text{th}}}{0.90} \pm \frac{P_e}{0.459}\right)_{\text{CCS}} - \left(Q_{\text{LHV}} \pm \frac{Q_{\text{th}}}{0.90} \pm \frac{P_e}{0.459}\right)_{\text{Ref}}}{\dot{m}_{\text{CO}_2}} \quad (5)$$

Depending on the technology and level of excess heat integration of the plant, some terms in Eq. (5) can be zero. The plus and minus signs represent an input or output of the plant, respectively. The equivalent energy consumption accounts for the heat requirement for solvent regeneration in the case of chemical and physical absorption. In the case of vacuum pressure swing adsorption (VPSA) and oxy-fuel combustion, the equivalent energy consumption accounts for the power required by sorbent regeneration and the ASU, respectively. The power requirement for the CO_2 compression unit is also accounted for all technologies, whenever is available. For solid looping cycles, the equivalent energy consumption is defined only as the thermal energy requirement met by the fuel consumption (Q_{LHV}), since these technologies integrate a steam cycle to recover the waste heat to meet their electrical energy requirement.

The economic performance was evaluated using the cost of CO_2 avoided (AC). For the sake of clarity, this metric is defined in Fig. 4. The cost of CO_2 avoided is defined as the cost associated with reducing the CO_2 emissions from the reference plant due to the CCS retrofit, excluding the additional CO_2 that needs to be captured to offset the energy penalty of the CCS retrofit.

All the costs reported in this review were adjusted to the year 2017 by using the Chemical Engineering Plant Cost Index (CEPCI), Eq. (6), and reported in Euro (€). The subscript i refers to the year of reported data. If a different currency is used in the reviewed studies, the costs were updated to the year 2017 and then converted to Euro.

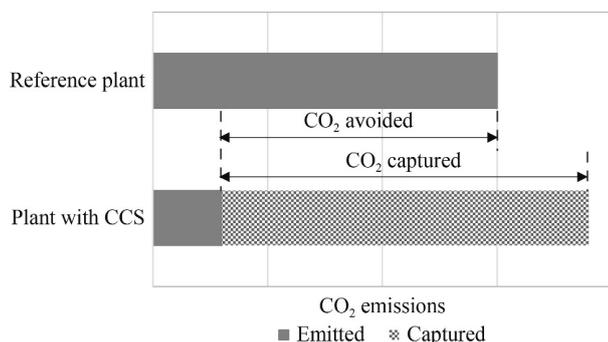


Fig. 4 Difference between CO_2 avoided and CO_2 captured.

$$AC_{2017} = AC_i \frac{CEPCI_{2017}}{CEPCI_i} \quad (6)$$

3.2 Iron and steel industry overview

The iron and steel industry is one of the main contributors to GHG emissions, accounting for 25.1% of the industrial CO_2 emissions in the world in 2018 [6]. Although the steel scrap-based production, which is based on secondary or recycled steelmaking process, generates around 4.5 times less CO_2 [82], the ore-based production is still expected to prevail, at least, in the next decade. It is due to the increasing steel demand and still low recycling rates of steel scrap [83]. Regardless of the steelmaking process, the following steps are present: raw material preparation, iron production and finally the steel production. Blast furnace, smelt reduction or direct reduction are the three main processes in iron production [84]. The ore-based production is carried out mainly through blast furnace-basic oxygen furnace (BF-BOF) or direct reduced iron electric arc furnace [85]. The iron production via BF-BOF is represented in Fig. 5. As can be seen, to meet the energy requirement of the iron and steel production, the off-gases from other operations are combusted in the power plant. This power plant can account for around 50% of the overall CO_2 emissions, while the blast furnace, the coke and sinter plants contribute to 26%, 13% and 10% of the total CO_2 emitted by this steelmaking plant, respectively [86]. According to Wiley et al. [40], the CO_2 concentration in the flue gases can vary from 8 vol% in the gas released in the sinter plant to 27 vol% in the exhaust gas from the coke plant. The flue gas from the blast furnace can contain up to 22 vol% of CO_2 . In 2004, to seek deep decarbonisation of the iron and steel industry, 48 European companies from 15 countries joined efforts and formed a consortium called ULCOS [87]. These partners have identified new iron and steelmaking processes, such as blast furnace with top gas recycling (TGR), smelting reduction (HIsarna), direct reduction (ULCORED) and electrolysis (ULCOWIN) to mitigate CO_2 emissions [82]. In the first process, TGR, O_2 is introduced in the blast furnace instead of air, eliminating N_2 presence and thus, increasing the CO_2 concentration in the flue gas. The second process, HIsarna, was developed to produce a high CO_2 purity stream via ore reduction with coal in pure oxygen rather than air. The ULCORED process relies on the production of direct-reduced iron by using natural gas, coke oven gas, or syngas from coal/biomass gasification as a reducing agent. Similar to the two previous routes, this process also needs to be coupled with CCS, but there is only one CO_2 source point (shaft furnace). The ULCOWIN process is a carbon-free technique where the iron oxides are reduced electrochemically. According to van der Stel et al. [88] and Eurofer: The European Steel Association [87], TGR and HIsarna process combined with CCS present a potential for CO_2 abatement that could reach the 60% and 80%, respec-

tively. In case of ULCOWIN, the potential CO₂ reduction can achieve the 98% when the electricity required to the process is generated from non-fossil fuel sources. Over the years, several CO₂ capture technologies have been assessed.

3.2.1 Chemical absorption (post-combustion amine scrubbing)

As mentioned above, after the power plant, the blast furnace is one of the main contributors to CO₂ emissions in iron and steel plants. For this reason, several researchers have been studying the CO₂ abatement measures for this specific part of the process. Ho et al. [84] have estimated the capture costs associated with CO₂ capture from a conventional blast furnace by amine scrubbing (monoethanolamine). It should be mentioned that the estimated costs only include the cost associated with CO₂ capture, and the transport and storage costs were not included in their estimation. They concluded that the cost of CO₂ avoided was around 59.7 €·t_{CO₂}⁻¹, with an equivalent energy consumption of 3.3 MJ_{th}·kg_{CO₂}⁻¹. However, replacing the conventional blast furnace with the Corex process could reduce the cost of CO₂ avoided to 45.6 €·t_{CO₂}⁻¹. In this process, the smelt reduction

process uses O₂ and coal instead of air and coke, which takes place in two separate reactors. These reactors, reduction shaft and the melter gasifier substitute the sinter and coke plants. This alternative resulted in a decrease in the equivalent energy consumption of around 7% (3.1 MJ_{th}·kg_{CO₂}⁻¹). The lower cost and power consumption was due to higher CO₂ concentration and no requirement for pre-treatment of flue gas.

Ho et al. [89] performed the techno-economic feasibility assessment of the amine scrubbing retrofit that used monoethanolamine as a solvent. This study has compared the CO₂ avoided cost for CO₂ capture from the flue gas of the TGR and Hismelt processes with the largest volume of CO₂. In this case, it was the flue gas of the blast furnace and the smelt reduction vessel, respectively. The costs associated with CO₂ transport and storage were neglected. This study concluded that CO₂ capture from the TGR was the least expensive option, resulting in the cost of CO₂ avoided of 35.1 €·t_{CO₂}⁻¹. Such figure was 34.3% lower than 53.4 €·t_{CO₂}⁻¹ reported for the Hismelt process. Interestingly, the energy requirement for both processes was comparable and reported to be 3.1 and 3.3 MJ_{th}·kg_{CO₂}⁻¹ for the TGR and Hismelt process, respectively.

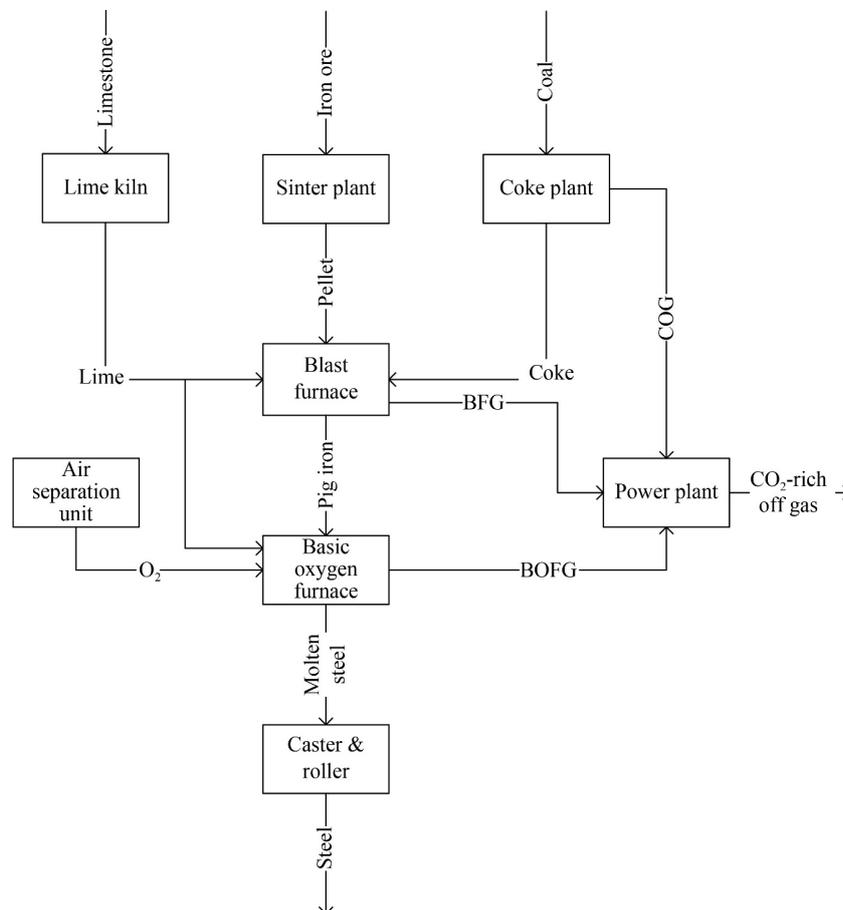


Fig. 5 Block flow diagram of an iron and steel plant with steel production via BF-BOF (BFG, blast furnace gas; BOFG, basic oxygen furnace gas; COG, coke oven gas).

