Review

CO$_2$ Capture, Use, and Storage in the Cement Industry: State of the Art and Expectations

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Abstract: The implementation of carbon capture, use, and storage in the cement industry is a necessity, not an option, if the climate targets are to be met. Although no capture technology has reached commercial scale demonstration in the cement sector yet, much progress has been made in the last decade. This work intends to provide a general overview of the CO$_2$ capture technologies that have been evaluated so far in the cement industry at the pilot scale, and also about the current plans for future commercial demonstration.

Keywords: CO$_2$ capture; cement; post-combustion; absorption; membranes; adsorption; calcium looping; oxyfuel; direct separation

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1. Introduction

The global production of cement reached 4.2 Gt in 2019 [1]. The main producer is China (55%), followed in the far distance by India (8%) [2]. The market is expected to grow in the forthcoming decades up to 12–23% by 2050, as a result of the increasing global population and urbanization trends, coupled with infrastructure development needs [3].

Figure 1 represents a state-of-the art cement manufacturing process. Limestone (or other calcium carbonate source) is ground with clay and other minor components, and fed to the preheater, where the raw meal countercurrently contacts the hot kiln exhaust gases in a series of vertical cyclones, being heated up to approximately 900 °C. At the bottom of the preheater, in the calciner, limestone decomposes to form calcium oxide, releasing CO$_2$ (see Reaction 1). This is referred to as process emissions, because they are inherent to the clinker manufacturing process. Most of the CO$_2$ is normally released from the limestone raw material in the preheater and the calciner [4]. The precalcined meal is heated up to 1450 °C in the rotary kiln, where the calcination is completed, and the calcium oxide reacts with silica, alumina, and iron oxides, to form the calcium silicates, aluminates, and ferrites that constitute the clinker. At the kiln outlet, the clinker is rapidly cooled in the grate cooler using incoming combustion air. Finally, the cooled clinker is mixed with gypsum and ground into a powder, to produce Portland cement, or with additional components—such as slag, fly ash, or limestone—that substitute part of the clinker, to produce blended cement. Each cement product has its own unique composition of raw materials, which depends strongly on the location of the cement plant and the availability of the raw materials.

$$CaCO_3 \quad CaO + \ CO_2 \tag{1}$$

Due to its size and the inherent characteristics of its production process, the cement sector is one of the main sources of anthropogenic CO$_2$, accounting for 8% of global emissions [5]. Up to 0.95 t of CO$_2$ are released per t of cement produced. The actual carbon footprint depends on the ratio of clinker to cement, the manufacturing process (dry or wet method), the level of heat recovery,

the fuel used, the moisture content of the raw materials, and the capacity of the plant, among other factors. Process emissions account for approximately 65% of the direct CO_2 emissions, whereas fuel combustion is responsible for the remainder [6]. Total CO_2 emissions have increased by 200% since 1990, mainly driven by the growth in cement demand, despite the decrease in the global average clinker ratio [7], the significant improvement on energy efficiency, and the increasing rate of alternative fuels used [3]. However, coal still remains the dominant fuel, with a share of 65% of the total fuel consumption [6]. On the other hand, the net climate impact of concrete products needs to account for the fact that 11–43% of the initial process emissions may be offset by gradual reabsorption of atmospheric CO_2 by the exothermic carbonation of hydrated cement [8].
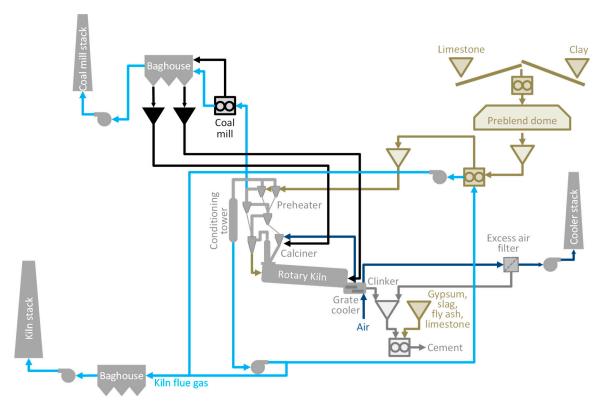

Figure 1. Scheme of a state-of-the art cement manufacturing process.

The Paris Agreement, ratified by 189 of the Parties, aims to limit global warming to well-below 2 °C above pre-industrial levels and to pursue efforts to restrain it to 1.5 °C. Limiting warming to 1.5 °C implies reaching net zero CO_2 emissions globally around 2050 [9]. Therefore, it is necessary to decarbonise the cement industry at a global scale.

All regions have improved the energy efficiency and reduced the carbon intensity of their cement industries. China, India, and the European Union, which are the three main cement producers, have adopted policy measures to reduce the carbon footprint of the cement industry, through emissions trading schemes, energy efficiency targets, replacement of old plants, new cement standards, and circular economy. Private-led initiatives are also gaining momentum. In 2015, 18 cement companies established the shared ambition to reduce their CO_2 emissions by 20–25% compared to business as usual by 2030, which represents a mitigation effort of about 1 Gt CO_2 [3]. The Global Cement and Concrete Association (GCCA) includes worldwide members that sum up to 50% global cement production capacity. In 2019, GCCA launched Innovandi, the Global Cement and Concrete Research Network, which intends to research areas such as the impact of co-processing, the efficiency of clinker production, the implementation of Carbon Capture Use and Storage (CCUS) technologies, the impact of clinker substitutes and alternative binders in concrete, low carbon concrete technologies, and re-carbonation [10]. Thirty companies from the cement and concrete sectors, have already committed to the initiative [11].

The European Cement Association (CEMBUREAU) has publicly stated its ambition to reach carbon neutrality along the value chain by 2050 [12]. HeidelbergCement has committed itself to reduce its specific net CO_2 emissions per ton of cement produced by 30% compared to 1990 levels by 2030, and has a vision to reach carbon neutral concrete by 2050 at the latest; this is the first cement company in the world to receive approval for science-based CO_2 reduction targets [13], and the only cement manufacturer that has been included in the CDP's Climate Change A list 2019 [14]. It aims to attain net zero carbon footprint by augmenting the percentage of CO_2-neutral feed materials and combustibles, arriving at lower clinker cement classes, and by CCUS. Dalmia cement has also committed to become carbon neutral by 2040. Other companies are heading the same way.

In the International Energy Agency's (IEA) 2 °C scenario (2DS), which is consistent with at least a 50% probability of limiting the average global temperature increase to 2 °C by 2100, the direct emissions of the cement sector are reduced by 24% by 2050. This implies a cumulative reduction of emissions of 7.7 Gt CO_2 compared to the IEA's Reference Technology Scenario (RTS), which considers the commitments by countries to limit carbon emissions, including the nationally determined contributions (NDC) pledged under the Paris Agreement, which would result in an average temperature increase of 2.7 °C [3]. With increasing climate ambition, the mitigation effort grows larger: in order to have a 50% probability of limiting the average global temperature increase to 1.75 °C, (IEA's Beyond 2 °C Scenario, B2DS) additional 3.2 Gt CO_2 of cumulative CO_2 emissions reduction would be necessary compared to 2DS [3]. Obviously, if the most ambitious climate objective of the Paris Agreement—i.e., limiting the global average temperature increase to 1.5 °C—is to be pursued, the mitigation effort would need to increase accordingly. However, according to CEMBUREAU's carbon neutrality roadmap, reaching net zero emissions along the cement and concrete value chain is achievable by 2050 [12].

There is a series of mitigation measures that can contribute to the decarbonization of the cement industry: improving energy and materials efficiency (on a life cycle approach); switching to less carbon intensive fuels; reducing the clinker content in the cement by partly replacing it with cementitious materials with lower carbon footprint; developing new, innovative and clean production technologies, including excess heat recovery (EHR) to power generation, integration of renewable power generation, CCUS; and improving transport efficiency [3].

As shown in Figure 2, in the 2DS, the largest cumulative direct CO_2 emissions reduction by 2050, compared to RTS, comes from the reduction in the clinker ratio, followed by CCUS, development of clean production technologies, fuel substitution, and thermal efficiency [3].

Broadly, CCUS prevents CO_2 from being released into the atmosphere by capturing it, and either using it, or injecting it in geological formations for permanent storage. Under the B2DS, the amount of CO_2 captured annually by 2050 more than doubles that of the 2DS (see Figure 3), and the share of total direct CO_2 emissions cumulatively captured increases from 25% to 63%. To achieve carbon neutrality by 2050, the role of CCUS would be even higher, as this is necessary to achieve deep levels of decarbonization, given the large share of process emissions in the cement sector. The cement industry undoubtedly needs to deploy CCUS technologies. Although there are not yet CCUS facilities operating in the cement sector at large scale, there are CCUS facilities currently operating at the power and industrial sectors with a total annual capacity close to 40 Mt CO_2 [15].

