

## Review article

# A review on CO<sub>2</sub> capture and sequestration in the construction industry: Emerging approaches and commercialised technologies

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## ABSTRACT

Reducing CO<sub>2</sub> emissions from the construction industry is the most imperative factor in the fight against climate change, which aims to reduce the average atmosphere temperature below 1.5 °C by the end of this century. Globally, the cement industry is responsible for approximately ~7 % of CO<sub>2</sub> emissions. About 9.95 Gt/y of CO<sub>2</sub> was emitted by the construction sector as of 2019, making it the highest contributor. The construction sector is forecast to reduce its CO<sub>2</sub> emissions by 16 % by 2030, leading to net-zero emissions by 2050. Thus, several measures have been implemented to mitigate significant CO<sub>2</sub> emissions from the construction industry by capturing and utilizing CO<sub>2</sub>. This paper reviews existing industrial-level CO<sub>2</sub> capture technologies in cement industries, such as amine scrubbing, oxy-combustion, direct capture, and calcium looping, as well as the costs and barriers associated with their use. Also presented a summary and comparison of utilizing CO<sub>2</sub> through accelerated mineral carbonation in cement-based materials, recycled aggregate, and calcium-rich solid waste. In addition to this, various commercialized technologies for mitigating CO<sub>2</sub> emissions (Carbon8, Calera Corporation, CarbonCure, Solidia, Blue Planet and Carbstone) and their methods of sequestering CO<sub>2</sub> as well as their technology readiness levels (TRL), %CO<sub>2</sub> uptake, and patent analysis for their technologies were discussed.

## 1. Introduction

Over the years, change in climate conditions has been one of the most significant concerns, as it affects the economic, social and environmental aspects of our lives. The increase in greenhouse gases (GHGs) in the atmosphere results from several human activities, contributing to a significant rise in the earth's temperature and thus threatening life on the planet [1,2]. Among various GHGs, viz., CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, etc., the major influencing gas is CO<sub>2</sub>, which is considered the major cause of global warming. Absorption of Infrared radiation by CO<sub>2</sub> gas causes its deformation and stretching-vibration force resulting in a warming effect [3]. Anthropogenic activities have contributed to global warming of 1.0 °C over pre-industrial levels. By 2030, it is likely to reach 1.5 °C if current CO<sub>2</sub> emissions levels continue [4].

Climate change was addressed at the United Nations Conference on Environment and Development (UNCED) in Rio de Janeiro, Brazil, in 1992 [5]. Under the United Nations Climate Change Secretariat (UNCCS 2009), increased CO<sub>2</sub> levels result in changes in climate indicators such as rainfall, heat, rise in temperatures and acidification of oceans [6]. The Paris Agreement in 2016 marked the historical transformation of global

climate change, as the world leaders from 195 nations agreed to combat climate change and its harmful effects [7]. According to the Paris Agreement, global warming should be reduced to 2 °C below pre-industrial levels by the end of the century [4]. During the Paris Agreement, India committed to reducing CO<sub>2</sub> emissions by 30–35 % by 2030 and creating a carbon sink of 2.5–3 billion tons of CO<sub>2</sub> equivalent by 2030 through additional forest cover [8,9]. At the Katowice Climate Summit (COP-24), one of the milestones in completing the Paris Agreement was approving the Paris Agreement Rulebook (Katowice Rulebook) [10]. To achieve the 1.5 °C targets by the end of the century, the International Panel on Climate Change (IPCC) recommends that CO<sub>2</sub> emissions should be maintained at 25–30 Gt CO<sub>2</sub> per year instead of the existing 52–58 Gt CO<sub>2</sub> per year [11]. A recent agreement on climate change COP-26 was held in Glasgow, the UK in November 2021 to review and discuss the status [12].

Globally, three main ways to mitigate CO<sub>2</sub> emissions are discussed in the literature.

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1. Reducing CO<sub>2</sub> emissions by using renewable energy, switching to cleaner fuels, improving efficiency, using nuclear power, and using carbon capture and storage techniques is possible [13–15].
2. CO<sub>2</sub> mitigation methods or negative emissions technologies will be deployed in the future to capture and sequester CO<sub>2</sub> from the atmosphere [6].
3. Thirdly, solar and terrestrial radiation management can be used to alter the earth's radiation balance [16,17].

The current study reviewed CO<sub>2</sub> capture and utilization in construction industries. This paper examines recent CO<sub>2</sub> capture and utilization developments, which concerns about global warming have driven. In the first part of the paper, we review the different capture methods of CO<sub>2</sub> in cement production, such as amine scrubbing, calcium looping, direct separation, and oxy combustion. We also discuss commercialized technologies involved in CO<sub>2</sub> capture and their challenges. In the second section, we discussed the CO<sub>2</sub> utilized in cement-based materials, recycled aggregate (RA) and solid wastes by ACT (accelerated carbonation technology). Later, we discussed the different commercially available technologies for CO<sub>2</sub> utilization in construction industries, including Carbon8, Calera, CarbonCure, Solidia, Blue Planet, and Carbstone. Further, patents published by these commercialized technologies were also reviewed.