Tsupari et al. [42] have studied the economic feasibility of amine scrubbing retrofit to the iron and steelmaking plant. The CO₂ capture from the largest source of emissions, power plant and blast furnace hot stoves, was evaluated. This study compared three different solvents, including monoethanolamine, advanced amine and low-temperature regeneration amine (“Low T”), as well as five different layouts for heat integration for solvent regeneration. Depending on the level of waste heat integration, the equivalent energy consumption was shown to fall between 0.5 (monoethanolamine) and 4.3 MJ_{th}·kg_{CO₂}⁻¹ (monoethanolamine). In the former case, the heat for solvent regeneration was covered by the waste heat from the steel plant. This, however, resulted in the lowest CO₂ capture capacity. The highest equivalent energy consumption, and subsequently the capture rate, was found for the case where all the steam produced in the iron and steelmaking plant was used for solvent regeneration. However, in such a case, no electricity was generated and the entire electricity requirement of the plant was met by the grid electricity. This study has also found that the electricity price played an important role in the cost of CO₂ avoided. If the price of electricity was fixed at 80 €·MWh⁻¹, the cost of CO₂ avoided was between 81.5–164.1 €·t_{CO₂}⁻¹ (“Low T” solvent) and between 91.3–180.6 €·t_{CO₂}⁻¹ (monoethanolamine). Nevertheless, if the cost of electricity increased to 100 €·MWh⁻¹, the corresponding cost of CO₂ avoided would also increase to between 90.3–186.4 and 92.2–205.8 €·t_{CO₂}⁻¹ for the “Low T” solvent and monoethanolamine, respectively. The “Low T” solvent presented lower costs due to lower heat duty for solvent regeneration, 3.4 against 3.0 MJ_{th}·kg_{CO₂}⁻¹, which translated into a lower energy penalty.

Dreillard et al. [90] have compared the techno-economic performance of three different amines when employed to capture CO₂ from the flue gases of TGR and blast furnace. A conventional amine (monoethanolamine) with two different concentrations (30 and 40 wt%) and a demixing solvent (blend amines) were selected as solvent. The latter involves the change of phase during the process. Their study has shown that the heat requirement for solvent regeneration of demixing solvent was lower by 20%–25% and 6%–9% than the one for monoethanolamine 30 wt% and monoethanolamine 40 wt%, respectively. This translated into an energy consumption in the range between 3.4 (monoethanolamine) and 2.7 MJ_{th}·kg_{CO₂}⁻¹ (demixing solvent) for blast furnace case and between 3.3 (monoethanolamine) and 2.4 MJ_{th}·kg_{CO₂}⁻¹ (demixing solvent) for TGR case. Such a reduction in the energy requirement can be explained by a higher CO₂ partial pressure in the flue gas from TGR. It should be noted that in both cases, there was no heat integration. Yet, it was not possible to estimate the equivalent energy consumption based on the data provided in their study. Similarly, the cost of CO₂ captured was reduced by 20%–28% when

the monoethanolamine 30 wt% was replaced by the demixing solvent. The cost of CO₂ captured decreased from 67.0 to 53.4 €·t_{CO₂}⁻¹ and from 56.6 to 40.9 €·t_{CO₂}⁻¹ to capture the CO₂ from blast furnace and TGR, respectively.

The CO₂ mitigation by amine scrubbing (monoethanolamine) was also assessed by Garðarsdóttir et al. [91]. They have estimated the CO₂ capture costs and energy consumption associated with the CO₂ capture from the power plant’s flue gas. The equivalent energy consumption for the CO₂ capture was 4.8 MJ_{th}·kg_{CO₂}⁻¹, which was higher than reported in previous studies. This can be attributed to the different levels of heat utilisation for solvent regeneration, which also impacts the CO₂ capture cost. Consequently, the cost of CO₂ captured estimated in this study was 51.4 €·t_{CO₂}⁻¹. It needs to be noted that this study reported the cost of CO₂ captured that, in general, are lower than the CO₂ avoided cost. This is because the former also includes the additional CO₂ that needs to be captured to offset the energy penalty of the CCS retrofit (Fig. 4).

Finally, Cormos et al. [92] have performed a techno-economic analysis of the CO₂ capture retrofit to the power plant, blast furnace hot stoves and lime and coke production via the amine scrubbing using methyl diethanolamine as a solvent. Their study estimated the CO₂ avoided cost of 73.5 €·t_{CO₂}⁻¹. This figure is slightly higher than that reported in previous studies. However, as opposed to previous studies by Ho et al. [84,89], this study accounted for the CO₂ transport and storage costs, showing the need for the complete life cycle costing of the CCS chains. Furthermore, the equivalent energy associated with the CO₂ capture was –1.6 MJ_{th}·kg_{CO₂}⁻¹. This means that the net power output of the plant retrofitted with CO₂ capture is higher than the one for the reference plant. This is the case because this study considered implementing a gas turbine that generated additional electricity and the steel plant off-gases were used to meet the heat and power requirements.

3.2.2 Physical absorption (Selexol)

To reduce the energy penalty associated with CCS, the low CO₂ partial pressure flue gas from iron and steel production can be pressurised and the CO converted into CO₂ via WGS reaction, thus increasing the CO₂ concentration. Physical absorption using Selexol as a solvent was the technology evaluated by Ho et al. [84] for such a case. This study has compared the Selexol process retrofit to the blast furnace and the Corex process. They estimated the cost of CO₂ avoided of 52.7 and 28.1 €·t_{CO₂}⁻¹, for blast furnace and Corex process, respectively. Similarly, the equivalent energy consumption for the Corex process was almost half (1.2 MJ_{th}·kg_{CO₂}⁻¹) of that estimated for the blast furnace (2.4 MJ_{th}·kg_{CO₂}⁻¹). This is due to the

higher content of CO in the Corex process stream before the water gas shift reaction (44%) compared to the CO content in the gas from the blast furnace (21%).

3.2.3 VPSA

Based on their previous studies, Ho et al. [89] have studied the implementation of VPSA, using zeolite 13X as adsorbent, as an alternative to reduce the cost of CO₂ capture. The economic feasibility of CO₂ capture from the flue gas, with the largest volume of CO₂, of 4 different processes, including conventional blast furnace, TGR, Hismelt and Corex, were compared. These points are normally selected due to economy of scale and, therefore, are likely to be prioritised for decarbonisation. The CO₂ capture was carried out at 1.5 bar and the adsorbent regeneration took place at 0.05 bar. Their study showed that the cost of CO₂ avoided from the flue gas of the TGR and Corex processes were the lowest among the considered processes, and were estimated to be around 26.7 €·t_{CO₂}⁻¹. The cost of CO₂ avoided from the flue gas of the blast furnace and the Hismelt processes were estimated to be around 31.6 and 35.1 €·t_{CO₂}⁻¹, respectively. The estimated equivalent energy consumption was estimated to be between 1.8 (Corex) and 2.4 MJ_{th}·kg_{CO₂}⁻¹ (Hismelt).

3.2.4 Solid looping cycles

Conversely to the other EII, such as cement, the solid looping cycles were considered for the decarbonisation of the iron and steel industry only recently. Therefore, techno-economic assessments are still very scarce. The integration of calcium looping to the iron and steel plant was proposed for the first time by Tian et al. [93]. In the considered process, the Ca and Fe ions in the iron slag were first separated using acetic acid. Then, the Fe-rich stream was recycled to the blast furnace and the Ca-rich stream, after precipitation and dry-distillation, was fed to the calciner. The spent CaO was recycled to the blast furnace. They concluded that the steel slag-derived sorbent could be employed as a CO₂ sorbent, presenting higher reactivity and lower degradation compared to limestone. Depending on the synthesis process, four of the steel slag-derived samples presented a CO₂ uptake between 0.08 and 0.11 g_{CO₂}·g_{sorbent}⁻¹ while the limestone stabilised for 0.07 g_{CO₂}·g_{sorbent}⁻¹ after 15 cycles. However, the enhanced CO₂ capture performance resulted in a higher cost of sorbent purchase that could be compensated by selling two by-products, high-purity CaO and acetone. Again, depending on the steel slag-derived sorbent synthesis, the final cost of this would result in values between 57.7 and 145.7 €·t⁻¹ compared to 102.0 €·t⁻¹ for limestone.

Based on their previous work, Tian et al. [94] have proposed a combined Ca-Fe looping to capture the CO₂ released from the blast furnace during the iron produc-

tion. The heat required to drive the endothermic reaction in the calciner was provided by the FeO oxidation. Similar to their previous work, the spent sorbent was utilised in the iron process as the CaO could be used in the blast furnace. The developed CaO-based, Fe-functionalised sorbent presented a medium CO₂ sorption capacity, around 0.16 g_{CO₂}·g_{sorbent}⁻¹, but very good cycling stability.

High energy consumption due to temperatures of around 900 °C during the calcination can be avoided by integrating calcium and chemical looping. In that case, the existing iron and steel power plant can be replaced by a steam cycle power plant. However, in order to produce a high-purity CO₂ stream, the ASU is still required by the oxy-fuel combustion, which is an energy-intensive process. To avoid the ASU requirement, Fernández et al. [95] and Martínez et al. [96] have proposed the combined Ca-Cu looping for H₂ production with CO₂ capture. In this process, the H₂ production was carried out by sorption enhanced water gas shift (SEWGS) and the heat required for the sorbent regeneration was met by reducing CuO to Cu. They concluded that this new process could effectively contribute to the CO₂ abatement in the iron and steel industry. The CO₂ direct emissions could be reduced in 30% by implementing the Ca-Cu process into the blast furnace. Furthermore, the part of the energy input could be recovered by the integration of high heat grade for electricity production.

A new Ca-Cu looping configuration was also considered by Fernández et al. [52]. While in the previous work by Fernández et al. [95], the blast furnace off-gas was used for H₂ production, in case of the new configuration, the coke oven gas and basic oxygen furnace gas were also used as reducing gas. This resulted in up to a 40% increase in the H₂-rich stream production that could be used for on-site power generation or to produce sponge iron, thus increasing the steel plant capacity. Unfortunately, the potential of calcium looping coupled with chemical looping cycles (Ca-CLC) was not completely demonstrated. Although Fernández et al. [52] have reported a specific energy consumption of around 1.5 MJ_{th}·kg_{CO₂}⁻¹, only Martínez et al. [96] have compared this technology with other CCS technologies. They found that the energy penalty associated with CO₂ capture was reduced when Ca-Cu looping was employed, the electricity imported from the grid decreased by more than a half, from 185.4 (monoethanolamine) to 69.1 kWh·t_{HRC}⁻¹ (Ca-Cu looping). Unfortunately, no economic analysis was performed to assess the viability of such a process.