This work intends to review the current state of the art of the CCUS technologies in the cement sector, including pilot scale demonstration studies and announced large scale projects that could start operation in the near term.
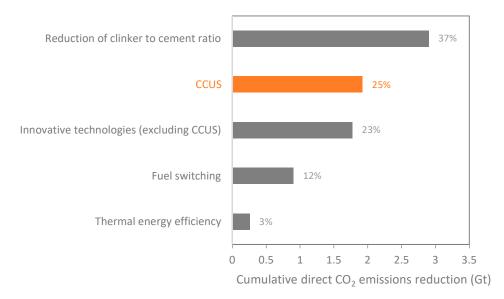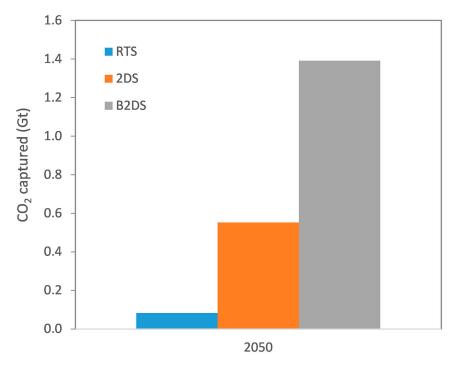
Figure 2. Global cumulative CO_2 emissions reductions by 2050 (2DS vs. RTS) (adapted from [3]).

Figure 3. Global direct CO_2 emissions from the cement sectors captured annually according to different IEA scenarios (data taken from [3]).

2. State of the Art of CO_2 Capture in the Cement Industry

In the Intergovernmental Panel on Climate Change's (IPCC) Special report on CCS, launched in 2005, four different types of CO_2 capture technologies were distinguished: post-combustion, pre-combustion, oxyfuel combustion, and industrial separation [16]. This classification differentiates industrial CO_2 capture from that carried out in the power sector. However, the separation technologies that can be used to capture CO_2 in industry can have many similarities with those of the power sector, depending on the specific process considered. In the case of the cement industry, CO_2 capture can be accomplished using post-combustion and oxyfuel combustion technologies.

Pre-combustion capture technologies would have limited mitigation potential in the cement sector, as they could deal only with the energy-related CO_2 emissions [17]. They might be of use

in new cement plants integrated with gasification technologies to produce syngas or H_2 fuel [18]. However, hydrogen flames have a relatively low emissive power, which makes them unsuitable for clinker manufacturing in conventional kilns [19]. New and more efficient hydrogen burners and cement kiln lines would be required.

A third type of CO_2 capture technology with great promise to be implemented in the cement sector is direct capture.

The state of the art of these groups of technologies in the cement sector will be discussed in the following sections.

2.1. State of the Art of Post-Combustion CO_2 Capture Technologies in the Cement Sector

A priori, conventional kilns of existing and new cement plants could be retrofitted with post-combustion CO_2 capture technologies relatively easily, without substantially modifying the cement manufacturing process. Only the energy management strategies and the start-up and shut-down procedures would be affected [19,20]. The inherent characteristics of the sector, the fact that actual kilns still have several life years ahead (expected 30–50 years lifetime), and that new kilns adapted to legal requirements are not thought to be built in the next years, reinforce the potential of post-combustion CO_2 capture technologies [21].

The kiln off-gas holds a temperature in the range of 85–105 °C to 150–180 °C, depending on the manufacturing process, and is at near atmospheric pressure [22]. These conditions are not ideal for the point of view of the separation process. On the other hand, the kiln-off gas presents a higher carbon dioxide concentration (up to 30% by volume after the preheater [19]) than other flue gas streams, including those of coal-fired power plants, which facilitates the capture step.

There are different post-combustion technologies that can be used to capture the CO_2 from the kiln-off gas: chemical absorption, membranes, and sorption with solids. A brief review of the state of the art of each of them is presented below.
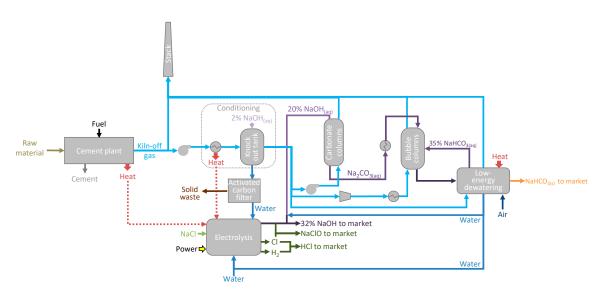
2.1.1. Chemical Absorption

SkyMine® Process

The largest demonstration of CCUS in the cement sector to date, started to operate in September 2015, at Capitol Aggregates' San Antonio cement plant in Texas, United States of America, with a capacity of over 75,000 t CO_2/y. The pilot plant was designed to make use of the SkyMine® process to capture 90% of the CO_2 from a slipstream of the cement plant, which represented approximately 15% of its total CO_2 emissions. The SkyMine® process produces marketable by-products, such as baking soda, hydrochloric acid, and bleach (see Figure 4), with a lesser CO_2 footprint than the conventional production processes, which contributes to additional CO_2 savings by product displacement (up to a total benefit of 224,110 t CO_2/y) [23]. The kiln-off gas, with an annual average composition of 29.58 vol % CO_2, 11.20 vol %H_2O, 10.22 vol % O_2, 0.04 vol % SO_2, 0.01 vol % NO, and 0.05 vol % CO, is cooled from approximately 125 °C to 35 °C. A diluted solution of NaOH is used to knock out SO_2 and NO_x. 60% of heavy metals (mainly Hg) are removed with the condensate, which is filtered with activated carbon to recycle water to the process. Conditioned flue gas is then fed to a multicolumn chemical absorption system, where a concentrated NaOH solution reacts countercurrently with the CO_2 from the flue gas in two packed absorbers working in parallel to form Na_2CO_3 (Reaction 2). Then, the saturated Na_2CO_3 solution is fed to bubble column reactors where it reacts with pressurized flue gas to yield $NaHCO_3$ crystals (Reaction 3). The resultant slurry is separated in a centrifuge into solids, which are sent to the dryer, and liquids, which are further processed to recycle the concentrated solution to the bubble column reactor and excess water to the process. The NaOH solution is generated through electrolysis of NaCl brine in a membrane cell that produces HCl as byproduct [23,24].

$$2NaOH_{(aq)} + CO_{2\,(g)}Na_2CO_{3(aq)} + H_2O_{(l)} \tag{2}$$

$$Na_2CO_{3(aq)} + H_2O_{(l)} + CO_{2\ (g)} 2NaHCO_{3(s)} \tag{3}$$

Figure 4. Simplified diagram flow of the SkyMine® process integrated in San Antonio cement plant (adapted from [23,24]).

According to available data, electrolysis is responsible for 87% of the total power requirement of the overall process, which is 21.12 MW$_e$. The specific power consumption results 8.3 GJ$_e$/t CO$_2$ captured, or 2.8 GJ$_e$/t CO$_2$ avoided, if the additional CO$_2$ savings achieved by the displacement of more energy-intensive commodities in the market are considered. The total installed cost of the project was estimated to be $125 M [23].

Amine Scrubbing

Chemical absorption with amine solutions has been extensively used in industry since 1930 [25], and has reached commercial scale demonstration in coal fired power plants [26,27].

Although specific process details would depend upon the particular facility, a generic process has three main steps: (i) flue gas pretreatment in a Direct Contact Cooler (DCC), with SO$_2$, NO$_x$, and dust particle removal; (ii) countercurrent contact of pretreated flue gas with an aqueous amine solution in an absorber column, where CO$_2$ reacts with the amine at 40–60 °C and atmospheric pressure, producing a decarbonized gas stream that is vented to the stack; (iii) regeneration of the spent solvent in the stripper column, at 100–120 °C and 1.5–2 atm (the operational values change slightly depending on the solvent used [28]), where a high purity CO$_2$ stream is recovered, while the lean solvent is sent back to the absorber column, closing the loop [29].

Aqueous solutions of alkanolamines, like monoethanolamine (MEA) and diethanolamine (DEA), were traditionally used because of their rapid reaction rates and low cost [30]. The development of second generation solvents and the optimization of the technology has led to substantial energy savings, up to 60% in the power sector [15].

The main differences between the flue gas of a power plant and that of a cement factory, are its temperature, composition, and different size distribution of their particulate matter. The higher temperature of kiln-off gases would cause thermal degradation of the amines and greater losses through evaporation [31], and hence requires further cooling. The higher CO$_2$ partial pressure in the cement flue gas is a priori advantageous, although it can also be a challenge for the CO$_2$ absorber, as more heat of absorption is released for a smaller volume of flue gas, and thus, the CO$_2$ absorber temperature is likely to increase, shifting the equilibrium in a less favorable direction [32].

SO$_2$ reacts irreversibly with amines to produce corrosive salts, while NO$_X$ can produce nitric acid resulting in additional corrosion and amine degradation [19]. To avoid the aforementioned
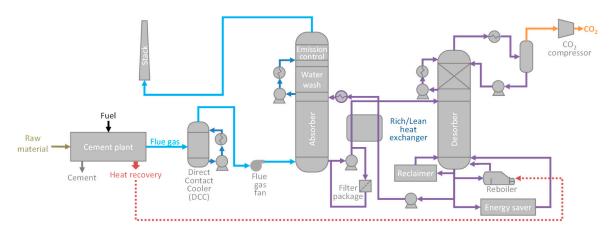
problems, it is necessary to remove the SO_2 and NO_2 levels below 10 and 20 ppm, respectively, prior to the absorber [33]. This pretreatment significantly increases the capital and operating cost of the overall process.