## 2. CO<sub>2</sub> capture in the cement industry

The cement industry plays a significant role in CO<sub>2</sub> emission, and it emits 0.5–0.6 tons of CO<sub>2</sub> per ton of cement production, which is ~7 % of the total CO<sub>2</sub> emission in the world [10,18–22]. At the beginning of cement production at a large scale, studies were carried out to improve energy efficiency, fuels reduction and optimize cost, however later on, due to environmental threats and global warming issues, cement industries were allocated to devote budget and time to mitigate the pollution strategies [23]. Approximately 576 million tons of CO<sub>2</sub>/year were emitted from the global cement industry in 1990 and increased to 1.4 billion tons in 2002. Within 24 years, CO<sub>2</sub> emissions increased almost three times and amounted to ~2.9 billion tons of CO<sub>2</sub> per year in 2021 [24–26]. Three sources mainly generate CO<sub>2</sub> during cement manufacturing [27,28].

1. 50 % results from calcination (decomposition of CaCO<sub>3</sub> to CaO and CO<sub>2</sub>) [29,30].
2. 40 % from the fuel combustion in the kiln (coal, waste, sewage sludge, etc.)

3. 10 % from the manufacturing operations include transportation and electricity.

Globally CO<sub>2</sub> emissions are estimated to be 35 % from China and India in 2020, while only 4 % are from the UK, Germany, and France combined [31]. The countries with the highest contribution of CO<sub>2</sub> from 2005 to 2021 are China, India, Europe, and the USA (Fig. 1) [31]. During COVID, the cement sector's CO<sub>2</sub> emissions did not grow but did not decline as much as oil, gas, and coal [26]. By applying the best strategies like alternative fuels, energy efficiency, clinker substitution and CCU, CO<sub>2</sub> emissions from cement industries can be reduced to 1.5 billion tons per year and 0.43-tons CO<sub>2</sub> per ton of cement production by 2050 (Fig. 2) [32]. Among global cement industries, Heidelberg Cement has pledged to mitigate CO<sub>2</sub> emissions by 25–30 % by 2030 compared to 1999 and was the first company to be approved by science-based CO<sub>2</sub> capture [33]. CO<sub>2</sub> captured in the cement industry by different technologies were distinguished, such as amine scrubbing, calcium looping, oxyfuel combustion and direct separation are summarized [34] and the pre-combustion process is not well suitable for CO<sub>2</sub> capture in the cement industry as they are suitable for the energy-related CO<sub>2</sub> emission source [35].

### 2.1. Amine scrubbing

The cement industry has used amine scrubbing technology since 1930 [36]. The process involves removing CO<sub>2</sub> from flue gases after combustion or before vents into the atmosphere. An amine solvent is used in the absorber column at 50 °C to produce a decarbonized gas stream, which is further regenerated by heating the solvent at around 120 °C in the stripper column, where a pure CO<sub>2</sub> gas is recovered [37]. The capturing method involves wet scrubbing with amine solvents such as monoethanolamine (MEA) and diethanolamine (DEA), as the rate of reaction of amine solvent is very high and lower cost [38]. During the process, a carbamate ion and a protonated amine were formed by the reaction of MEA and CO<sub>2</sub>, which further reacted with H<sub>2</sub>O molecules to form a bicarbonate ion (Fig. 3) [39].

Among the widely used amines in amine scrubbing technology, MEA is considered a primary amine, DEA is considered a secondary amine, and MDEA (methyldiethanolamine) is considered a tertiary amine [40]. According to Sharif et al., 2020 [41], the absorption of CO<sub>2</sub> by amine solvents is characterized by high CO<sub>2</sub> solubility, lower regeneration energy for solvent regeneration, and fast kinetic rates. Singh and Versterg 2008 [42] investigated amine activity performance and reported that amine solvent activity is highly dependent on chain lengths, alkyl

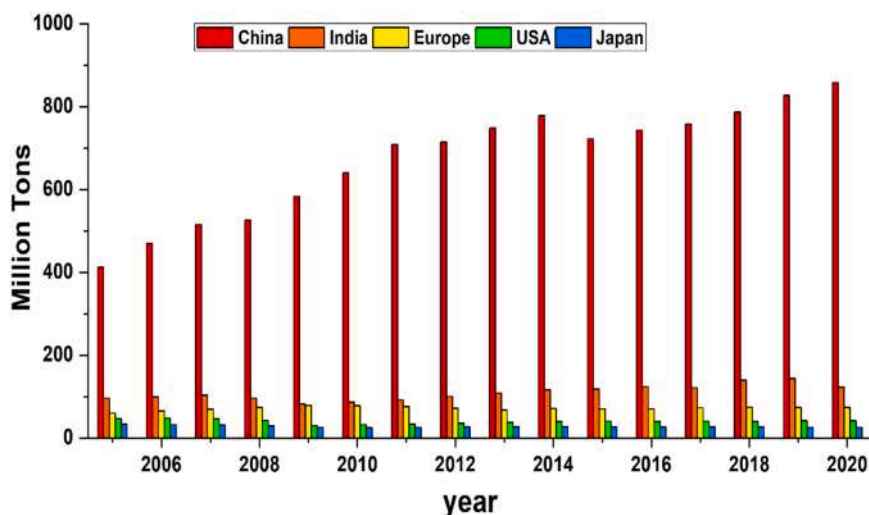


Fig. 1. Top countries with the highest CO<sub>2</sub> emission annually from cement industry (2005–2019) [31].