Tian et al. [19] were the first authors to present economic data on solid looping integration to the iron and steel industry. They have proposed a new decarbonisation concept that relies on the inherent CO₂ capture capability of the steelmaking process. The CO₂ emissions of the power plant, where the coke oven gas and blast furnace gas are combusted, were captured at the lime kiln. The integration of calcium looping in the lime production (Ca-LP), resulted in the cost of CO₂ avoided of between 12.9

and $16.3 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$, with an equivalent energy consumption of $2.8 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$. Such figures seem to be the best option in the mid and long-term.

Cormos et al. [92] have evaluated the calcium looping potential to capture the CO_2 from the power plant, blast furnace, hot stoves, lime and coke production. Because this work considered the decarbonisation of four points without integration of calcium looping in the iron and steel process, unlike what was considered in the previous work, the operating costs were higher. Their study presented the CO_2 avoided cost of $68.9 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$ and the energy penalty of $1.3 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$. The former figure is 4–5 times higher, while the latter figure is more than 50% lower than the values reported by Tian et al. [19]. This can be attributed to the implementation of a combined gas turbine.

3.2.5 Discussion

The CO_2 capture from the flue gas of the blast furnace, one of the main sources of CO_2 emissions in this industry, has been extensively studied. The comparative techno-economic performance of the CO_2 capture technologies for decarbonisation of this industry is represented in Fig. 6. It should be noted that only the studies that reported both thermodynamic and economic performance data are represented in that figure. Amine scrubbing, the most commonly studied technology, presents the highest mean cost of CO_2 avoided ($104.6 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$) and the highest mean equivalent energy consumption ($2.7 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$). The range for the equivalent energy consumption for this technology was wide, reflecting the different levels of waste heat integration considered. The current literature shows that using alternative solvents, for example, replacing monoethanolamine with methyl diethanolamine, can reduce the heat regeneration duty and, thus, the cost associated with CO_2 capture. However, the major cost reductions reported in the literature result from

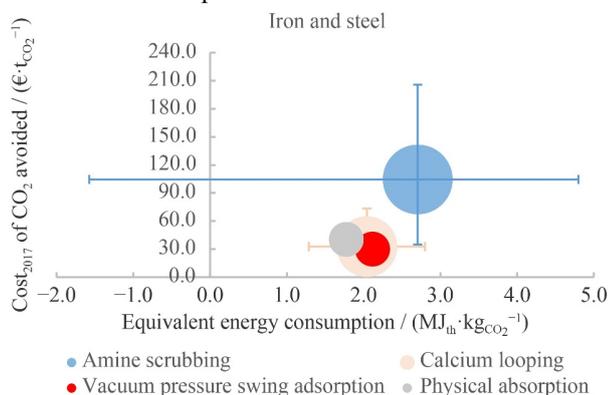


Fig. 6 Techno-economic performance of different CO_2 capture technologies for decarbonisation of iron and steel industry: equivalent energy consumption vs. mean CO_2 avoided cost (Error bars represent the range of figures found in the literature. The bubbles without error bars have only one source. The area of each bubble is proportional to the number of studies reviewed).

consideration of alternative technologies, such as the VPSA or emerging calcium looping. The retrofits of these technologies to the iron and steel plants resulted in the cost of CO_2 avoided of as low as 30.0 and $32.7 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$, respectively. However, the techno-economic assessments of using alternative CO_2 capture technologies for decarbonisation of the iron and steel industry are still very limited. It needs to be emphasised that even for amine scrubbing, the current literature presents a large discrepancy in the results of the techno-economic assessments, with the cost of CO_2 avoided reported to vary between 35.1 and $205.8 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$. This can be attributed to the selection of system boundaries, different assumptions and methods used in the economic assessment, as confirmed by Tsupari et al. [42]. Furthermore, the techno-economic performance indicators also depend on the volume and composition of the flue gas to be treated. Although it is difficult to choose the best technology and, to date, the studies are very limited, calcium looping appears to be a promising option for the decarbonisation of the iron and steel industry. The concept Ca-LP, proposed by Tian et al. [19], showed a CO_2 avoided cost in the range as low as 12.9 – $16.3 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$ due to the heat waste recovery and the reduction in the material cost once the limestone used as the sorbent is fully integrated into the steelmaking process. Regardless of the limited number of assessments presented in the current literature, Fernández et al. [52] showed that combined Ca-CLC is a promising technology for decarbonisation of the iron and steel industry, mostly because of its low specific energy consumption of around $1.5 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$.

Although not discussed in this work, SEWGS technology was shown to be also a promising CO_2 capture technology in the iron and steel industry [97,98]. Manzolini et al. [98] have shown that SEWGS can be employed in an iron and steel plant to capture the CO_2 from the off-gases and produce H_2 . The H_2 is then burnt in the power plant to meet the electricity requirement of the iron and steel plant. They have compared the SEWGS performance with that of a conventional amine scrubbing and found that the former presents a lower cost of CO_2 avoided (33 against $38 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$ for amine scrubbing) and lower specific energy consumption for CO_2 avoided (1.9 against $2.5 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$ for amine scrubbing).

3.3 Cement industry

The cement industry accounted for around 26.4% of the total global industry emissions in 2018 [6]. 40% of these emissions are released during the heat generation for the kiln and the remaining 60% results from the calcination process [29,99,100], in which limestone is converted into CO_2 and CaO at $> 900 \text{ °C}$. The CO_2 content in the exhaust stream after the raw mill, depending on the level of air leaking in the system, can reach up to 22 vol% [25]. Figure 7 shows the simplified process of a cement plant. Since limestone is the cheapest raw material available for

cement production, the abatement of these 60% emissions can only be achieved with a decrease in production [13,100]. Although cement production has decreased in China, which is the largest cement producer globally (2.3 billions tonnes per year), the Indian and some African countries are emerging markets [101]. Therefore, the mitigation of CO₂ emissions from cement production can only be achieved by CCS. Because the flue gas produced in this industry include a high content of CO₂, the application of CCS appears to be a feasible decarbonisation option [5,8].

3.3.1 Chemical absorption (post-combustion amine scrubbing)

Barker et al. [99] were one of the first to study the feasibility of amine scrubbing (monoethanolamine) integration in the cement industry. They found that the cost of CO₂ avoided (116.8 €·t_{CO₂}⁻¹) for this process was higher than that for coal-fired power plants (42.4 €·t_{CO₂}⁻¹). This can be attributed to the lower volume of CO₂ emissions produced by the cement plant, the requirement of a unit for SO_x and NO_x removal, being normally part of existing power plants, and the need for installing a steam generator. The major cost of amine scrubbing is associated with the steam supply for the stripper. It was found that the CO₂ capture translated into an equivalent energy consumption of 5.2 MJ_{th}·kg_{CO₂}⁻¹, for a CO₂ capture rate of 77%, where the extra energy demand was met by coal-fired CHP. However, the study by Barker et al. [99] showed that the replacement of monoethanolamine by other amine solvents with lower heat duty of regeneration and the location of cement plant in an industry cluster with CO₂ capture, enhancing the CO₂ transport scale, could reduce the cost of CO₂ avoided up to 50% (in case of the latter).

The CO₂ avoided cost associated with amine scrubbing (monoethanolamine) implementation in a cement plant was also estimated by Ho et al. [84] and Atsonios et al. [102]. Their work showed lower costs of CO₂ avoided (59.7 and 72.4 €·t_{CO₂}⁻¹, respectively) than that reported by Barker et al. [99]. However, their work did not account

for the transport and storage costs. Ho et al. [84] have estimated an energy penalty of 1.5 MJ_{el}·kg_{CO₂}⁻¹, which corresponds to an equivalent energy consumption of 3.3 MJ_{th}·kg_{CO₂}⁻¹. They concluded that this figure could be reduced by considering solvents with a lower energy requirement, such as methyl diethanolamine, or utilisation of the excess heat available in the cement plant, which was not considered in this work. A higher value was obtained by Atsonios et al. [102], as the estimated equivalent energy consumption was 4.4 MJ_{th}·kg_{CO₂}⁻¹. While Ho et al. [84] assumed that the steam and power required for the carbon capture was delivered by an external source (CHP plant), Atsonios et al. [102] considered an on-site coal-fired boiler to meet the heat requirement for solvent regeneration.

Gomez et al. [103] have compared the economic performance of a cement plant using amine scrubbing as CO₂ capture. Two kinds of amines, the conventional monoethanolamine and a phase-change amine (demixing solvent) were considered as solvent. They have found that the cost of CO₂ capture was reduced by around 50% with the phase-change amine (43.9 €·t_{CO₂}⁻¹) when compared with the conventional monoethanolamine (89.1 €·t_{CO₂}⁻¹). Unfortunately, there was not enough data available to estimate the equivalent energy consumption. However, the authors have found that the heat requirement for solvent regeneration, met by steam, was lower for the phase-change amine.

Zhou et al. [104] have done a comparative study between amine scrubbing (monoethanolamine) with CHP and amine scrubbing with imported steam and electricity. Assuming an 85% CO₂ capture rate, they concluded the cost of CO₂ avoided was comparable in both cases. The second option was marginally less expensive (68.3–113.1 against 78.5–128.4 €·t_{CO₂}⁻¹), mainly due to lower capital costs and the higher CO₂ avoided. The latter was due to the lower electricity emission factor assumed for the electricity imported, since the indirect emissions associated with the electricity and steam required by the CO₂ capture plant were also accounted on the estimation of CO₂ avoided. An equivalent energy requirement of 8.1 and 5.3 MJ_{th}·kg_{CO₂}⁻¹ was found for the first and second

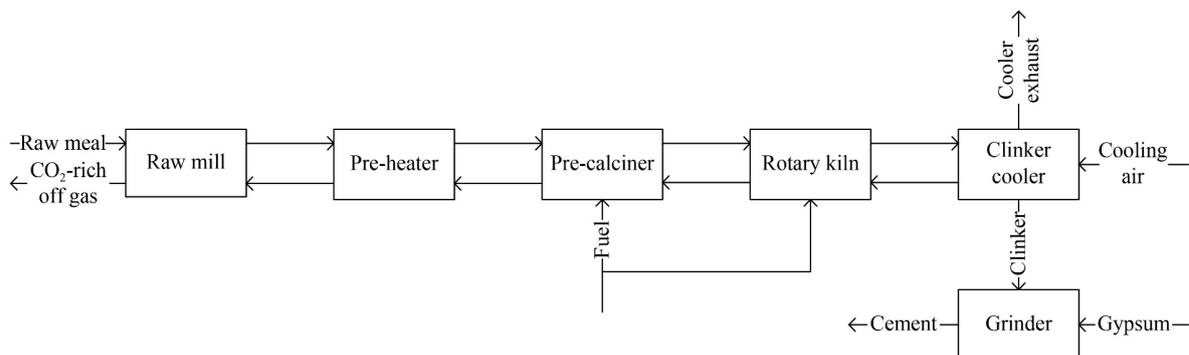


Fig. 7 Block flow diagram of a cement plant.

options, respectively. These figures are higher than the values reported above with similar configurations. This can be explained by different assumptions made in these studies, such as electric efficiency and specific heat requirement for solvent regeneration. The lower energy consumption for the configuration with energy import compared to that with on-site generation is in agreement with the previous results. Since the heat and power production on-site results in a rise of CO₂ produced and then captured, this translates into a higher energy requirement.