The main disadvantage of amine scrubbing is its high energy demand, mostly driven by the solvent regeneration, which contributes with 50–80% of the total energy requirements [34]. Moreover, in a cement plant there is no source of low pressure steam available to regenerate the solvent: at least 2 GJ/t CO_2 captured would need to be provided via a combined heat and power facility (CHP), or through waste heat recovery [20]. Furthermore, in modern cement plants, a significant fraction of waste heat is already used to dry the raw materials and preheat the raw meal. According with some simulations, no more than 15% of the additional thermal energy needs can be recovered from the cement kiln [35]. However, the actual amount of surplus waste heat is site dependent; this is strongly influenced by the moisture content of the raw materials. Calculations conducted for the Norcem's Brevik cement plant, in Norway, where the raw materials are relatively dry, indicate that using only waste heat, approximately 40% of the total CO_2 emitted by the plant could be captured, which correspond to 0.4 Mt CO_2/y [22]. More extensive heat integration (and costly) could lead to even higher capture rates, up to 85% of total emissions, using only waste heat [36].

The cost of the technology is dependent on the site location, the specific technology used, the steam source, the fuel price, and plant specific characteristic. The estimated cost for the retrofit of a cement plant with the reference MEA-based absorption technology is 80 €/t CO_2 avoided [37].

Although amine scrubbing is considered the benchmark technology for CO_2 capture in the cement industry, it is yet to be demonstrated at large scale in a cement plant. Norway's Longship project might be the first to achieve that milestone, being the first of its kind in the world. Aker Solutions' Advanced Carbon Capture (ACC^{TM}) technology (see Figure 5) and its S26 amine solvent, will be used to capture 0.4 Mt/y of the CO_2 emitted by the Norcem's cement factory located in Brevik, Norway, making use of available waste heat. The captured CO_2 will be liquefied and temporarily stored at Brevik facilities, and then shipped to an onshore terminal at Øyrgarden, on the Norwegian west coast, from where it will be transported by pipeline to an offshore storage location under the North Sea within the Northern Lights transport and storage project. The Norwegian Government has recently approved the final investment decision [38]. Operation is expected by 2023 [39]. Total investment in Longship project is estimated to be NOK 25 billion, although this also covers CO_2 capture at Fortum Oslo Varme's waste incineration facility [38].

Figure 5. Generic flow sheet of an ACC^{TM} process integrated in a cement plant.

Aker Solutions has extensively tested its ACC^{TM} technology making use of its advanced amine solvent S26 at pilot scale in Brevik factory for 18 months [40]. The Mobile Test Unit (MTU), installed at Brevik in April 2014, has a flue gas capacity up to 1000 Nm^3/h, and consists of an absorber with a diameter of 0.4 m and a packing height up to 18 m, a desorber with a diameter of 0.32 m and a packing

height of 8 m, and a proprietary anti-mist design. Typically, a slipstream of 450 Nm^3/h of the cement kiln flue gas was taken just before the stack, downstream of the selective non-catalytic reduction (SNCR), electrostatic precipitator (ESP), spray dryer flue gas desulfurization (FGD), and baghouse filter, with the following composition: 7.5 vol % O_2, 17.8 vol % CO_2, 18.2 vol % H_2O, 0–130 mg/Nm^3 SO_2, 180–250 mg/Nm^3 NO_x and 5–10 mg/Nm^3 dust. This corresponds to a CO_2 removal rate of 140–150 kg/h which is the full capacity of the reboiler [32]. SO_2, NO_x, and dust particles were knocked out from the flue gas with the condensate [22]. During field testing, which included over 5500 h, stable operation on flue gas from cement kiln was demonstrated, with 90% CO_2 capture, low amine consumption (<0.15 kg/t CO_2), low build-up of degradation products (heat stable salts mainly formed by slip of SO_2 through pre-scrubber), with low nitrosamine formation, low corrosiveness, and low emissions. The energy demand, without heat integration, for a 90% CO_2 removal rate is approximately 3 GJ/t CO_2, or 2.7 GJ/t CO_2 with the ACC^{TM} Energy Saver, and this can be entirely satisfied using the available low grade heat from the cement plant. Aker Solution's ACC^{TM} technology has been scaled-up and the S26 solvent qualified at Technology Centre Mongstad [36]. The technology has currently a technology readiness level (TRL) of 8 [39].

To date, the largest demonstration of CO_2 capture using amine absorption technology in the cement industry, with a capacity of 50,000 t CO_2/y, is located in Anhui Conch's Baimashan plant in the city of Wuhu, in the Ahnui province, China [41]. The pilot facility, which started to operate in 2018, captures 3% of the total CO_2 emissions of the cement factory. The CO_2 produced, with a purity of 99.99%, is transported by trucks and sold to industrial customers [42]. However, due to the limited local market for CO_2, the company has no plans to expand.

Dalmia Cement announced in 2019 its intention to build a large scale CCUS facility at one of its cement plants in Tamil Nadu, India, with a capacity of 0.5 Mt CO_2/y [43]. The facility will make use of Carbon Clean's CDRMax$^®$ technology, which combines the use of a proprietary solvent (amine promoted buffer salts, APBS) with novel heat integration, to provide CO_2 capture at an estimated cost of $40/t CO_2. The technology can be flexed to manage CO_2 concentrations in the source gas from 3% to 25% and to produce CO_2 with purities between 95–99.9% [44]. The partnership will explore possible uses for the CO_2 captured, including direct sales to other industries, or chemical manufacturing. No completion date or budget has been disclosed yet.

Chilled Ammonia Process

The chilled ammonia process (CAP) makes use of an ammonium aqueous solution to absorb CO_2 from the flue gases at ambient pressure and low temperature. This entails the use of low grade heat. Moreover, the solvent is not affected by oxygen or acidic trace components present in the incoming flue gas. The main chemical reactions involved in CAP are listed below [45]:

$$CO_{2\ (g)} \leftrightarrow CO_{2(aq)} \tag{4}$$

$$2NH_{3(aq)} + H_2O_{(l)} + CO_{2\ (aq)} \leftrightarrow (NH_4)_2CO_{3(aq)} \tag{5}$$

$$(NH_4)_2CO_{3(aq)} + H_2O_{(l)} + CO_{2\ (aq)} \leftrightarrow 2(NH_4)HCO_{3(aq)} \tag{6}$$

$$(NH_4)_2CO_{3(aq)} \leftrightarrow NH_2CO_2NH_{4(aq)} + H_2O_{(l)} \tag{7}$$

Reactions 4–7 are reversible and the direction depends on pressure, temperature and concentration.

A simplified flow diagram of CAP process is shown in Figure 6. The flue gas is first conditioned in a combined DCC and SO_2 absorber, where the flue gas is cooled and SO_2 is scrubbed with ammonia. Liquid ammonium sulphate $(NH_4)_2SO_4$ is formed as a by-product, which can be commercialized as a fertilizer. Optionally, ammonia can be recovered in a dedicated recovery unit, leading to a gypsum by-product [45]. The conditioned flue gas is then sent to an absorption column where CO_2 is removed by an ammonia solution at temperatures around 12–13 °C. To reduce the ammonia loses with the decarbonized flue gas, a water wash section is installed at the top of the absorber; ammonia is recovered

from the wash water in a dedicated stripper and recycled to the CO_2 adsorber. To further reduce the emissions of ammonia to the atmosphere, an acid wash section fed with H_2SO_4 is installed between the water wash and the stack. The spent solvent is regenerated in the CO_2 desorber at around 25 bar [46].
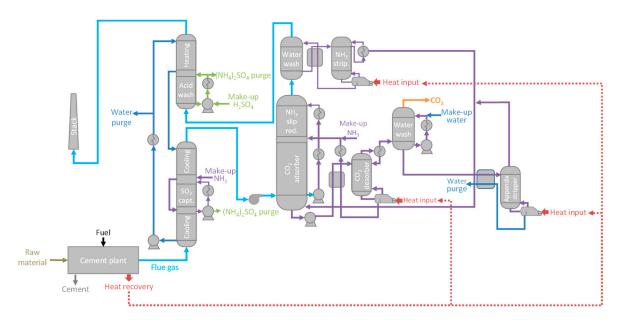

Figure 6. Process flow sheet of a CAP process integrated in a cement plant (adapted from [46]).