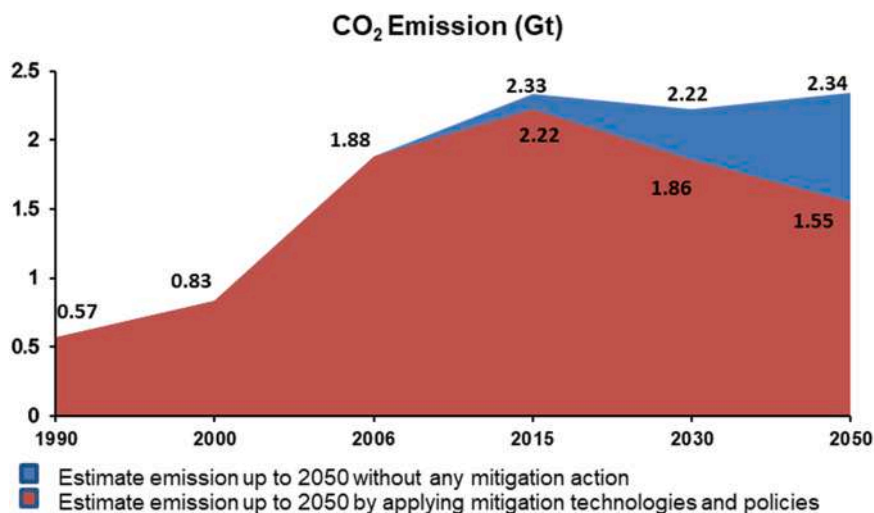


Fig. 2. Globally CO<sub>2</sub> emission from 1990 to 2050 with and without any mitigation strategies [32].

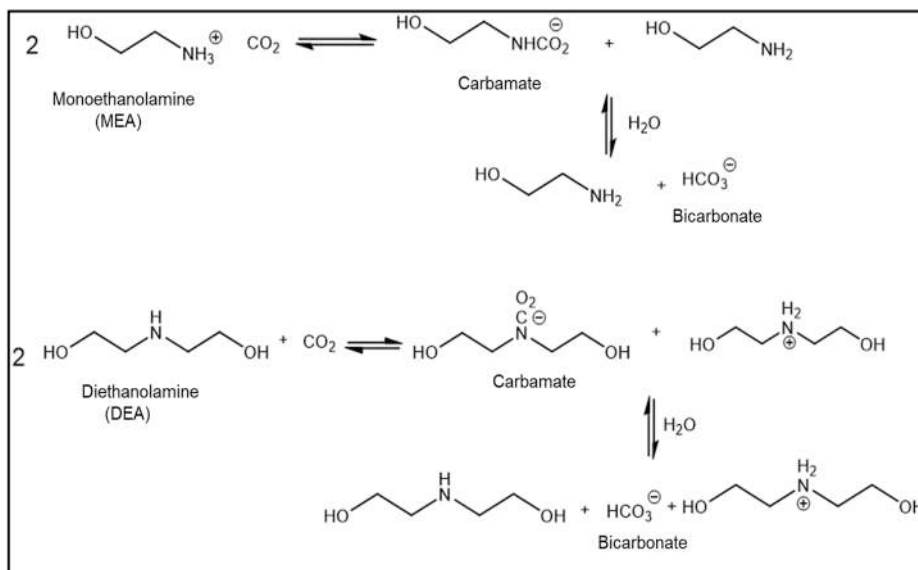


Fig. 3. Mechanism of CO<sub>2</sub> capture in aqueous amine solution (MEA & DEA) [39].

group position, and abundance of functional groups. Budzianowski 2016a [43] found that primary and secondary amine solvents' regeneration energy is very high compared to tertiary amine.