A conventional amine, monoethanolamine, was the solvent selected by Gardarsdottir et al. [26] to capture 90% of the CO₂ present in the flue gas stream from the cement kiln. This required an equivalent energy consumption of 5.2 MJ_{th}·kg_{CO₂}⁻¹, the same figure obtained by Barker et al. [99]. The former considered that 96% of the steam required for solvent generation was delivered by a natural gas-fired boiler and the remaining by from the waste heat. Regarding the economic analysis, a comparable figure with the previous works was obtained, 78.5 €·t_{CO₂}⁻¹.

The techno-economic analysis of retrofit amine scrubbing (monoethanolamine) to cement industry was also assessed by Markewitz et al. [105]. Four different cases were compared. In two of them, the plant was an electricity and steam importer, but with different levels of flue gas air leak, in the third case, the steam was provided by an on-site coal boiler, and in the fourth, a coal-fired CHP plant was implemented. They found that the heat requirement demand varied between 3.5–3.8 MJ_{th}·kg_{CO₂}⁻¹, for cases 2, 3 and 4, respectively. Similar to the heat demand, the equivalent energy consumption did not differ too much for the different cases (4.8–5.2 MJ_{th}·kg_{CO₂}⁻¹). As shown by the previous work of Zhou et al. [104], cases 1 and 2 (with energy import) presented the lowest equivalent energy consumption, 4.8 and 4.9 MJ_{th}·kg_{CO₂}⁻¹. Regarding the economic analysis, they concluded that the second case is the cheapest (80.8 €·t_{CO₂}⁻¹), while in cases 1, 3 and 4, the cost of CO₂ avoided were 85.6, 120.5 and 106.9 €·t_{CO₂}⁻¹, respectively.

Cormos et al. [92] performed a techno-economic analysis for CO₂ capture by amine scrubbing (methyl diethanolamine) with CHP to meet the additional energy requirement. The estimated equivalent energy consumption was around 4.8 MJ_{th}·kg_{CO₂}⁻¹. This figure is lower than the previous studies [26,99,105] where on-site CHP was considered as the source to meet the additional energy demand but still comparable. The cost of CO₂ avoided was estimated to be around 86.9 €·t_{CO₂}⁻¹.

3.3.2 Oxy-fuel combustion

Unlike post-combustion amine scrubbing, oxy-fuel combustion technology requires some modification in the cement production process. Because the fuel combustion

with air is replaced by a mixture of O₂ with recycled CO₂, an ASU is required. Furthermore, the retrofit of oxy-fuel combustion to cement process alters the gas temperature, the heat exchange between gas and reactor, and the bed material that affect the quality of clinker [106,107]. Therefore, the pre-calciner and/or rotary kiln, depending on type of oxy-fuel combustion (partial or full), need to be adapted or redesigned and a second pre-heater is required in parallel with the first pre-heater, in case of partial oxy-fuel combustion [107]. Barker et al. [99] evaluated the cost associated with oxy-fuel combustion retrofitted to the pre-calciner in the cement industry. They concluded that to achieve a capture rate of 52%, the cost of CO₂ avoided (43.7 €·t_{CO₂}⁻¹) was almost 1/3 of post-combustion amine scrubbing. Similarly to the cost, the equivalent energy consumption (1.8 MJ_{th}·kg_{CO₂}⁻¹) was less than 1/3 of the energy requirement of monoethanolamine. However, the latter could achieve a higher CO₂ capture rate (77%).

Rodríguez et al. [46] also studied the feasibility of pre-calciner replacement by an oxy-fuel circulating fluidised bed calciner. They estimated a cost of CO₂ avoided of 13.8 €·t_{CO₂}⁻¹ with extra electricity consumption, compared with the reference plant, in the range 0.5–0.7 MJ_{el}·kg_{CO₂}⁻¹. Nonetheless, this additional requirement due to CO₂ capture was covered by the integration of the Rankine cycle for the recovery of the heat waste. The estimated equivalent energy consumption was 1.2 MJ_{th}·kg_{CO₂}⁻¹ for a CO₂ capture rate of 89%. It should be mentioned that although the cost of CO₂ avoided was around half of the figure estimated by Barker et al. [99], it did not take into account the costs associated with CO₂ transport and storage.

The retrofitting of oxy-fuel combustion to pre-calciner was also assessed by Zhou et al. [104]. Their study reported values between 38.7 and 62.2 €·t_{CO₂}⁻¹, which are comparable with those estimated by Barker et al. [99]. Similar to this study, the authors also concluded this cost was around half of the cost for post-combustion amine scrubbing and the equivalent energy consumption (2.0 MJ_{th}·kg_{CO₂}⁻¹) was less than half of the figure obtained for monoethanolamine, in the case of electricity and steam import. However, this performance was obtained with a lower capture rate (62% against 85%) because only the CO₂ from the calciner was captured and thus, the CO₂ from the kiln was released to the atmosphere.

Gardarsdottir et al. [26] proposed the integration of oxy-fuel combustion in cement plant, but in that case, the CO₂ released during the fuel burnt in the kiln was also captured which means the process conditions were changed. They have estimated a CO₂ avoided cost of around 41.5 €·t_{CO₂}⁻¹, almost half of post amine scrubbing cost. This figure was also confirmed by previous studies. It should be emphasised that in that case, the CO₂ emissions from the kiln were also captured. As proposed by

Rodríguez et al. [46], the extra electricity requirement ($0.7 \text{ MJ}_{\text{el}} \cdot \text{kg}_{\text{CO}_2}^{-1}$) was covered by the power generated by a Rankine cycle. The equivalent energy consumption was $1.7 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$, which was in agreement with the values obtained in the previous studies.

3.3.3 Solid looping cycles

For the integration of the solid looping cycles in the cement plants, two configurations have been studied over the years. In the tail-end process, the CO_2 is captured from the flue gases via the post-combustion route. In the integrated process, the calcium looping purge stream, which comprises mainly CaO, is used as a feed stream in the cement plant, the carbonator is integrated with the pre-heater of the clinker production process and the oxy-fuel calciner replaces the pre-calciner of a conventional cement plant. As calcium looping has been extensively explored in the power industry, which along cement industry is a major contributor for CO_2 emissions, Romeo et al. [108] proposed the integration of power and cement plants with a common calcium looping process. This new layout benefitted from the waste energy from the carbonation reaction and clinker cooling, which was used to produce extra power in the steam cycle, and the CaO from the calcium looping was integrated with the cement process. The equivalent energy consumption was estimated to be around $0.7 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$, for a CO_2 capture rate of 94%. The cost of CO_2 avoided associated with this symbiosis was $12.0 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$, which is lower than for a standalone cement or power plant with CO_2 capture. It should be noted that this figure neglected the CO_2 transport and storage costs.

Rodríguez et al. [46] have proposed the substitution of cement pre-calciner by calcium looping process, this was an oxy-fuel calciner connected with a carbonator. In this case, part of the CaO formed in the calciner was directed to the carbonator, where the CO_2 , from flue gas generated in the kiln, was captured. The remaining CaO stream was fed to the kiln to produce the clinker as in the conventional process. Although the integration of calcium looping to cement process showed an electrical penalty comprised between 0.6 and $0.7 \text{ MJ}_{\text{el}} \cdot \text{kg}_{\text{CO}_2}^{-1}$, mainly due to ASU and CO_2 compression, this was covered by the waste heat recovery. This translated into an equivalent energy consumption of $1.3 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$ with 99% of the CO_2 emissions captured. The CO_2 avoided cost was as low as $22.3 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$. However, this cost would be higher if the costs associated with CO_2 transport and storage had been included.

The techno-economic feasibility of retrofitting calcium looping to a cement plant was also evaluated by Atsonios et al. [102]. In this study, the authors took into consideration the technical specifications of the cement process. They showed that fuel composition, S content, plays an important role in the calcium looping

performance and CaO purge quality. They found that coal with lower S content was the fuel that minimised the fresh limestone make-up. The specific heat requirement for sorbent regeneration was $4.6 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$ which is slightly higher than the one for monoethanolamine regeneration ($4.3 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$) in the same work. However, this heat was then recovered in the steam cycle to produce electricity, sold to the grid. For this case, they estimated a cost of CO_2 avoided of $70.1 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$, which is higher than previous works as well as the equivalent energy consumption ($3.0 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$). Like the previous ones, this figure did not include CO_2 transport and storage costs.

Since calcium looping technology has an energy penalty due to ASU, Diego et al. [109] proposed a new concept that did not require one. In order to minimise the calciner heat requirements, a double calcium looping was implanted. In the first cycle, the flue gases from the cement plant and air-fired combustor, were fed to the carbonator, where the CO_2 was captured, and then the CaCO_3 was preheated before entering the calciner. Here, the heat requirements were met by a stream of hot CaO, previously overheated in the air-fired combustor, constituting the second looping cycle. In that case, the CaO stream was a heat carrier. The estimated equivalent energy consumption, for a capture rate of 94%, was $1.6 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$. However, this figure could be reduced to $0.9 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$ if the capture efficiency was reduced to 58%. The cost of CO_2 avoided estimated for both cases was 39.1 and $25.2 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$, respectively. As these figures are comparable to those reported for a single calcium looping scheme, it is difficult to justify the superiority of the new scheme.