Within the European funded CEMCAP project (2015–2018), TRL of GE's CAP process for cement plants was advanced to TRL 6. Three experimental campaigns were carried out at a 1 t CO_2/d pilot plant of GE's Technology Center in Växjö, Sweden. The first campaign evaluated the effect of high CO_2 concentration in the absorber; the second campaign studied the combined DCC and SO_2 absorber, which reduces the SO_2 content below 1 ppm; and the last campaign investigated the flue gas water wash, which reduces the NH_3 slip from 1% to below 200 ppm [47]. Tests proved that operating conditions could be adapted to work under cement plant conditions, with CO_2 recovery rates of 90%, using a similar design than that of power plants. Moreover, the higher CO_2 concentrations of cement plant flue gases allowed enhancing the NH_3 absorber design, reducing its height and complexity. CAP is ready to be demonstrated at a scale of 100,000 t CO_2 captured/y in a cement plant [48]. For a standard 1 Mt clinker/y plant, the expected steam consumption for a 88% capture rate is 2.37 GJ steam/kg CO_2 [47]. The specific primary energy consumption per CO_2 avoided (SPECCA) of the CAP process for a 90% CO_2 capture rate is 3.75 GJ/t CO_2 captured [46], while the estimated cost of CO_2 avoided is around 66.2 €/t CO_2 [37].
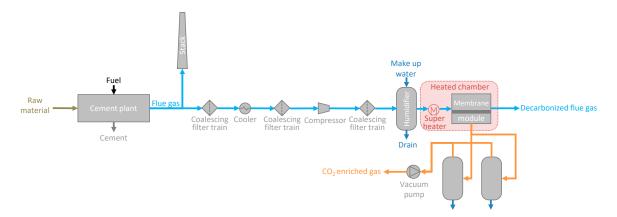
2.1.2. Membrane Separation

Gas separation membranes are based in physical and/or chemical interactions between the different gas components and the membrane material: one component passes through the membrane faster than another, being separated by the pressure gradient and the different diffusivities of the molecules. Membranes can be ceramic, polymeric (the most common), or hybrid. Selectivity is provided by the membranes themselves, and permeation rates would depend on parameters such as the relative molecular sizes, or their diffusion coefficient in the membrane. The driving force for the permeation across the membrane is the difference in the partial pressure of the components at both ends.

Membranes can be designed for a wide variety of roles, including carbon capture in the cement industry [19]. The higher concentration of CO_2 in cement flue gas compared to other sources, is advantageous for all separation technologies, and especially for membranes, because of the larger partial pressure driving force [49].

Membrane based separation selectivity is low, and only a fraction of the inlet CO_2 can be captured. In addition, the CO_2 purity is also limited, so usually multistage operation is required to fulfill the desired product standards. Membranes could theoretically reach capture rates larger than 80%, although only up to 60–70% recovery has been demonstrated at laboratory scale [17]. Purity obtained is about 90% (in a typical two-stage process), but it can be increased over 95% through new stages addition or by adding a low-temperature liquefaction unit [50]. Moreover, membranes are usually sensitive to sulfur traces, and polymeric membranes are mostly intolerant to high temperatures. Nevertheless, membrane systems do not require regeneration (although they require to be operated under pressurized conditions, or vacuum, which imply a high energy demand), and their overall footprint is lower than that of other technologies [17].

Membrane separation was demonstrated at pilot scale in Norcem CO_2 capture project (2013–2017), in Brevik cement plant, in Norway. The pilot facility, shown in Figure 7, was designed and constructed by Yodfat Engineers and operated by the Norwegian University of Science and Technology (NTNU) and DNV GL. This consisted of a one stage membrane module with 12 cassettes, with 2 flat sheets of polyvinylamine (PVAm) based fixed-site carrier (FSC) membranes per cassette, made by NTNU, with a total membrane area of approximately 1.5 m^2. Initial testing, carried out in 2014 with a slipstream of the plant flue gas downstream the SNCR, ESP, FGD, and baghouse filter, showed that it was difficult to achieve a stable and high performance of the membrane system. A CO_2 purity up to 72 vol % was achieved for short periods of time, when all process parameters were well controlled in the single stage FSC membrane system. The membranes withstood extended exposure to cement flue gas without showing damage, with promising recoveries in the range of 60–70% [22]. Nevertheless, the membrane efficiency in the plat-and-frame module was low, and suffered water condensation/corrosion issues.

Figure 7. Simplified flow diagram of the pilot membrane unit tested at Brevik cement plant (adapted from [51]).

In the MemCCC project, Air Products' commercial hollow fiber membrane modules were coated at NTNU with the PVAm based FSC membrane (up to 18 m^2), and tested at Brevik plant for 9 months at different conditions (including high NO_x and SO_2 concentrations) showing stable performance, and up to 70 vol % purity in a single stage. The techno-economic analysis for 80% CO_2 recovery and a purity of 95 vol %, showed a power consumption of 1.20 GJ_e/t CO_2 captured, and an estimated cost of 46.54–48.49 €/t CO_2 captured [51]. These results could be enhanced adding new stages or a low-temperature liquefaction unit [52].

In membrane-assisted CO_2 liquefaction (MAL) technology, the bulk separation of CO_2 is carried out by permeation through a membrane module, resulting in a permeate with a moderate purity, which is fed to a low temperature liquefaction unit, where it is conditioned, compressed, and cooled typically between −55 and −50 °C, leading to a high purity CO_2 liquid product and a gas stream that is vented to the stack, as shown in Figure 8.
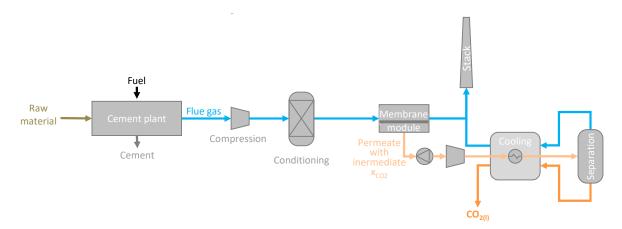
Figure 8. MAL system integrated in a cement plant (adapted from [53]).

Within CEMCAP project, MAL has been evaluated for cement production. Two polymeric membrane materials, perfluoropolymer and PEBAX-based membranes, were evaluated at bench scale at TNO, in Eindhoven, Netherlands, using CO_2/N_2 mixtures reaching high separation factors and selectivities of 20 and 45%, respectively, and a 78 mol % CO_2 concentration in the permeate side for the perfluoropolymer membranes, reaching the concentration requirements for membrane assisted CO_2 liquefaction. At the same time, CO_2 was separated and purified in a laboratory pilot plant at SINTEF Energy Research in Trondheim, Norway, with a capacity of 10–15 t/day. The low-temperature liquefaction process showed stable and robust, and it is easily scalable to a larger capacity [48]. Nevertheless, tests included only N_2/CO_2 mixtures. It is still necessary to carry out additional tests containing typical kiln-off gas impurities to establish the realistic obtainable purity of the captured CO_2. The estimated cost of retrofitting this technology to cement plants is 83.5 €/t CO_2 avoided, which is significantly higher than that of competing technologies [37].

2.1.3. Sorption with Solids

Adsorption is a surface process where the adsorbate molecules, originally present in the bulk fluid phase, tend to concentrate onto the surface of a solid adsorbent. When the phenomenon is not restricted to the surface of the solid, the term 'sorption' is preferred. Solids' sorption-based separation processes are led by selective attractions between a particular adsorbate, present in the bulk fluid mixture, and a solid sorbent. There are two principal mechanisms of gas sorption by solids: physical sorption (or physisorption) and chemical sorption (or chemisorption). These are differentiated by the nature of the interaction between the sorbate and the sorbent: intermolecular forces (van der Waals) or chemical bonds formation, respectively.

Sorption with Solids at Low Temperature

Candidate physisorbents for CO_2 capture are carbon materials, zeolites, alumino-phosphates (AlPOs), alumino-silico-phosphates (SAPOs), or metal–organic frameworks (MOFs) [54]. Chemisorbents generally include an active phase, such as amines or carbonates, supported on a porous support. Sorbent development seeks to achieve larger CO_2 adsorption capacities, faster kinetics, larger selectivity towards CO_2, milder regeneration conditions, better stability, tolerance to impurities, and lower costs.

In order to operate in a continuous basis, the adsorbent can be regenerated by decreasing the pressure, in pressure swing adsorption (PSA) processes, by increasing its temperature in temperature swing adsorption (TSA) processes, by using a purge gas in concentration swing adsorption (CSA) processes, or by a mixture of the previously mentioned methods of regeneration [55]. As the flue gas of a cement plant is at near atmospheric pressure, the adsorbent can be regenerated at subatmospheric

pressures in vacuum swing adsorption (VSA) processes. Process development seeks to reduce the overall energy consumption of the capture processes, while meeting the purity and recovery constraints.

The main advantage of adsorption-based CO_2 capture processes is their negligible environmental impact, with no toxic emissions or wastes, and without corrosion problems. Moreover, adsorption presents scope to reduce the energy penalty of the benchmark technology, amine absorption, although it has a lesser development stage.