Amine scrubbing is considered a benchmark technology for CO<sub>2</sub> capture in the cement industry. The first globally CO<sub>2</sub> capture technology based on amine scrubbing that achieved the milestone in the cement industry was Norway's Longship. Aker Solution Advanced Carbon Capture technology (ACCT<sup>M</sup>) and its S26 solvent used by Norcem's cement factory in Brevik, Norway, capture ~400,000 Mt of CO<sub>2</sub>/year. The plant can capture 40 % of its total CO<sub>2</sub> emissions using only waste heat, and its technology readiness level (TRL) is 8 [44]. In December 2020, Heidelberg Cement Group, another CO<sub>2</sub> capture technology, will construct its first full-scale CCS facility at the NORCEM cement plant in Brevik, which can capture ~400 kt CO<sub>2</sub>/year [45]. Recently, Heidelberg Cement Group announced that it would construct CCS facilities at its cement plant in Slite, Gotland Island, Sweden, which capture ~1.8 Mt CO<sub>2</sub>/year. This facility is accounted for around 3% of CO<sub>2</sub> emissions in Sweden [46]. The world's largest amine-based CCS was built at the Baimashan cement plant in late 2017 by Anhui Conch Group, Wuhu, China. The CO<sub>2</sub> capturing capacity of the plant is ~50Mt CO<sub>2</sub>/year

(Table 1) [47].

An improved Amine Promoted Buffer Solution (APBS), APBS-CDRMax®, developed by Carbon Clean Solutions Limited (CCSL), was used by different commercialized technologies. The solvent designed by APBS-CDRMax® has a high CO<sub>2</sub> absorption rate, higher capacity of CO<sub>2</sub> capture and lower regeneration energy than MEA solvent. Compared with the MEA solvent, it also has a lower corrosion rate, solvent degradation rates and operating costs are also very low [48]. Kentucky Utilities E.W. Brown Power Generation Station in Harrodsburg, Kentucky, uses the CDRmax solvent for different test conditions campaigns at the 0.7 MWe pilot scale CO<sub>2</sub> Capture. The solvent regeneration energy ranged from 2.9 to 3.3 GJ/ton CO<sub>2</sub>, which is a 30 % lower regeneration rate than MEA [49]. The Dalmia Cement Group built a large CCUS facility using Carbon Clean's technology, CDRMax®, at their cement plant sites in Tamil Nadu, India, in 2019. The facility can capture 0.5 Mt CO<sub>2</sub>/year and the estimate capturing cost is approximately \$40/t CO<sub>2</sub> (Table 1) [50].

In the recent decade, the mixture of amine solvents has solved many of the current challenges regarding the direct use of primary, secondary and tertiary amines for CO<sub>2</sub> capture. The mixed amine solution has a

**Table 1**  
Different technology and CO<sub>2</sub> captured rates in cement plants worldwide.

Project	Location	CO <sub>2</sub> capture	Ref.
<ul style="list-style-type: none"> <li>• Amine-based absorption technology.</li> <li>• The largest CO<sub>2</sub> capture plant operated in 2018 almost captured 3% of the total CO<sub>2</sub> emission from the cement industry.</li> <li>• CO<sub>2</sub> is produced by this plant with a purity of 99.9%.</li> <li>• The cost of the project was \$10 M</li> </ul>	AnhuiConch's Baimashan plant (Wuhu, China)	50,000 t CO <sub>2</sub> /year	[259,260]
<ul style="list-style-type: none"> <li>• Amine-based scrubbing technology.</li> <li>• Used of Carbon Clean CDRMax® technology combined with proprietary solvent (Amine Promoted Buffer Salts -APBS)</li> <li>• Produced CO<sub>2</sub> purity between 95% and 99.9%.</li> <li>• The project cost was \$40/t CO<sub>2</sub>.</li> <li>• Calcium looping technology</li> <li>• ITIR (Industrial Technology Research Institute) with TCC (Taiwan Cement Company) installed a calcium looping pilot in 2013</li> <li>• Demonstrate HECLOT (High-Efficiency Calcium Looping Technology)</li> <li>• The captured rate was about 85%</li> <li>• The estimated cost was \$30 t CO<sub>2</sub>.</li> <li>• Aker Solutions ACCT<sup>M</sup> and its S26 amine solvent</li> <li>• capture CO<sub>2</sub> gas emitted by the Norcem's cement factory.</li> <li>• Complete in 2023.</li> <li>• Direct capture technology -Low Emissions Intensity Lime and Cement (LEILAC)</li> <li>• European funded from 2016 to 2020 in which CO<sub>2</sub> is direct capture from the lime and cement industry.</li> <li>• Direct capture technology</li> <li>• successful of LEILAC, LEILAC-2 was launched on April 7, 2020, and ended in the year 2025</li> <li>• CEMCAP project was launched in 2015 and ended in 2018</li> <li>• CO<sub>2</sub> is used for cooling the clinker.</li> <li>• Cost of CO<sub>2</sub> capture technology in cement</li> </ul>	Dalmia Cement project (Tamil Nadu, India)	0.5 Mt CO <sub>2</sub> /year	[261,262]
	TCC's Ho Ping cement plant (Hualien, Taiwan)	1 t CO <sub>2</sub> /h	[89,263]
	Brevik, Norway	400,000 Mt CO <sub>2</sub> /year	[264] {add reference}
	Heidelberg Cement's plant (Lixhe, Belgium)	25,000 t CO <sub>2</sub> /year	
	Western Europe	100,000 t CO <sub>2</sub> /year	[109,265]
	Hannover	–	[63]