As mentioned before, two approaches can be adapted to retrofit calcium looping into a cement plant, integrated and tail-end. De Lena et al. [20] compared three different levels of integration, 100%, 50% and 20%. It should be mentioned that when there is no full integration, it is considered a tail-end process. The technical analysis showed that the calcium looping 100% integrated had the lowest extra electricity consumption of 0.8 against $0.9 \text{ MJ}_{\text{el}} \cdot \text{kg}_{\text{CO}_2}^{-1}$ for only 20% of integration, though, the latter was the only option that resulted in an electricity exporter. The equivalent energy consumption ranged between 2.5 (full integration) and $2.9 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$ (20% of integration). They estimated a cost of CO_2 avoided in the range 51.0 – $57.4 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$ for partial and full integration, respectively, although the latter with a CO_2 capture rate around 3.2 points percent higher than the case with 20% of integration.

Cormos et al. [92] have also studied the feasibility of calcium looping as a tail-end technology for the retrofit of the cement plant. As proposed by the previous authors, the extra electricity demand to drive the calcium looping plant was generated by a steam cycle that recovered the

high-grade heat available in the process. This study reported a similar cost ($57.8 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$) to that estimated by De Lena et al. [20], though with a higher equivalent energy consumption ($3.9 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$) and a lower CO_2 capture rate (90%). It is important to note that the estimated cost for calcium looping was 33% lower than the figure obtained in the same study for amine scrubbing.

3.3.4 Discussion

Calcium looping has been extensively studied in the cement industry as an end-pipe technology or fully integrated into the process. As can be seen from the techno-economic comparison between the CO_2 capture technologies (Fig. 8), the cheapest technology is the oxy-fuel combustion ($40.0 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$) followed by calcium looping ($42.9 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$). Amine scrubbing presents the highest mean cost ($92.0 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$) as well as the highest equivalent energy consumption $5.1 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$, which is mainly due to the high heat requirement for amine regeneration. Unlike the iron and steel industry, where the heat requirement for solvent regeneration or at least part of it can be met by the integration of waste heat, in the cement industry, this needs to be delivered by an external source such, as a boiler or a CHP plant implemented on-site or be off-site but delivering the required energy. The results showed that the implementation of a boiler or CHP on-site results in higher equivalent energy consumption since the CO_2 emissions generated by them are also captured in the CO_2 capture plant. Although calcium looping and oxy-fuel combustion also result in an energy penalty, the additional fuel demand in the calciner for calcium looping and the electricity demand of the ASU for both technologies, a part of high-heat grade can be recovered with a Rankine cycle. Although oxy-fuel combustion resulted in the lowest equivalent energy consumption (1.7 versus $2.2 \text{ MJ}_{\text{th}} \cdot \text{kg}_{\text{CO}_2}^{-1}$ for calcium looping), the CO_2 capture rate is, in general, lower than the values reported for calcium looping technology. In summary, calcium looping is a feasible technology for this industry and an alternative to overcome the high efficiency penalty presented by amine scrubbing.

3.4 Petroleum refining industry

Accounting for 14.3% of the industrial CO_2 emissions in the world in 2018 [6], petroleum refineries are the largest industrial worldwide energy consumer [113]. These industries require 15 vol% of the total primary demand for oil and 9 vol% of gas consumed globally. Johansson et al. [110] categorised the EU refineries by size, considering the simple refinery designs with no conversion units through to the complex refinery designs that consider hydrocracking and catalytic cracking processing units. The latter can also accommodate an integrated gasification combined cycle for the conversion of

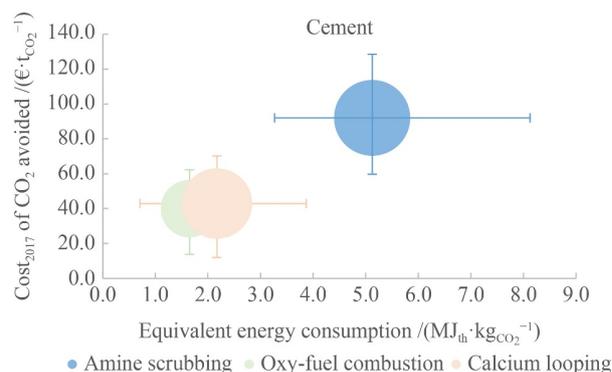


Fig. 8 Techno-economic performance of different CO_2 capture technologies for decarbonisation of cement industry: equivalent energy consumption vs mean CO_2 avoided cost (Error bars represent the range of figures found in the literature. The area of the bubble is proportional to the number of works reviewed).

solids and heavy fuels into heat and power along with lighter products. Figure 9 represents a simple diagram of a refinery with conversion. The main challenge in the petroleum industry is the fact that the CO_2 emissions come from a range of diverse sources, leading to variable CO_2 content in the exhaust gases. According to van Straelen et al. [111], between 20% and 60% of the CO_2 emissions are released by furnaces and boilers. The remainder part comes from different sources: the utilities (electricity and steam) produced to feed the refinery (can be between 20% and 50% of the total emissions), the fluid catalytic cracker (20%–35% of the total emissions), and from other sources during H_2 manufacturing (5%–20% of the total emissions). The type of fuel burnt also plays an important role in the flue gas composition [112]. In general, the CO_2 concentration varies from 4 vol% in the CHP gas turbine, and can reach 20–99 vol% in the gas stream from the pressure swing adsorption (PSA) unit used in the H_2 purification [111]. Then, a combination of multiple flue gases, to be treated in a single CCS unit, may be necessary which is not simple to accommodate in the plant and thus has not been tested yet [13]. The demand for chemical products, such as high-value chemicals, ammonia and methanol, has been increasing and this growth is expected to continue [113]. As these heavily rely on fossil fuels, CCS is a viable route to decarbonise the petroleum refining industry.

3.4.1 Chemical absorption (amine scrubbing)

Amine scrubbing has been the most studied CCS technology in the petroleum refining industry. The costs associated with the retrofit of the post-combustion amine scrubbing (monoethanolamine) into a complex refinery was assessed by van Straelen et al. [111]. They have studied the cost of CO_2 avoided for different CO_2 emissions point sources, with a diverse volume of gas to be treated and CO_2 concentration. For a capture rate between 85% and 90% of a stream with 8 to 9 vol% of CO_2 , the cost of CO_2 avoided was in the range 97.2 – $129.6 \text{ €} \cdot \text{t}_{\text{CO}_2}^{-1}$,

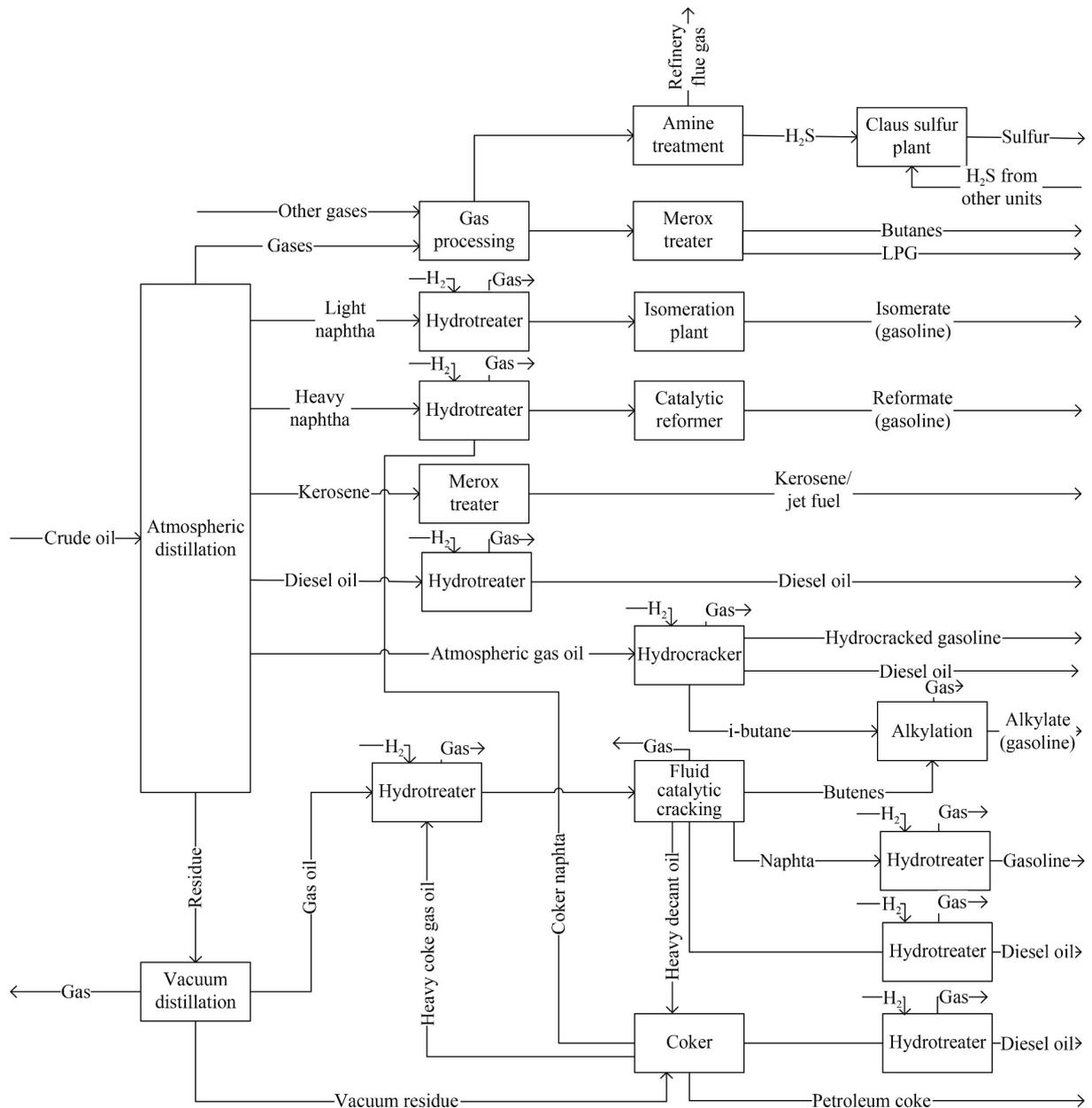


Fig. 9 Simplified diagram of a conversion refinery plant.

although these costs did not include the cost of CO₂ transport and storage. These figures correspond to a combined stack, corresponding to 40% of the total CO₂ emissions of the refinery. Due to the scale of flue gases streams to be treated, the CO₂ capture plant would require the implementation of a dedicated utility plant, with a boiler and a Rankine cycle steam cycle being the most feasible option. In that case, the equivalent energy consumption would be 3.8 MJ_{th} · kg_{CO₂}⁻¹. They concluded that CO₂ capture costs depend strongly on the volume gas to be treated and its CO₂ concentration. If the annual CO₂ captured (8 vol% CO₂ content in the flue gas) dropped from 2000 to 500 kt the cost of CO₂ avoided increased by around 20%. The reduction in the level of CO₂ in the flue gas, from 12 to 8 vol% translated into a rise up to 25% on

the cost of CO₂ avoided.