The Research Triangle Institute (RTI)'s solid sorbent technology was evaluated against Aker solution's ACC^{TM} amine technology, membrane separation, and GE's regenerative calcium cycle (RCC) within Norcem CO_2 capture project (2014–1017). RTI's technology makes use of a high capacity chemisorbent (35 wt % Polyethyleneimine, PEI, loaded on silica), which flows countercurrently with flue gas in a multi-staged fluidized moving bed (SFMB) sorber, in a TSA process, where the sorbent circulates continuously between the sorber and the sorbent regenerator (also a SFMB) (see Figure 9). This type of reactor provides efficient heat management, critical to the chemisorption process, as any temperature rise in the absorber would reduce the CO_2 capacity of the sorbent. RTI evaluated the sorbent exposure to actual cement flue gas using an automated sorbent test rig (ASTR), which was installed at the Brevik cement plant, in Norway, during the first phase of the project. A slipstream of the flue gas was cooled, and the condensate knocked out. This demonstrated an effective way to reduce the contaminants that could impact RTI's sorbent, as most of them were removed with the condensate. Tests showed that exposure to 100 ppm of SO_2 caused a 30% drop in the CO_2 capacity of the sorbent. A RTI's prototype, with a sorbent inventory of 75 kg, was operated at Brevik cement plant between September and November 2016. Steady 80–90% CO_2 capture rate was demonstrated with an energy consumption of 2.4 GJ/t CO_2 avoided (without heat recovery), which is lower than that of the ACC^{TM} process. According to the technical and economical assessments carried out, the cost of CO_2 capture, for a minimum of 85% capture rate, without heat integration, is 45.8 €/t CO_2 avoided. For a minimum of 85% capture rate, but taking advantage of the available waste heat within the cement plant, it falls to 40.7 €/t CO_2 avoided. Using all the low grade heat available, without a minimum capture rate restriction, the cost falls to 38.6 €/t CO_2 avoided [56].
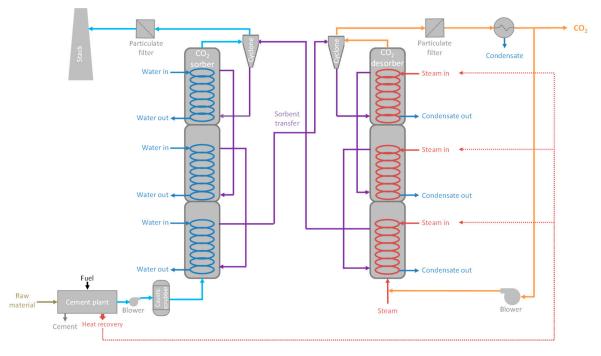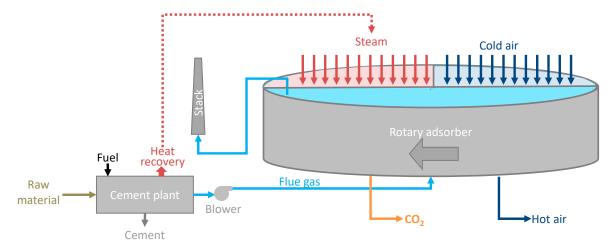

Figure 9. RTI's solid sorbent technology (adapted from [56]).

Adsorption-based technology might be close to commercial scale demonstration in the cement sector: in January 2020, Svante (formerly Inventys), LafargeHolcim, Oxy Low Carbon Ventures, and Total, launched a joint study to assess the viability and design of a commercial-scale carbon capture facility at the LafargeHolcim's Portland Cement Plant in Florence, Colorado, United States of America [57]. This initial scoping study was successfully completed in June 2020, and in September 2020 the United States Department of Energy's National Energy Technology Laboratory (DOE-NETL) awarded $1.5 million of federal funding to support the advancement of the LH CO2MENT Colorado Project. This would be the largest demonstration in the cement sector, with a capacity of up to 2 Mt of CO_2 captured annually [58]. The carbon capture facility would employ Svante' CCS technology, which is an intensified TSA process that makes use of a patented architecture of structured adsorbent (spaced sheets), and a proprietary process cycle design, the Veloxotherm^{TM} process, where a rotary adsorber is used to capture the CO_2 from the flue gas, and release it by displacement with steam (and vacuum, VTCSA), regenerating the adsorbent in a continuous manner and in a single unit (see Figure 10). Svante's process claims to be material agnostic: it can make use of different adsorbents, from carbons, to functionalized-silicas or metal–organic frameworks. The latter have high potential due to sharper temperature and pressure swing adsorption and desorption, which leads to lower parasitic energy loads, faster kinetic rates, and higher working capacity compared to conventional adsorbents, although they are still in development. As advanced adsorbents develop, cost reduction can be expected [59]. According to Svante's CEO, the abatement cost for the cement plant would be roughly $50/t CO_2. This fact, combined with the 45Q tax credit, which provides $35/t CO_2 stored through enhanced oil recovery (EOR), and the current CO_2 prices, of around $20 t CO_2, makes the project economically feasible [60].

Figure 10. Simplified flow diagram of Svante's Veloxotherm^{TM} process integrated in a cement plant.
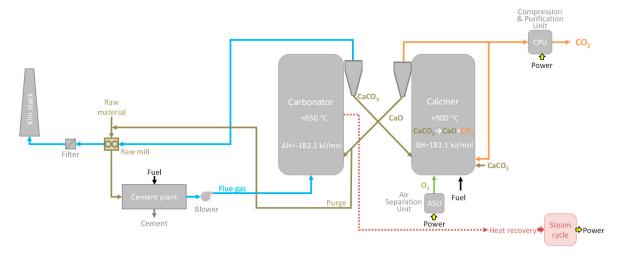
Svante's technology is being demonstrated at LafargeHolcim's cement plant in Richmond, British Columbia, Canada, at the smaller pilot scale of 1 t CO_2/d within the CO2MENT project (2019–2022), funded by the CO_2 Capture Project (CCP), the province of British Columbia and Canada's federal government. The project was launched in partnership with Total, with the objective to build the world's first full-cycle solution to capture and reuse CO_2 from a cement plant [61]. CO_2 capture started by the end of 2019 [59]. The CO_2 captured is currently being used for synthetic fuels production and injection in concrete and fly ash.

Calcium Looping

Calcium looping (CaL) is one of the most promising CO_2 capture technologies for the cement sector. It is based on the reversible carbonation (Reaction 8), which is generally carried out in two interconnected circulating fluidized beds: a carbonator and a calciner (see Figure 11).

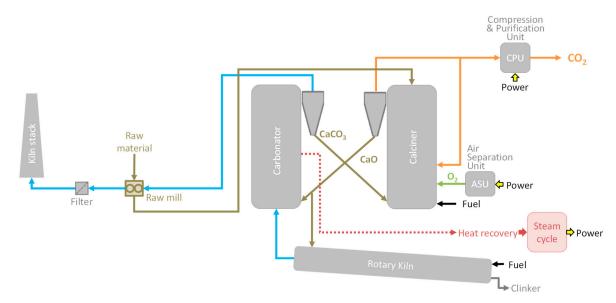$$CaO + CO_2 \rightleftharpoons CaCO_3 \qquad (8)$$

Figure 11. Simplified diagram flow of a CaL process installed in a cement plant: tail-end configuration (adapted from [46]).

In the carbonator, CaO is put into contact with the flue gas containing CO_2 at 600–700 °C. The $CaCO_3$ formed is then sent to the calcination reactor, where it is heated up to 890–930 °C, to recover the CO_2 product and to regenerate the CaO, which is sent back to the carbonator reactor [62]. In order to obtain a pure stream of CO_2, it is necessary to use oxyfuel combustion in the calciner, which has the drawback of the requirement of a cryogenic air separation unit (ASU), which significantly increases the CAPEX and OPEX. An emerging alternative is to heat the calciner indirectly, avoiding the need of an ASU. The outlet CO_2 enriched stream is sent to a compression and purification unit (CPU) where it can reach purities larger than 95% [63]. The sorbent tends to sinter and lose CO_2 capture capacity over cycles, so it is necessary to add a make-up stream of fresh $CaCO_3$ and to extract a purge stream rich in CaO to avoid the build-up of inert species [63]. However, the latter is not a problem if the CaL process is integrated in a cement plant that can make use of such purge as a feedstock for cement production (see Figure 11). This technology can offer capture rates up to 98% [64]. One of the inherent advantages of this technology is that a large part of the energy introduced into the calciner can be recovered as high temperature heat (≈650 °C) in the cooled carbonator to produce electricity with high efficiency [20,63].

There are two CaL configurations that could be implemented in the cement industry: the tail-end configuration (post-combustion technology), which has already been shown in Figure 11, and the integrated configuration, which is shown in Figure 12. In the integrated configuration, the calcination is integrated in the calciner of the cement kiln, which leads to a higher energy efficiency. Due to the small particle size required for the cement kiln, the integrated calciner is an entrained flow reactor [46]. Fuel consumption is larger in the tail-end option, although the high temperature heat can be recovered in a steam cycle to generate power. The specific energy consumption is larger for the tail-end technology: the SPECCA of a tail end configuration with 20% integration is 4.42 GJ/t CO_2 (4.07 GJ/t CO_2 for 50% integration), whereas for the fully integrated configuration the SPECCA is reduced to 3.17 GJ/t CO_2 [65]. The estimated cost related to the retrofitting of tail-end and integrated CaL to cement plants is 52.4 and 58.6 €/t CO_2 avoided, respectively [37].