**Table 1 (continued)**

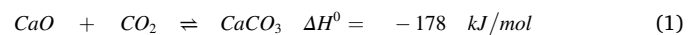
Project	Location	CO <sub>2</sub> capture	Ref.
plants is almost \$44/t CO <sub>2</sub>			
<ul style="list-style-type: none"> <li>• Oxyfuel combustion technology-ECRA.</li> <li>• Three of the four-phase were completed, and phase IV was underway.</li> <li>• The project's estimated cost was \$84.1 M.</li> </ul>	Heidelberg Cement plant (Italy) & Lafarge Holcim plant (Austria)	–	[119]

high CO<sub>2</sub> absorption capacity and lower regeneration energy during the desorption of CO<sub>2</sub>. Table 2 summarizes the recent literature on different methods of optimizing amine solvent mixtures to enhance CO<sub>2</sub> absorption efficiency and regeneration energy [51–61].

The main disadvantage of the amine scrubbing method is its higher regeneration energy, as almost 50–80% of energy is used in the solvent regeneration process of the overall CO<sub>2</sub> capture cost. The estimated cost of MEA-based scrubbing technology is around \$80/t CO<sub>2</sub> [62,63]. Various contaminants in the flue gases, including SO<sub>2</sub>, NO<sub>2</sub>, etc., poison the absorption solvent, which is crucial to the operation of the scrubber unit. SO<sub>2</sub> reacts with amine solvent forming corrosive salt. Similarly, NO<sub>2</sub> reacts with the solvent to form HNO<sub>3</sub> resulting in the degradation of the amine solvent [64]. The concentration of SO<sub>2</sub> and NO<sub>2</sub> should be limited to 10 ppmv and 20 ppmv, respectively, in the amine absorption process [65,66]. Solvent degradation can also occur in the presence of acidic components like HCl, which reduces the effectiveness of the MEA absorption process [67]. The environmental risk associated with amine solutions during CO<sub>2</sub> capture is escaping amine solutions into the atmosphere [68]. According to a study, every million tons of CO<sub>2</sub> captured with 30 % MEA aqueous solutions releases ~80 tons of MEA into the atmosphere [68]. A study by Xie et al. [69] found that atmospheric reactions of ethanolamine produce harmful isocyanic acids and cancer-causing nitrosamines. The advantages and disadvantages of amine scrubbing technology are summarized in Table 3.

## 2.2. Calcium Looping

Shimizu first proposed calcium looping technology in 1990 [70,71]. This method has a reversible reaction between CaO and CO<sub>2</sub> at high temperatures to form CaCO<sub>3</sub>. It is a regenerative process that uses CaO-based energy to effectively release CO<sub>2</sub> from combustion through carbonation-calcination reaction cycles (Eq.1).



This process comprises two interconnected reactions, i.e., carbonation and calcination. Carbonation reaction occurs in the carbonator where CO<sub>2</sub> is reacted with CaO at high temperatures (600–750°C), giving a solid CaCO<sub>3</sub>, which is an exothermic reaction (Eq. 1) [72,73]. CaCO<sub>3</sub> formed in the carbonator is sent to the calciner for calcination reaction, where CaCO<sub>3</sub> is heated up to ~950°C to obtain CO<sub>2</sub> gas and CaO. The reaction is endothermic. CaO formed in the calciner is further sent back to the carbonator for reaction (Fig. 4) [72,73]. Impurities such as NO<sub>2</sub> and SO<sub>2</sub> in the flue gas can remove easily by the CO<sub>2</sub> processing unit [74,75]. Repeated calcination and carbonation processes are observed to affect the performance of the sorbent. Sorbent deactivates due to sintering and irreversible sulphation during the processing [76–79]. As sintering becomes more severe at higher CO<sub>2</sub> partial pressures and calcination temperatures, the calcination temperature is set as low as possible to maintain sorbent regeneration [76,79,80]. In a recent study, Lisbona et al. propose using cement plants as carbon capture hubs for industrial clusters of carbon emitters by implementing calcium looping carbon capture, thereby reducing overall CO<sub>2</sub> capture costs. In the proposed work, carbonators utilize flue gases generated by different processes within the cluster as calciners. As a result of using a CO<sub>2</sub>

**Table 2**

Summary of mixed amine scrubbing technology studied by various researchers.