Ho et al. [84] also studied the economic feasibility of CO₂ capture from a combined stack with 9 vol% of CO₂ concentration. They concluded that the implementation of a CO₂ capture unit, post-combustion amine scrubbing (monoethanolamine), introduced an electrical power penalty of 1.6 MJ_{el} · kg_{CO₂}⁻¹, which translated into an equivalent energy consumption of 3.5 MJ_{th} · kg_{CO₂}⁻¹. For a 90% of capture rate, the cost of CO₂ avoided was equal to 76.3 € · t_{CO₂}⁻¹, a lower figure than the one reported by van Straelen et al. [111]. Since the CO₂ concentration and volume captured (1000 kt per year) were similar and in both cases, the cost associated with the CO₂ transport and storage was excluded, this difference maybe is due to the different route assumed for the source of energy to meet

the energy requirement by the CO₂ capture plant. While these authors assumed that the energy was delivered by an external source, the previous study considered the implementation of a CHP plant.

Berghout et al. [114] compared the retrofits of the pre- and post-combustion amine scrubbing in five different plants (two refineries, two petrochemical plants and a steam reforming hydrogen plant) in a long- (2040–2050) and short- (2020–2025) term analysis. Therefore, the latter assumes that commercially ready CO₂ capture technologies are employed and the former considers new technologies still in development. While monoethanolamine was selected as the solvent for the post-combustion retrofits, a mixture of methyl diethanolamine and 3 wt% of piperazine was chosen in pre-combustion. This was justified by higher absorption capacity, chemical stability and lower regeneration heat requirement. In the post-combustion, the CO₂ present in the flue gases (4–16 vol% CO₂ concentration) from furnaces, boilers and the catalytic cracker, depending on the plant type, was captured. In the pre-combustion capture, the CO₂ was captured from a high-pressure gas leaving the steam methane reformer, followed by the WGS reactor. The main techno-economic results are shown in Table 2. As can be seen for refineries, the equivalent energy consumption was the lowest for pre-combustion (between 1.1 and 1.2 MJ_{th}·kg_{CO₂}⁻¹) retrofits than for post-combustion for either short (3.4–4.0 MJ_{th}·kg_{CO₂}⁻¹) and long term (2.6–3.3 MJ_{th}·kg_{CO₂}⁻¹). This can be attributed to the selection of a solvent with lower regeneration duty in the pre-combustion and the waste heat from H₂ production was enough to meet the energy requirement for the solvent regeneration, though this was achieved with high fuel consumption and high CO₂ emissions. Regarding the chemical plant, the equivalent energy requirement was again lower for pre-combustion (1.1 MJ_{th}·kg_{CO₂}⁻¹) than post-combustion at short- (4.0–4.7 MJ_{th}·kg_{CO₂}⁻¹) and long- terms (2.1–3.3 MJ_{th}·kg_{CO₂}⁻¹) for both plants. Even though the cost is dependent on CO₂

concentration and volume to be treated, in general, pre-combustion presented higher CO₂ avoided costs (89.6–172.1 €·t_{CO₂}⁻¹) than post-combustion (71.1–120.5 €·t_{CO₂}⁻¹) due to higher fuel and capital costs. Regarding the H₂ plant, the post-combustion energy requirement in the short- and long- term (5.8 and 4.8 MJ_{th}·kg_{CO₂}⁻¹) was higher than the figure reported for pre-combustion (3.1 MJ_{th}·kg_{CO₂}⁻¹) and higher costs of CO₂ avoided, too. This can be explained by the lower heat solvent regeneration used in the pre-combustion, which had an impact on energy requirement and so, on the energy expenses. Furthermore, in the pre-combustion, the CO₂ was captured from a high-pressure gas rather than a flue gas at atmospheric pressure. However, these figures were obtained at the expense of a lower CO₂ capture rate (56%, pre-combustion, against 80% and 89%, post-combustion). It should be noted that the specific CO₂ emissions factors for the additional fuel and electricity imported (in this case at short and long term) were accounted in calculation of the CO₂ emissions captured.

Post-combustion amine scrubbing (a mixture of methyl diethanolamine and piperazine) was also considered by Fernández-Dacosta et al. [115] to assess the techno-economic feasibility of CO₂ capture (with an efficiency of 95%) from an H₂ plant in a refinery. They evaluated two scenarios, CCS and CCUS. In the latter, 10% of the CO₂ captured was used for polyol production. Unlike previous studies that the CO₂ avoided cost was estimated, these authors determined the break-even CO₂ cost, 49.2 €·t_{CO₂}⁻¹, which accounts for the CO₂ captured instead of avoided. This figure means that implementation of CCS would be economically more attractive than without CO₂ capture. Regarding CCUS, they concluded that could be a feasible option since part of CO₂ is replacing an expensive feedstock. The equivalent energy consumption was around 4.1 MJ_{th}·kg_{CO₂}⁻¹ for both cases, which is comparable with the values reported by other authors.

Table 2 CO₂ capture rate, equivalent energy consumption and cost of CO₂ avoided for post- and pre-combustion

Item		Post-combustion		Pre-combustion
		Short-term	Long-term	Short-term
Refineries	CO ₂ capture rate/%	86–85	89–79	82–72
	Equivalent energy consumption/(MJ _{th} ·kg _{CO₂} ⁻¹)	3.4–4.0	2.6–3.3	1.1–1.2
	Cost of CO ₂ avoided/(€·t _{CO₂} ⁻¹)	78.3–82.4	71.1	89.6–92.7
Chemical plants	CO ₂ capture rate/%	80–84	80–88	100
	Equivalent energy consumption/(MJ _{th} ·kg _{CO₂} ⁻¹)	4.7–4.0	3.3–2.1	1.1
	Cost of CO ₂ avoided/(€·t _{CO₂} ⁻¹)	94.8–120.5	83.5–98.9	117.5–172.1
Steam reforming H ₂ plant	CO ₂ capture rate/%	80	89	56
	Equivalent energy consumption/(MJ _{th} ·kg _{CO₂} ⁻¹)	5.8	4.8	3.1
	Cost of CO ₂ avoided/(€·t _{CO₂} ⁻¹)	117.5	101.0	62.8

3.4.2 Oxy-fuel combustion

As mentioned before, Berghout et al. [114] have compared different CO₂ capture technologies in five plants in a long- (2040–2050) and short- (2020–2025) term analysis. In the case of oxy-fuel combustion, this study assessed the performance of CO₂ capture retrofit to two refineries and two chemical plants. The oxy-fuel combustion was applied to the boilers, furnaces and catalytic cracker. The techno-economic performance in a short and long term analysis is present in Table 3. Although the oxy-fuel combustion presented the highest consumption of electricity, due mainly to the ASU, this technology presented the lowest cost in the refineries and chemical plants. Namely, the cost of CO₂ avoided was 24.7–58.7 and 38.1–127.8 €·t_{CO₂}⁻¹, respectively. The equivalent energy consumption was shown to range between 1.9 (long term) and 2.8 MJ_{th}·kg_{CO₂}⁻¹ (short term) for the refineries, with a CO₂ capture rate between 65% and 76%. For the chemical plants, the energy consumption was higher in the range 2.7–3.9 MJ_{th}·kg_{CO₂}⁻¹ (short term) and 2.2–5.2 MJ_{th}·kg_{CO₂}⁻¹ (long term), but higher CO₂ capture rates were achieved (88%–100%).

3.4.3 Discussion

Unlike the two previous EIIs, in the last decade, very few techno-economic data have been published in the literature for the petroleum refining industry. To date, amine scrubbing was the most studied technology, while solid looping technology has not been widely considered for this industry. Only one paper was found in the literature [49], dated for more than one decade and then not reviewed in this study. CLC was the technology studied in the referred paper and no data about calcium looping was found. Regarding the economic feasibility, oxy-fuel combustion seems to be the least expensive technology with a mean CO₂ avoided cost of around 80.6 against 102.4 €·t_{CO₂}⁻¹ for amine scrubbing, Fig. 10. It should be noted that only the results of one source, for oxy-fuel combustion, is represented in this figure. Once again, some discrepancies are visible in the CO₂ avoided costs, which is strongly dependent on the volume of gas to be treated and CO₂ concentration. This is particularly

important in this industry due to its heterogeneity and numerous points of CO₂ emissions. Although the oxy-fuel combustion presents the lowest mean cost, as well as the lowest equivalent energy consumption (3.0 against 3.2 MJ_{th}·kg_{CO₂}⁻¹ for amine scrubbing), the implementation of this technology can be limited to individual sources of CO₂. On the contrary, deployment of the amine scrubbing, as an end-pipe technology, can be retrofitted to capture the CO₂ from combining streams. It is worthwhile to mention that the mean equivalent energy consumption for amine scrubbing is very similar to oxy-fuel combustion because the pre-combustion was shown to offer better opportunities for heat integration.

3.5 Pulp and paper industry

The pulp and paper industry is the 4th energy-intensive industry, being responsible for 2% of the global industry emissions in 2018 [6]. These CO₂ emissions are released along the process, and for the extraction, manufacturing and transport of raw materials [116]. Although the CO₂ emissions depend on the operation conditions, the recovery boiler is the major contributor to the CO₂ released in the pulp and paper plants. The CO₂ produced in the recovery boiler during the black liquor combustion accounts for up to 75% of the total CO₂ emissions [91]. The power boiler, whose purpose, along with the recovery boiler, is to meet the heat and power requirements of the plant, is another source of CO₂ emissions. The remaining CO₂ emissions are released during the calcination of lime mud and the combustion of fuel, usually of fossil origin, in the lime kiln. The highest concentration of CO₂ occurs in the flue gas produced in the lime kiln, around 20 vol%, while the recovery and power boiler generates lower CO₂ purity streams, 13 and 12 vol%, respectively [21,44,117].

Although paper and paperboard production has increased by more than 25% in the last two decades, energy consumption has seen a rise of only 6%. This can be explained by the fact that more recycled paper has been produced [118]. However, in order to achieve deep decarbonisation, CCS has been appointed as a path to follow in this industry [5]. The simplified process of a pulp and paper plant is represented in Fig. 11.