Figure 12. Simplified diagram flow of a CaL process installed in a cement plant: integrated configuration (adapted from [46]).
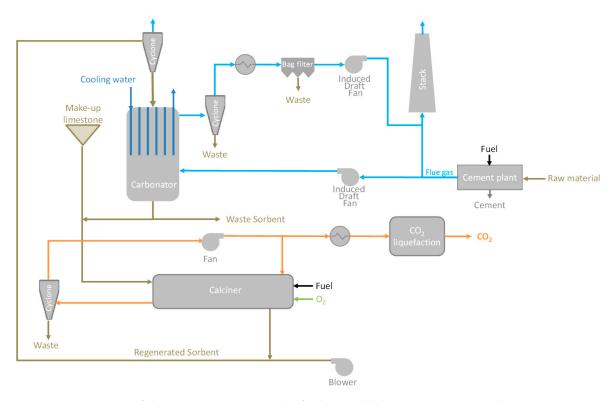
Industrial Technology Research Institute (ITRI) in cooperation with the Taiwan Cement Company (TCC) installed in 2013 a pilot CaL facility in the TCC's Ho Ping cement plant in Hualien, Taiwan, to demonstrate its High Efficiency Calcium Looping Technology (HECLOT). The pilot facility, which simplified flowsheet is shown in Figure 13, mainly consists on a bubbling fluidized bed carbonator with a diameter of 3.3 m and a height of 4.2 m, whose temperature was controlled by means of 36 water-cooled double steel jackets suspended from its top, and a rotary kiln calciner, with a diameter of 0.9 m and a length of 5 m. The pilot successfully captured 1 t CO_2/h from a slipstream of 3.1 t/h of flue gas with 20–25% CO_2 (equivalent to 1.9 MW_{th} out of 300 MW_{th} of the whole plant), with capture rates above 85% [66]. This is the world largest CaL pilot plant. In 2014, ITRI received an R&D 100 Award for this technology breakthrough in the environmental technologies category. According to ITRI, the estimated cost of the technology is less than \$30/t CO_2 [67]. The design of cascade-cyclone can increase the mixing efficiency of limestone and hot stream to increase the calcination efficiency above 90%. Oxyfuel combustion cyclone calciner with flue gas recirculation can reduce energy consumption and produce CO_2 purities above 90%. Integration of steam hydration can increase CO_2 capture efficiency and sorbent carbonation conversion, reducing sorbent consumption, recirculation rate, and thus the volume of the reactors and the energy consumption. The economic and feasible scale is considered to be a 50 MW demonstration plant [68]. TCC aims to reach zero emissions at its industrial facilities, and is developing HECLOT technology at the Ho Ping Cement plant not only to capture CO_2 but to reduce the cost of cement production. It has committed US \$19.1 M to expand the technology, and it estimates that by 2025 it will capture 0.45 Mt CO_2/y [69].

GE's RCC technology was evaluated within the Norcem CO_2 capture project (2013–2017). Pilot testing was conducted at the Institute of Combustion and Power Plant Technology at the University of Stuttgart (IFK), in Germany, under representative operation conditions focusing on the validation of process models [70]. The end of the pipe benchmarking carried out for Brevik, indicated a SPECCA of 3.13 GJ/t CO_2 avoided. Oxyfuel combustion RCC integration options provide attainable solutions for efficiently capturing CO_2 form cement plants, with lower specific energy consumption (SPECCA of 1.45 GJ/t CO_2 avoided). Emerging indirectly heated RCC concepts provide prospects to lower the overall cost by the ASU elimination [71].

Within CEMCAP project (2015–2018), the tail-end CaL (fluidized bed) was evaluated at two existing pilot facilities: the 200 kW_{th} rig at IFK, and the 30 kW_{th} pilot at INCAR-CSIC, in Spain [72]. CO_2 capture efficiencies up to 98% were attained. Testing involved the evaluation of different operation

conditions, such as high CO_2 concentration (up to 33 vol %; wet basis), high limestone make-up ratios, looping ratios up to 20 mol CaO/mol CO_2, and carbonator temperatures between 600 and 710 °C. Tail-end CaL was demonstrated under industrially relevant conditions, and is considered to be ready for the demonstration at larger scale in the cement industry [73].

Figure 13. Set-up of the 1.9 MW$_{th}$ HECLOT pilot facility installed at Ho Ping cement plant in Taiwan (adapted from [66]).

The integrated CaL configuration, or entrained flow CaL, is a promising option, although with a lesser degree of maturity than that of the tail-end CaL configuration [48]. This technology is being further developed within the European funded *CLEAN clinKER* production by calcium looping process (CLEANKER) project (2017–2021), which aims at demonstrating the entrained flow CaL technology at TRL 7 through the design, construction and operation of a pilot system in the Buzzi Unicem's cement plant in Vernasca, Italy. The pilot plant includes an entrained-flow carbonator with a 250 mm diameter in the up-flow section and 350 mm diameter in the downstream section, which treats a slipstream of approximately 1000 Nm^3/h of the kiln flue gas, and an entrained-flow oxyfuel calciner, connected to the kiln. The carbonator operates as an adiabatic reactor with an inlet adiabatic mixing temperature of 600 °C. The temperature rise due to the exothermic carbonation reaction is mitigated by heat losses and by air in-leakages. The pilot plant uses as sorbent the same raw meal that is used for cement production [74]. The project intends to demonstrate that CaL is an optimum alternative for CCUS in cement industry, with CO_2 capture efficiencies larger than 90%, ensuring that the clinker quality is not affected, and setting the basis for the industrial exploitation of the technology [75,76]. The two first years of the project were devoted to the detailed design and construction of the CaL demonstration system, and to the characterization of raw meals. Up to the project closure in September 2021, efforts are focused in the experimental campaigns, analysis of results, evaluation of the scale-up of the technology, economic analysis, and the life cycle assessment of the integrated system. The project also comprises mineralization tests with the captured CO_2 [74].

2.2. State of the Art of Oxyfuel Combustion CO$_2$ Capture Technologies in the Cement Sector

The configuration of an oxyfuel combustion cement plant is shown in Figure 14. Oxyfuel combustion refers to the combustion of the fuel in oxygen, instead of air. To control the temperature in the kiln, part of the flue gas must be recirculated (see Figure 14). As a consequence, both the material conversion in the kiln and the operational specifications of the overall process differ from those of conventional kilns [77]. The kiln exhaust would be mainly composed of CO$_2$ and steam, which can be easily knocked out by condensation. The resulting CO$_2$ stream (up to 80% mol CO$_2$, dry basis) would only need to be purified in a relatively simple cryogenic CPU unit to achieve 95% CO$_2$. Higher purities could be achieved by cryogenic distillation [4].
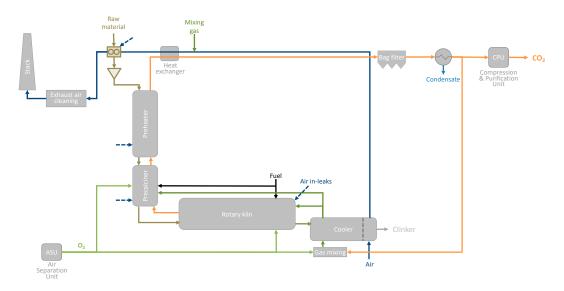

Figure 14. Configuration of an oxyfuel cement plant (adapted from [77]).

Oxyfuel combustion improves fuel efficiency, and provides a relatively low cost option for CO$_2$ abatement in cement plants compared to other technologies. Nevertheless, this method implies re-building and re-engineering the cement plant in order to minimize air ingress and optimize the heat recovery system. The main economic and energetic penalties arise from the need of an ASU, and the need to develop new kilns adapted to oxyfuel combustion conditions [78].

There is experience of oxygen enrichment in cement plants to improve throughput, or to enable the use of alternative fuels, but not for CO$_2$ abatement, and not at high levels of O$_2$ enrichment. Moreover, oxyfuel combustion capture has not been demonstrated yet at commercial scale in other sectors, and has an earlier stage of development than post-combustion capture technologies [4].

The European Cement Research Academy (ECRA) has been conducting research on oxyfuel combustion carbon capture since 2007. ECRA's CCS project phases I, II, and III have been completed, and phase IV is underway [79]. This might include the demonstration of oxyfuel technology at industrial scale in two European cement plants: Heilderberg Cement's plant in Colleferro, Italy, and Lafarge Holcim's plant in Retznei, Austria, which was announced in 2018 [80]. The project aims to advance the technology to TRL 7-8.