Solvent	Ratio	CO <sub>2</sub> removal efficiency %	Method	Temperature (°C)	Regeneration energy (GJ/tCO <sub>2</sub> )	Ref.
MEA	35 wt% MEA	85	solvent development	123.7	3.10	[51]
	30 wt% MEA	95.9	solvent development	114–130	3.1–5.4	[52]
	28 wt% MEA	80.49	modification	120–121	3.98	[53]
	30 wt% MEA	90	solvent development	136	3.99	[54]
AMP	30 wt% AMP	96.39	solvent development	95–109	2.1 (MJ/kgCO <sub>2</sub> )	[55]
MDEA + PZ	10 wt% MDEA+ 30 wt% PZ		modification	110	2.39	[56]
	20 wt% MDEA+ 10 wt% PZ	90	solvent development	95–110	3.0–4.1 (MJ/kg CO <sub>2</sub> )	[57]
	MDEA+PZ = 30–40 wt%		multi-objective optimization	100–120	2.76	[58]
1-MPZ + PZ	30 wt% 1-MPZ+ 10 wt% PZ	94	solvent development	124.7	2.988	[59]
MEA + sulfolane	20 wt% 1-MPZ+ 10 wt% PZ		solvent development	120	2.5	[60]
	4 M MEA + % 5 M sulfolane		solvent development	70	1.159	[61]
MEA+ 1-propanol	30 wt% MEA+ 40 wt% 1-propanol	90	solvent development	136	2.40	[54]
TETA +TMPDA	30 wt% MEA+ 40 wt% TMPDA	90	solvent development	136	1.83	[54]

**Table 3**Advantage and disadvantage of different CO<sub>2</sub> capture technologies [102,111, 266,267].

CO <sub>2</sub> capture Technology	Advantage	Disadvantage
Amine Scrubbing	<ul style="list-style-type: none"> <li>• Adsorption capacity is high</li> <li>• The technology is well known and is widely used in various industries</li> <li>• Recovery rate of up to 95%</li> <li>• Thermally stable</li> </ul>	<ul style="list-style-type: none"> <li>• Loss of sorption capacity over multiple cycles</li> <li>• Significant energy requirement for the solvent regeneration</li> <li>• Degradation and equipment corrosion</li> <li>• Solvent emission has negative impact on environment</li> </ul>
Oxy Combustion Capture	<ul style="list-style-type: none"> <li>• Concentration of CO<sub>2</sub> in flue gas is high (60%)</li> <li>• CO<sub>2</sub> separation is easier without N<sub>2</sub></li> <li>• NO<sub>x</sub> free emission</li> <li>• Mature ASU system</li> <li>• Low volume of gases involves smaller equipment size</li> </ul>	<ul style="list-style-type: none"> <li>• Energy penalty due to the air separation unit</li> <li>• CO<sub>2</sub> recycle required to control combustion temperature</li> <li>• Significant energy requirement for separation of O<sub>2</sub> from air</li> <li>• May present corrosion problems</li> </ul>
Direct Separation	<ul style="list-style-type: none"> <li>• High CO<sub>2</sub> removal efficiency</li> <li>• Can achieve net zero or even net negative emission</li> </ul>	<ul style="list-style-type: none"> <li>• The concentration of CO<sub>2</sub> in air is low (420 ppm) which make the process energy intensive</li> </ul>
Calcium Looping	<ul style="list-style-type: none"> <li>• The technology is well known</li> <li>• No thermal formation of NO<sub>x</sub></li> <li>• Exhaust gas stream are not harmful</li> </ul>	<ul style="list-style-type: none"> <li>• Large scale demonstration is not available</li> </ul>

capture hub approach, small plants could save approximately 10% on operating costs, while cement plants could save about 5% [81].

Calcium looping has two configurations for CO<sub>2</sub> capture in the cement industry, i.e., integrated configuration and tail-end configuration. Several studies have explored the concept of integrated configuration technology with the cement industry to capture CO<sub>2</sub> [82–85]. A cement kiln with an integrated calciner performs calcination within the calciner in an integrated configuration [86]. The tail-end configuration involves capturing CO<sub>2</sub> from the clinker process using a carbonator at the end of the pipe. In the cement industry, the tail-end configuration is more suitable for large-scale exhibitions and more mature than the integrated configuration [87,88].

The integrated technology was further developed under the European funded CLEAN clinker (CLEANKER) project from 2017 to 2021, which aimed the project at TRL7 in Buzzi Unicem's Cement plant in Vernasca, Italy. Fuel consumption in the tail-end configuration is more than in the integrated configuration [87]. The estimated cost of tail-end

and integrated configurations in the cement industry is approximately \$55/t CO<sub>2</sub> and \$61/t CO<sub>2</sub>, respectively [63].

Taiwan Cement Company (TCC) and Industrial Technology Research Institute (ITRI) installed a calcium looping driver at Ho Ping's cement plant in Hualien, Taiwan, in 2013 to demonstrate high-efficiency Calcium Looping Technology (HECLOT). The plant captured 1 ton of CO<sub>2</sub> per hr. from 3.1 tons of flue gas (having ~25 % CO<sub>2</sub>) (Table 1). The captured rate was more than 85% and the capturing cost was approximately \$30/t CO<sub>2</sub> [89]. It was the largest calcium looping plant in the world and ITRI received more than 100 R&D awards for this technology [90]. An additional calcium looping project, CEMCAP (2015–2018) (tail-end calcium looping), was tested on two pilot plants: a 30 kWth rig at INCAR Spain and a 200kWth rig at IFK with an efficiency of CO<sub>2</sub> up to 98 % [91] (Table 1).