Table 3 CO₂ capture rate, equivalent energy consumption and cost of CO₂ avoided for oxy-fuel combustion

Item	Short-term	Long-term	
Refineries	CO ₂ capture rate/%	65–73	70–76
	Equivalent energy consumption/(MJ _{th} ·kg _{CO₂} ⁻¹)	2.6–2.8	1.9–2.2
	Cost of CO ₂ avoided/(€·t _{CO₂} ⁻¹)	53.6–58.7	24.7–31.9
Chemical plants	CO ₂ capture rate/%	100–95	100–88
	Equivalent energy consumption/(MJ _{th} ·kg _{CO₂} ⁻¹)	2.7–3.9	2.2–5.2
	Cost of CO ₂ avoided/(€·t _{CO₂} ⁻¹)	82.4–127.8	38.1–74.2

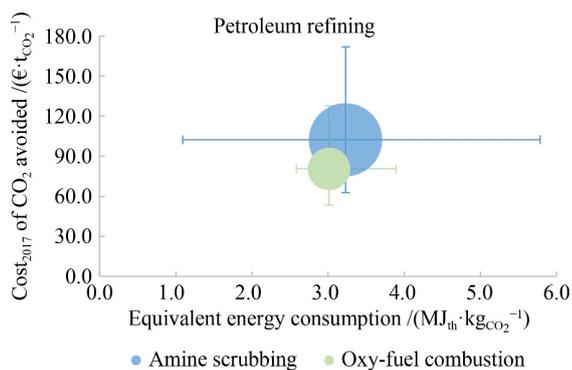


Fig. 10 Techno-economic performance of different CO₂ capture technologies for decarbonisation of petroleum refining industry: equivalent energy consumption vs mean CO₂ avoided cost (Error bars represent the range of figures found in the literature. The area of the bubble is proportional to the number of works reviewed).

3.5.1 Chemical absorption (post-combustion amine scrubbing)

In the last decade, amine scrubbing has been the focus of attention in 85% of the studies carried out about CCS retrofitted to pulp and paper plants.

Hektor and Bertsson [119] evaluated the techno-economic feasibility of CO₂ capture from the flue gases in the recovery boiler by amine scrubbing (monoethanolamine), with a CO₂ capture rate of 90%. This study was performed for a pulp plant and for an integrated pulp and paper plant. Since this technology demands steam for the solvent regeneration, five different configurations have been assessed to satisfy this additional steam requirement: 1) upgrade of biofuel boiler, 2) implementation of a nat-

ural gas combined cycle (NGCC) to replace the biofuel boiler; 3) recovery of low-grade heat available by a heat pump and 4) thermal process integration with a larger biofuel boiler or 5) with NGCC. They concluded that for the integrated pulp and paper plant, NGCC and the process integration with NGCC alternatives were the best options in terms of thermodynamic performance, as these resulted in an energy surplus. It should be noted that the reference plant was an electricity importer. These two options have an equivalent energy consumption of -0.9 and -0.8 MJ_{th}·kg_{CO₂}⁻¹, respectively. The negative sign means the CO₂ capture does not represent an energy penalty for the plant because the CO₂ capture was associated with an improvement of the process (implementation of NGCC). The remaining options required additional electricity importation, which translated into an equivalent energy consumption between 2.4 and 3.4 MJ_{th}·kg_{CO₂}⁻¹. Under different energy market scenarios, they concluded that to capture 90% of CO₂ emissions, the CO₂ avoided cost ranged between 32.9 and 57.9 €·t_{CO₂}⁻¹ for the pulp plant and between 22.7 and 75.0 €·t_{CO₂}⁻¹ for the integrated pulp and paper plant, for a future scenario in 2020. These costs are only valid for transportation until 500 km. It is noteworthy that the costs were estimated based on the CO₂ allowance price of 27 and 43 €·t_{CO₂}⁻¹ that reflect the current trends in CO₂ pricing.

Application of amine scrubbing, Fluor Corporation's Econamine FG Plus[®], into a primary recovery boiler was also assessed by McGrail et al. [3]. As mentioned previously, this technology requires additional steam, which the authors fulfilled with a larger biomass boiler, integrated in the power island (37 MW_e), that replaced the

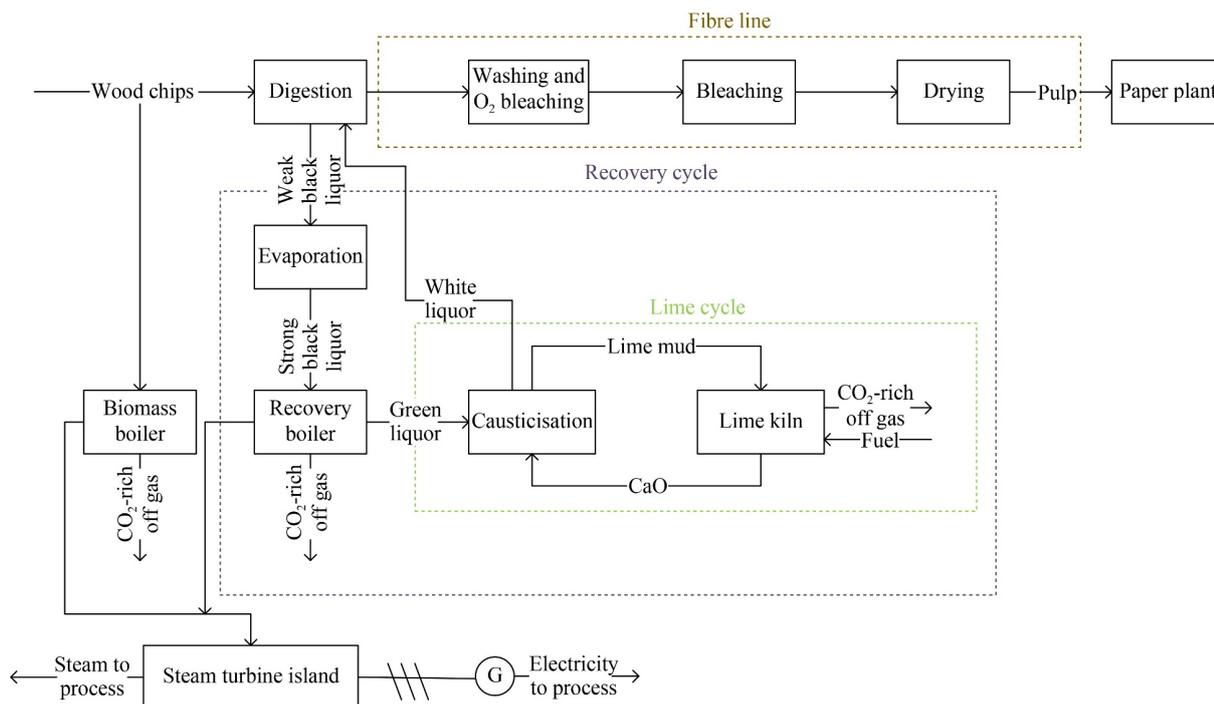


Fig. 11 Simplified diagram of a pulp and paper plant.

existing natural gas and hog boilers. This boiler meets the steam requirements for both the paper and CCS plants, whereas producing enough steam to ensure an electricity surplus. This new design would permit to work without CO₂ capture and thus the additional steam used to produce more electricity. Comparing the new plant with and without CO₂ capture, the equivalent energy consumption was 2.2 MJ_{th}·kg_{CO₂}⁻¹. They estimated a CO₂ avoided cost of around 52.5 €·t_{CO₂}⁻¹, in the same order of magnitude as the previous work, but only 62% of CO₂ is captured.

A detailed evaluation of amine scrubbing (monoethanolamine), retrofitted to a pulp plant and integrated pulp and paper plant, was carried out by Onarheim et al. [43]. They assessed different six CO₂ capture configurations, CO₂ capture from 1) recovery boiler, 2) power boiler, 3) lime kiln and stream combinations from 4) recovery and power boilers, 5) recovery boiler plus lime kiln and 6) recovery and power boilers plus lime kiln, which corresponded to an overall CO₂ capture rate between 9.1% (configuration 3 in both cases) and 90% (configuration 6 in the pulp plant). They found that the excess energy of the standalone pulp plant was enough for the CCS unit. However, in the integrated pulp and paper plant, this did not meet the energy requirements for configurations 1 and 4–6. Depending on the capture level, the equivalent energy consumption was within the range 2.0–3.0 MJ_{th}·kg_{CO₂}⁻¹ for the pulp plant. On the other hand, the integrated pulp and paper plant has shown an equivalent energy consumption between 2.2 and 5.4 MJ_{th}·kg_{CO₂}⁻¹. Similarly, to the study of Hektor and Berntsson [119] the cost associated with the integrated pulp and paper plant was higher than for a standalone pulp plant. The cost of CO₂ avoided, assessed under various market scenarios, was in the range of 72.4–90.7 €·t_{CO₂}⁻¹ for the first one, while the second plant presented costs comprised between 53.0 and 67.3 €·t_{CO₂}⁻¹. These figures are valid for an overall CO₂ capture rate of between 60% and 90%.

Garðarsdóttir et al. [91] have assessed the cost associated with CO₂ capture, amine scrubbing (monoethanolamine), in a pulp and paper plant in Sweden. They found that, for a CO₂ capture rate of 90%, the cost of CO₂ capture was 63.0 €·t_{CO₂}⁻¹, which had slightly favoured affected by an increase in CO₂ concentration. This translated into an equivalent energy requirement of 5.0 MJ_{th}·kg_{CO₂}⁻¹, which is in agreement with the results obtained by Onarheim et al. [43]. Note that these figures correspond only to the capture of CO₂ from the flue gases produced by the recovery boiler, with a CO₂ concentration of 13 vol%.

Amine scrubbing was also studied by Nwaoha and Tontiwachwuthikul [44], which proposed the use of 2-amino-2-methyl-1-propanol blend with a conventional monoethanolamine to capture the flue gas from recovery boiler, lime kiln and power boiler. Although it was not possible to estimate the equivalent energy consumption, this would be lower for the cases where the 2-amino-2-

methyl-1-propanol blend with a conventional monoethanolamine is used, once its regeneration duty (3.2–4.7 MJ_{th}·kg_{CO₂}⁻¹) is lower than monoethanolamine (4.2–5.2 MJ_{th}·kg_{CO₂}⁻¹). They have compared the performance of the blend with the one by a single monoethanolamine under different configurations. They found that the cost of CO₂ captured was lower for the cases which in the 2-amino-2-methyl-1-propanol blend with a conventional monoethanolamine was used (108.0–110.5 €·t_{CO₂}⁻¹), compared to 115.1–123.3 €·t_{CO₂}⁻¹ (monoethanolamine). They have also concluded that the costs of CO₂ captured could be reduced with new 2-amino-2-methyl-1-propanol-based amine blends and the process configurations proposed in this study.