CEMCAP project (2015–2018), involved oxyfuel testing: a prototype that used CO$_2$ for cooling the clinker was manufactured by IKN and evaluated at a Heidelberg Cement's plant in Hannover; and an oxyfuel burner prototype, POLFLAME, was constructed by Thyssenkrupp based on a commercial kiln burner design. An existing oxyfuel combustion facility at IFK was also modified to entail testing under representative conditions of cement manufacturing. The results showed that the clinker quality and the cement strength were not affected by the oxyfuel operating conditions [48], advancing the technology to TRL 6. The estimated cost for the retrofit of oxyfuel combustion technology to cement plants is 42.4 €/t CO$_2$ avoided [37].

2.3. State of the Art of Direct CO₂ Capture Technologies in the Cement Sector

Low Emissions Intensity Lime and Cement (LEILAC), is a European funded project running between 2016 and 2020, which aims at demonstrating direct capture of process emissions at the lime and cement industries. A 60 m high pilot plant was built at Heidelberg Cement's plant in Lixhe, Belgium, with a capacity of 25,000 t CO_2/y (240 t/d of raw meal feed for cement production, or 190 t/d ground limestone), which is the equivalent to 5% of the factory's total CO_2 emissions. It makes use of Calix's technology, in which the conventional calciner is re-engineered to indirectly heat the limestone, using a direct separator reactor (DSR), which is a special steel tube that acts as a large heat exchanger (see Figure 15). In the pilot, natural gas is burned along the external furnace, heating the exterior of the DSR up to approximately 1000 °C. The raw material is dropped in the interior of the DSR, from its top, and falls slowly down, being heated by both conductive and radiative heat transfer from the reactor wall, causing it to calcine, releasing CO_2. At the base, the solids and gases are separated [81]. This way, the process emissions arising from the limestone calcination are isolated from those resulting from fuel combustion, which can be addressed using alternative fuels or with other CCUS technologies. The main advantage of the technology is its low energy penalty, which is only related with heat losses. Moreover, according to the developers of the technology, the capital cost is comparable to conventional equipment. On the other hand, its main challenge is the high temperature required for limestone calcination, which is significantly above than that of other applications of Calix's direct CO_2 separation technology. During pilot testing, special attention is being addressed to ensure that enough heat is put into the DSR and that the heat is placed at the right places. The capture rate is being evaluated from the difference in the CO_2 content of the powder before and after the DSR. Early results from the pilot plant are promising, although testing will be extended until the end of 2020 to gradually increase operational throughputs, temperatures and evaluate materials durability. LEILAC's has advanced the technology to a TRL of 6–7.
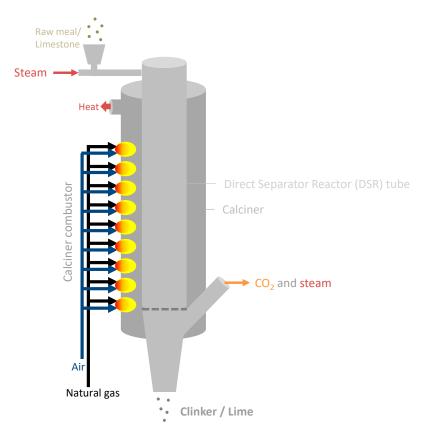

Figure 15. Scheme of LEILAC's direct separator reactor (adapted from [81]).

Based on the success of LEILAC project, LEILAC 2 was launched on 7 April 2020, and will run to the end of 2024. Central aspects of the LEILAC2 project are the further scale-up of the technology to an industrial level, the full process integration into an existing cement plant, and the heat supply by means of renewable energy to achieve carbon neutral CO_2 separation [14]. A CO_2 separation plant with capacity to capture 100,000 t CO_2/y (four-fold scale-up of LEILAC's pilot), which represents approximately 20% of a cement plant's process CO_2 emissions, or 100% of a large lime kiln's process emissions, will be built in a HeidelbergCement's plant in Western Europe, although the actual location is yet to be determined [82].

2.4. Comparison of CO_2 Capture Technologies Evaluated or Planned in the Cement Sector

Table 1 summarizes relevant information of the CO_2 capture technologies that have been evaluated in the cement sector, or those that are under planning stage. As can be seen from the table, the activity is concentrated in the last 7 years.

Caution is needed when comparing energy consumption and cost between technologies, due to the different assumptions of the technology developers. Costs depend strongly on factors such as the geographical location, the carbon tax, the steam source, the electricity mix, the electricity price, the fuel price, and the plant specific characteristics. To choose the best suited technology for a specific plant, specific technical and economic evaluations need to be performed. However, the efforts of certain projects, like Norcem CO_2 capture project and CEMCAP, which have compared different capture technologies based on a common basis, have provided relatively fair comparisons.

As shown in Table 1, within CEMCAP project, oxyfuel combustion CO_2 capture was found to provide the lowest cost per CO_2 avoided, followed by CaL, CAP, and MAL. Oxyfuel combustion CO_2 capture also showed the lowest specific primary energy consumption per CO_2 avoided (SPECCA), followed by integrated CaL, MAL, CAP, and tail-end CaL.

The lowest cost of CO_2 capture reported in Table 1 corresponds to HECLOT technology, with \$30/t CO_2. However, this value, which was taken from an ITRI's publication, has not been peer reviewed and must be taken with caution.

The second lowest cost reported in Table 1 corresponds to the CDRMax® technology. However, we must bear in mind that this cost is not site specific for the Dalmia cement's plant but a generic cost provided by Carbon Clean, and thus must also be taken with caution.

The third lowest cost shown in Table 1 corresponds to RTI solid sorbent technology, which was evaluated within Norcem CO_2 capture project.

The cost of Svante's technology is also amongst the lowest shown in Table 1. This has been taken from a public declaration of Svante's CEO regarding the commercial Lafarge Holcim cement CO_2 capture project, which has not been peer review, and thus must also be taken with caution.

However, cost is not the only factor to consider for the retrofit of a cement plant with CO_2 capture: technology maturity, possible effects over product quality, space requirements, and the need for utilities are important factors that need to be considered.

To date, chemical absorption with liquid solvents have reached the largest demonstration scale in the cement sector, with the SkyMine process at the front, with 75,000 t CO_2/y, followed by amine-based Anhui Conch's project, with 50,000 t CO_2/y. However, no operational performance data of those facilities are publicly available. On the other hand, amine absorption has reached commercial scale demonstration in the industry and power sector, and hence is considered the most mature technology. In the Longship Project, this has swayed the argument towards Aker Solution's ACC^TM technology over RTI solid sorbent technology, despite the lower energy consumption of the later shown within Norcem CO_2 capture project (see Table 1).

Figure 16 summarizes the current TRL of CO_2 capture technologies in the cement sector. On one end we find membranes, with the lowest TRL, 4, and in the opposite end, chemical absorption, with TRL 8. In between, the trio formed by adsorption, oxyfuel, and direct capture, with TRL 6, and CaL, with TRL 7.

Table 1. Summary of carbon capture technologies evaluated and planned in the cement sector
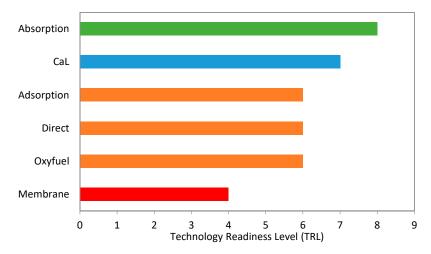
Technology	Project	Test Location	Operation	Scale of Field Test	Type	Energy Consumption	Cost
SkyMine®	SkyMine® Beneficial CO_2 Use Project	Capitol Aggregates' San Antonio cement plant, Texas, United States of America	2015–	76,488 t CO_2/y	CCU	8.3 GJ_e/t CO_2 [23]	$125 M [a] [23]
Aker Solution's ACC[TM]	Norcem CO_2 capture project	Norcem's cement plant, Brevik, Norway	2014–2015	450 Nm^3/h kiln flue gas 140–250 kg CO_2/h	capture	3.0 GJ/t CO_2 [b,c,d] 2.7 GJ/t CO_2 [b,d] 0 GJ/t CO_2 [b] [36]	NA
Aker Solution's ACC[TM]	Norway's full chain CCS	Norcem's cement plant, Brevik, Norway	2023–	400,000 t CO_2/y	CCS	NA	NA
Amine scrubbing	Anhui Conch Cement project	Anhui Conch's cement plant in Wuhu, China	2018–	50,000 t CO_2/y	CCU	NA	$10 M
CDRMax®	Dalmia Cement project	Dalmia cement's plant in Tamil Nadu, India	NA	500,000 t CO_2/y	CCU	NA	$40/t CO_2 [44]
CAP	CEMCAP	GE's Technology Center in Växjö, Sweden	2015–2018	-	Individual components tested: absorber, DCC-scrubber and water wash	2.37 GJ/t CO_2 [b] [47] 3.75 GJ/t CO_2 [e] [46]	66.2 €/t CO_2 [37]
Membranes	Norcem CO_2 capture project	Norcem's cement plant, Brevik, Norway	2014	NA	capture	NA	NA
Membranes	MemCCC project	Norcem's cement plant, Brevik, Norway	2016	37 Nm^3/h kiln flue gas	capture	1.20 GJ_e/t CO_2 [51]	47–48 €/t CO_2 [d] [51]
MAL	CEMCAP	TNO, Eindhoven, Netherlands and SINTEF Energy Research, Trondheim, Norway	2015–2018	-	Individual components tested separately: membranes and liquefaction	3.22 GJ_e/t CO_2 [e] [46]	83.5 €/t CO_2 [37]
RTI solid sorbent technology	Norcem CO_2 capture project	Norcem's cement plant, Brevik, Norway	2016	NA	capture	2.4 GJ/t CO_2 [b,c,d] [56]	45.8 €/t CO_2 [c,d] 40.7 €/t CO_2 [d] 38.6 €/t CO_2 [56]
Veloxotherm[TM]	LafargeHolcim Cement Carbon Capture	Holcim Portland cement plant, Colorado, United States of America	2023–	2,000,000 t CO_2/y	CCUS (EOR)	NA	$150 M $50/t CO_2 [60]