Currently, calcium looping technology faces the problem of high calcination temperatures and decaying CO<sub>2</sub> capture efficiency. Several studies have shown that the sintering of CaO gradually intensifies with increased calcination temperature, resulting in the decay of CO<sub>2</sub> capture performance [76,92]. We have summarized recent literature and attempted to reduce the calcination temperature. Anbalagan et al. [93] and Haji-Sulaiman et al. [94] investigated that impurities reduced the decomposition temperature and increased limestone calcination rates. It can also be stated that the energy required for the decomposition of limestone is lower than pure CaCO<sub>3</sub> decomposition [95]. According to Valverde et al. [96], milled limestone can regenerate CaO at 900 °C more than raw limestone calcination at 950 °C. Dolomite (CaMg(CO<sub>3</sub>)<sub>2</sub>) is an abundant material and also a low-cost precursor used to prepare CaO-based sorbents because of the homogeneous distribution and inert MgO within the dolomite-derived sorbents [97]. Doping limestone with salts having a low melting point, such as KCl, CaCl<sub>2</sub>, NaCl, Na<sub>2</sub>CO<sub>3</sub>, KMnO<sub>4</sub>, and CaBr<sub>2</sub> is another way to reduce the calcination temperature. Al-Jeboori et al. [98] studied that low doses of CaCl<sub>2</sub> or MgCl<sub>2</sub> doping in limestone increased the pores by around 100 nm, which promotes the carbonation reaction of CaO.

### 2.3. Direct Separation

A direct separation technology involves circulating air through regenerative filters to directly capture CO<sub>2</sub> from the atmosphere. Since 1999, direct separation has decreased atmospheric CO<sub>2</sub> concentration [99,100]. Carbonates are formed from this reaction, which can be calcined to generate CO<sub>2</sub>, while hydroxide streams are recirculated in a closed loop. McLaren [101] states this direct separation technique has a Technology Readiness Level (TRL) level of 4–6, with a potential CO<sub>2</sub> capture capacity of 10 Gt/year. Currently, the technology has a TRL of 7 [102]. Several companies are developing pilot and commercial facilities, including Climeworks, Global Thermostats, and Carbon Engineering [102,103]. With Sunfire and Audi, Climeworks built and operated a pilot



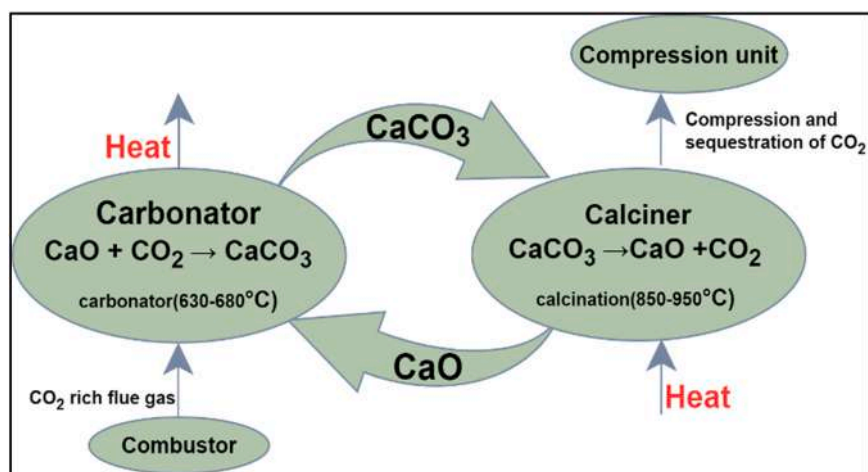


Fig. 4. Principle of calcium looping process [72,73].

plant in Dresden in 2014 that captured 80 % of the air's CO<sub>2</sub> [104].

According to Benhelal et al. [29], CO<sub>2</sub> is captured from the lime and cement industry without being exposed to air or combustion gases. The direct separation technology developed by Calix, an Australian company, captured approximately 64–70 % of the CO<sub>2</sub> produced by a standard cement plant [105,106]. LEILAC is a low-emission intensity lime and cement developed through a European-funded project between 2016 and 2020. In the LEILAC system, the heat from the exhaust gas is transferred to the limestone through a steel vessel called a direct separator reactor (DSR). CO<sub>2</sub> gas captured from this system is almost pure [107] in approximately 25,000 t CO<sub>2</sub>/year, almost 5 % of total CO<sub>2</sub> emissions from the factory (Table 1). After the completion of LEILAC 1, LEILAC 2 project was launched on April 7, 2020, and will be completed by 2025. [108]. LEILAC 2 is almost four times larger than LEILAC 1 and can capture 100,000 t CO<sub>2</sub>/year (Table 1) [108]. After examining different locations, the LEILAC 2 demonstration project is built at the Heidelberg Cement plant in Hanover, Germany. The project's main features were improved technological advancement at the industrial level, a complete integration process in the existing cement plant, and the provision of renewable energy [109].