3.5.2 Solid looping cycles—calcium looping

The calcium looping retrofit to a pulp and paper plant was proposed for the first time by Santos et al. [21]. The inherent decarbonisation of the Kraft process, by integration of calcium looping in the existing lime cycle, was techno-economically assessed for different design configurations and under market scenarios. They found that an integrated pulp and paper plant could turn from an electricity importer to an electricity exporter. The equivalent energy consumption estimated was 3.7 MJ_{th}·kg_{CO₂}⁻¹. This study showed that the calcium looping performance is superior to post-combustion amine scrubbing, once the retrofit of latter to pulp and paper plant in general translated in a decrease of net power production if the retrofit of CCS to the plant is not linked to any process improvement. Furthermore, in some cases, the implementation of an auxiliary boiler is necessary to meet the energy requirement. The CO₂ avoided cost to capture 90% of the CO₂ emitted by the plant, was 39.0 €·t_{CO₂}⁻¹, under the baseline scenario.

3.5.3 Discussion

From the four EIIs, the pulp and paper industry has been the one with less attention being paid by the researchers. As a result, the techno-economic data of CO₂ capture retrofitted to this industry is scarce. Only two technologies, amine scrubbing and calcium looping, were assessed to date. Figure 12 shows the equivalent energy consumption versus the cost of CO₂ avoided. However, to ensure a fair comparison, only the data for integrated pulp and paper are represented. Calcium looping presented the lowest mean avoided cost (39.0 €·t_{CO₂}⁻¹) against amine scrubbing (62.7 €·t_{CO₂}⁻¹). This can be explained by the reduction of material costs, as part of fresh limestone is replaced by lime mud from the Kraft process, and the plant achieves an additional revenue from sales of the electricity exported to the grid. While the equivalent energy consumption is similar for both technologies (2.9 against

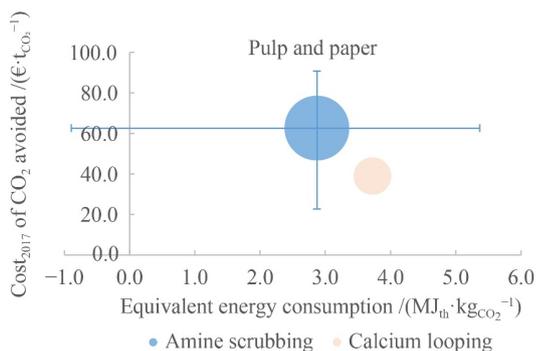


Fig. 12 Techno-economic performance of different CO₂ capture technologies for decarbonisation of pulp and paper industry: equivalent energy consumption vs mean CO₂ avoided cost (Error bars represent the range of figures found in the literature. The bubble without error bars has only one source. The area of the bubble is proportional to the number of works reviewed).

3.7 MJ_{th} · kg_{CO₂}⁻¹ for amine scrubbing and calcium looping, respectively), calcium looping presents a clear advantage to amine scrubbing. Since the calcium looping temperatures (600–900 °C) are higher than amine scrubbing (40–50 °C), there is a higher heat recovery potential. This was confirmed by Santos et al. [21], who showed that an importer pulp and paper industry could turn into an electricity exporter by integration of calcium looping in the lime cycle. As can be seen in Fig. 12, the range of equivalent energy consumption for amine scrubbing is high due to the different configurations assumed to deliver the additional energy requirement by the CCS plant. The cases that consider the upgrade of the reference plant present the lowest equivalent energy requirement, with the cases with NGCC implementation showing values below zero since the efficiency of the process is enhanced.

4 Perspective for decarbonisation of EIs

As can be seen in Fig. 13, the current literature shows a substantial uncertainty in the estimates of the cost of CO₂ avoided for CCS. The most mature technology, amine scrubbing, is also the technology that presents higher costs associated with the highest uncertainty. The cost of CO₂ avoided varies between 22.7 (pulp and paper) and 205.8 € · t_{CO₂}⁻¹ (iron and steel). Although there is limited evidence in the current literature, these costs could be reduced by using phase-change amines. These are promising amines as their regeneration heat requirement has been shown to be lower than that for conventional amines. On the other hand, the costs associated with CO₂ capture using calcium looping were shown to be one of the lowest for the three industries analysed (iron and steel, cement and pulp and paper). It is noteworthy to mention that these figures are based on studies considering natural CaO-based sorbents. These sorbents are

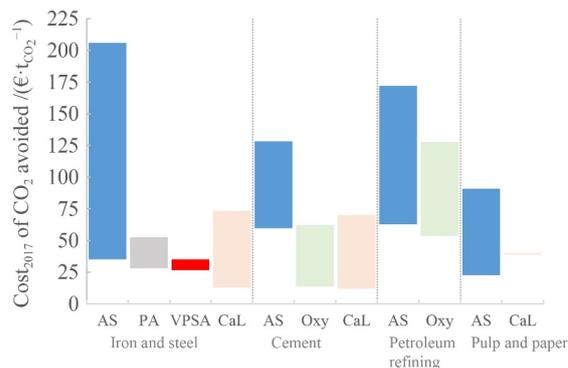


Fig. 13 Cost of CO₂ avoided of each technology for Energy Intensive Industries (AS: amine scrubbing; PA: physical absorption; CaL: calcium looping; Oxy: oxy-fuel combustion; VPSA: vacuum pressure swing adsorption).

characterised with lower sorbent cost but have a high rate of degradation compared to synthetic sorbents. The consideration of synthetic sorbents with higher sorption capacity would overcome this drawback, but at the expense of higher sorbent costs. The CO₂ avoided cost varied between 12.0 and 73.5 € · t_{CO₂}⁻¹ for cement and iron and steel industries, respectively. In the pulp and paper industry, the cost falls within this range. Nevertheless, it should be noted the economic feasibility evaluation is very limited for the iron and steel industry and the pulp and paper industry, with only one economic assessment published to date for the latter one. Besides, because there is no standardised methodology for techno-economic assessment, a transparent and fair comparison of the different technologies for decarbonisation of EIs is challenging. It is, therefore, crucial to standardise the assumptions and assessment frameworks for techno-economic assessment, as well as, to systematically assess the uncertainty in techno-economic performance via stochastic modelling. Such an approach would account for the uncertainty associated with the technology operating conditions and the market conditions. There is some evidence in the current literature indicating that these challenges are gradually being addressed. Garcia and Berghout [120] have proposed a cost method to assess the CCS costs in the cement and iron and steel industries, which could be extended to the other two EIs. More recently, Roussanaly et al. [121] have proposed some recommendations that should be taken into consideration for a fair comparison between economic studies. These guidelines can be applied to different industries. It is also important to emphasise that beyond the standardised techno-economic assessments, it is crucial to assess the impact of the integration of CO₂ capture technology in the industrial process on product quality. This is especially important when the CO₂ capture technology requires modification of the original process, such as in the case of oxy-combustion and calcium looping retrofits.

Although there are around world CCS pilot plants already in operation, the carbon capture technologies still need to scale up to a large scale. Due to its process

characteristic, the implementation of CCS in the petroleum refining industry is demanding. This industry is characterised by high heterogeneity of flue streams which, in most of the studies, are assumed to be combined. This assumption may lead to misleading results, overestimation of thermodynamic performance and underestimation of costs since, in practice, this is hard to be accomplished and, up to this point, not put in practice. In the pulp and paper industry, because the primary source of energy is biomass, the CO₂ emissions are considered neutral and so, not accountable in the energy sector and no incentive for carbon capture. For this reason, the inclusion of biogenic emissions in the European Union Emissions Trading System and/or on the attribution of credits for them is the only way to unlock CCS implementation in this industry.

Overall, it is important to keep assessing the techno-economic feasibility of CCS as well as CCU potential that would be useful to identify possible CCS/CCU clusters. Besides, the potential of achieving negative carbon emissions by use of BECCS should also be the focus of future research.

5 Conclusions

This work presented a review of the state-of-the-art in CCS technology for decarbonisation of the main four EIIs, including iron and steel, cement, petroleum refining and pulp and paper industries. Only the papers published in the academic literature in the last decade were reviewed.

CCS is a feasible option for the decarbonisation of the four EIIs analysed, presenting costs between 12.0 (calcium looping retrofitted to cement industry) and 205.8 €·t_{CO₂}⁻¹ for amine scrubbing implementation in the iron and steel industry. A direct comparison between CO₂ capture technologies is difficult to be done since there is no methodology and assumptions standardisation, which has an impact in the techno-economic performance. Regarding the economic analysis, while some authors estimated the cost accounting for the CO₂ avoided, others considered the cost of CO₂ captured. Furthermore, the costs of CO₂ transport and storage, in some studies, are also neglected. Besides, the estimation of the specific energy consumption of each technology is a tough task because, in some studies, it is not clear what is taken into account as the power consumption required for CO₂ compression. Therefore, a wide range in the thermodynamic and economic analysis was observed and the identification of the best CO₂ capture technology is challenging. Thus, these observed discrepancies cause some uncertainty and, therefore, a delay in the technology deployment at a commercial scale.

The review of the current literature has indicated that,

to date, there is no dominant CO₂ capture technology. However, high-temperature solid looping cycles seem to be an emerging technology with the potential to be implemented across the EIIs studied, excluding petroleum refining due to lack of data. Importantly, one of the main drawbacks of amine scrubbing, which is the high-heat requirement for solvent regeneration, could be overcome by new amines and new configurations. However, as this technology operates at lower temperatures than solid looping cycles, the heat recovery potential is significantly diminished, resulting in lower overall process efficiency. Furthermore, some authors showed that calcium looping can also be integrated into iron and steel, cement and pulp and paper, using the inherent potential of the process for CO₂ capture. Since the limestone used as a sorbent in calcium looping is partial or totally integrated in the industrial process, there is a reduction in the material cost too. Further work should focus on standardising the techno-economic assessment of technologies for industrial decarbonisation to support industry and policy decision makers in deriving reliable decarbonisation pathways.

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