Table 1. *Cont.*

Technology	Project	Test Location	Operation	Scale of Field Test	Type	Energy Consumption	Cost
Veloxotherm[TM]	CO2MENT	Lafarge's Richmond British Columbia cement plant, Canada	2019–2022	1 t CO_2/d	CCU	NA	$28 M
HECLOT	ITRI's Calcium Looping Pilot	Ho Ping Cement plant, Taiwan	2013–	1 t CO_2/h	capture	NA	$30/t CO_2 [67]
RCC	Norcem CO_2 capture project	Institute of Combustion and Power Plant Technology at the University of Stuttgart (IFK), Germany	2013–2017	-	-	3.13 GJ/t CO_2 [e,h] 1.45 GJ/t CO_2 [e,i] [71]	NA
CaL	CEMCAP	IFK, Stuttgart University, Germany INCAR-CSIC, Spain	2015–2018	-	-	4.42 GJ/t CO_2 [e,f,h] 4.07 GJ/t CO_2 [e,g,h] 3.17 GJ/t CO_2 [e,i] [65]	52.4 €/t CO_2 [h] 58.6 €/t CO_2 [i] [37]
Entrained CaL	CLEANKER	Buzzi Unicem Vernasca cement plant, Italy	2019–2021	1000 Nm^3/h kiln flue gas	capture and mineralization testing	NA	NA
Oxyfuel combustion	CEMCAP	HeidelbergCement plant, Hannover	2015–2018	-	oxyfuel prototype testing: clinker cooler and burner	1.63 GJ/t CO_2 [e]	42.4 €/t CO_2 [37]
Oxyfuel combustion	ECRA's CCS project	HeidelbergCement plant in Coleferro, Italy Lafarge Holcim plant in Retznei, Austria	NA	NA	NA	NA	80 M€ [80]
Direct capture	LEILAC	Lixhe, Belgium	2020	25,000 t CO_2/y	NA	NA	NA
Direct capture	LEILAC 2	Heidelberg cement plant in Western Europe	2020–2024	100,000 t CO_2/y	NA	NA	NA

[a] Installed costs; [b] Specific reboiler duty (SRD); [c] Without heat recovery; [d] 85% capture rate; [e] Specific primary energy consumption per CO_2 avoided (SPECCA); [f] 20% integration; [g] 50% integration; [h] tail-end; [i] integrated; NA: Not available.

Figure 16. Current technology readiness level of CO_2 capture technologies in the cement sector (adapted from [83]).

3. Future Challenges and Prospects for CO_2 Capture in the Cement Industry

CO_2 capture is a necessity in the cement sector, where up to 65% of CO_2 emissions come from the manufacturing process, and thus cannot be avoided by other means. The Faster Innovation Case (FIC) recently published by the IEA, which is consistent with reaching net-zero emissions in 2050, would see 5 CCUS facilities of 1 Mt/y each month in the cement sector through 2050. Moreover: the increasing share of bioenergy in the fuel mix of cement kilns, which does not require significant equipment retrofits, if combined with CCUS, could lead to net CO_2 removals from the atmosphere, thus contributing to the achievement of net-zero emission across the entire economy [84].

CO_2 capture in the cement sector is now close to commercial demonstration. There are four large scale projects at different stages of development: Norway's Longship Project, which will make use of amine-based Aker solutions' ACC^{TM} technology to capture 0.4 Mt CO_2/y by 2023; Dalmia cement project, which will make use of another amine scrubbing technology, CDRMAX$^®$, to capture 0.5 Mt CO_2/y; LafargeHolcim cement carbon capture, which would make use of the Svante's adsorption-based Veloxotherm^{TM} process to capture up to 2 Mt CO_2/y; and TCC is developing HECLOT technology to capture 0.45 M tCO_2 by 2025. Although at a lower scale, LEILAC 2 will also demonstrate direct CO_2 capture at the significant scale of 0.1 Mt CO_2/y by 2024.

While direct CO_2 capture and oxycalcination are promising technologies that have great potential to be implemented in new cement plants, the prospects for their use in the retrofitting of existing plants is less likely. Given the existing overcapacity, and the age and life ahead of existing facilities, the construction of new cement plants integrated with CCUS at a relevant scale for climate mitigation seems unlikely.

Post-combustion CO_2 capture technologies are the preferred option to retrofit existing facilities, as the CO_2 is captured from the exhaust gas of the cement plant, thus not affecting the existing cement production process. Among the available post-combustion technologies, chemical absorption with liquid solvents is the most mature technology: to date, it has reached the largest demonstration scale at cement plants, and provide the least risky pathway for the retrofitting of existing facilities. Moreover, solvent and process development are expected to lead to further cost reductions as the technology deploys in the cement sector, as it has previously occurred in the power sector. In fact, this is the technology selected by two of the commercial-scale projects in the pipeline. On the other hand, although with a lesser technology readiness level, solid sorbents based post-combustion CO_2 capture processes also show great promise. Svante's adsorption and HECLOT technologies are already on commercial scale development.

Further research and development is urgently needed in order to reduce the cost of the capture step and to increase the technology readiness level of emerging technologies to make CCUS an economically viable and safer option for cement producers in the forthcoming CO_2 neutral economy.

Abbreviations

2DS	IEA's 2 °C scenario
ACC^{TM}	Aker Solution's Advanced Carbon Capture technology
AIPOs	Alumino-phosphates
ASU	Air separation unit
ASTR	Automated sorbent test rig
B2DS	IEA's Beyond 2 °C scenario
CaL	Calcium looping
CAP	Chilled Ammonia Process
CAPEX	Capital expenditure
CCP	Carbon Capture Project
CCS	CO_2 capture and storage
CCU	CO_2 capture and utilization
CCUS	CO_2 capture, utilization and storage
CHP	Combined heat and power facility
CLEANKER	Clean clinker production by calcium looping process
CPU	Compression and purification unit
CSA	Concentration swing adsorption
DCC	Direct contact cooler
DOE-NETL	United States Department of Energy's National Energy Technology Laboratory
CDRMax$^{®}$	Carbon Clean's amine-based CO_2 capture technology
CEMBUREAU	The European Cement Association
DEA	Diethanolamine
DSR	Direct separator reactor
ECRA	European Cement Research Academy
EHR	Excess heat recovery
EOR	Enhanced oil recovery
ESP	Electrostatic precipitator
FGD	Flue gas desulfurization
FSC	Fixed-site carrier
GCCA	Global Cement and Concrete Association
IEA	International Energy Agency
IFK	Institute of Combustion and Power Plant Technology at the University of Stuttgart
ITRI	Industrial Technology Research Institute
HECLOT	High Efficiency Calcium Looping Technology
LEILAC	Low Emissions Intensity Lime and Cement
MAL	Membrane-assisted liquefaction
MEA	Monoethanolamine
MOFs	Metal organic frameworks
MTU	Mobile test unit
NDC	National determined contributions
NTNU	Norwegian University of Science and Technology

OPEX	Operational expenditures		
PSA	Pressure swing adsorption		
PVAm	Polyvinylamine		
RCC	GE's Regenerative Calcium Cycle		
RTI	Research Triangle Institute		
RTS	IEA's Reference Technology Scenario		
SAPOs	Alumino-silico-phosphates		
SFMB	Staged fluidized moving bed		
SNCR	Selective non catalytic reduction		
SPECCA	Specific primary energy consumption for CO_2 avoided		
TCC	Taiwan Cement Company		
TRL	Technology readiness level		
TSA	Temperature swing adsorption		
VSA	Vacuum swing adsorption		
VTCSA	Vacuum, temperature, and concentration swing adsorption		

Symbols and Colors Notation

Symbol	Name	Line	Name
	Baghouse filter		Air
	Coalescing filter		Air in-leaks
	Cooler		Clinker/cement
	Centrifugal pump		CO_2 product
	Compressor		Flue gas
	Cyclone		Fuel
	Filter		Heat
	Heater		Heat recovery
	Heat exchanger		Other process streams
	Fan/blower		Power
	Mill		Raw meal/feedstock
	Reboiler		Solvent/sorbent
	Rotary kiln		Steam
	Silo		Water
	Stack		
	Tower/vessel		
	Vacuum pump		

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