A significant advantage of direct separation technology is that it can be used anywhere. Using this technology, it is theoretically possible to reach zero net emissions or even to produce negative net emissions. Low concentrations of SO<sub>2</sub> and NO<sub>2</sub> in atmospheric air result in moderate degradation of sorbents [110]. Low concentrations of SO<sub>2</sub> and NO<sub>2</sub> in atmospheric air result in moderate degradation of sorbents [110]. Since ambient air contains 420 ppm of CO<sub>2</sub>, it is relatively low compared with flue gases at power plants. As a result, the cost of direct separation is high. Direct separation technology pilot plants cost \$94–232/t CO<sub>2</sub> but are expected to drop to \$60/t CO<sub>2</sub> by 2040, increasing their competitiveness [111,112]. Both direct separation and calcium looping technology appear to be progressing rapidly. The advantages and disadvantages of both calcium looping and direct separation technology are summarized in Table 3.

#### 2.4. Oxy-combustion capture

Oxy-combustion technology is promising for CO<sub>2</sub> capture in the cement industry (CSI/ECRA, 2009). However, development is expected to take longer than amine scrubbing technology [113]. Oxy-combustion technology uses pure O<sub>2</sub> for the combustion of fuels. During this process, oxygen combustion increases the kiln temperature, which can cause structural damage [114,115]. Parts of the flue gas rich in CO<sub>2</sub> must be recycled back into the kiln to maintain the temperature. The resulting CO<sub>2</sub> stream is purified in a simple cryogenic central processing unit (CPU), achieving almost 95 % of CO<sub>2</sub>. Higher purity of CO<sub>2</sub> is also

achieved by using cryogenic distillation [116]. Contaminants present in the flue gas, like SO<sub>2</sub>, particles, etc., are removed with the help of desulphurization and a standard electrostatic precipitator, respectively. Oxyfuel combustion can improve fuel efficiency and capture CO<sub>2</sub> at a lower cost in cement plants. According to Hörtl et al., oxyfuel combustion applies to the combustion of low-calorific fuels, such as agricultural waste and municipal waste biomass, and the flame's temperature can be controlled by adjusting the flue gas recycling rate [117,118].

The European cement research academy's (ECRA's) long-running research project has been conducting research on CO<sub>2</sub> capture since 2007. Phases I, II, and III of ECRA's CCS project were completed and continuing the phase IV project [119,120]. The two European cement plants demonstrated industrial-scale oxy-combustion CO<sub>2</sub> capture in 2018, the Lafarge Holcim plant in Retznei (Austria) and the Heilderberg Cement plant in Colferferro (Italy) [121], where clear steps were taken to build an oxyfuel kiln. Such kilns were intended to provide insight into the industrial performance of technology that provides high CO<sub>2</sub> emissions for further carbon capture and the cost is approximately \$84.1 M. CEMCAP (2015–2018) is another oxyfuel testing project manufactured by IKN wherein CO<sub>2</sub> was used for cooling the clinker and incorporated into the Heilderberg Cement plant in Hannover, Germany. The oxy-combustion technology produces high purity of oxygen (95 %) in the Air Separation Unit (ASU) [122]. The estimated cost of the technology in the cement plants is almost \$44/t CO<sub>2</sub> (Table 1) [63]. As the world's first and largest oxy-fuel demonstration power plant retrofitting an existing PC-fired boiler, the Callide oxy-fuel power plant with 30 MW capacity commenced in 2011 in Queensland, Australia [123]. The plant consists of four units, each with a capacity of 30 MW. A recent literature review of oxy-fuel combustion demonstrations at pilot and industrial scales is provided in Table 4 [123–125]. It is expected that the success of

Table 4

Projects on oxy-fuel combustion technology (Pilot and industrial demonstration) [123–125].

Companies	Year	Project name	Capacity (MW <sub>e</sub> ) and New/Retrofit	Location
ENDESA, CIUDEN and Foster	2015	Compostilla (OXY-CFB-300)	320, New	Spain
Vattenfall	2015	Janschwalde	250, New	Germany
FutureGen Alliance	2015–2016	FutureGen	210, Retrofit	USA
Black Hills Corporation	2016	Black Hills Power	100, New	USA
KEPCO	2016–2018	Youngdong	100, Retrofit	South Korea

these demonstrations will lead to the greater commercial potential.

### 3. CO<sub>2</sub> utilization in the construction industry

#### 3.1. Mineral carbonation

Mineral Carbonation (CO<sub>2</sub> mineral storage) is an accelerated form of natural carbonation or weathering carbonation of natural silicate rocks in which CO<sub>2</sub> is permanently stored in the form of thermodynamically stable carbonate (CaCO<sub>3</sub>/MgCO<sub>3</sub>) (Eq. 2) [126].

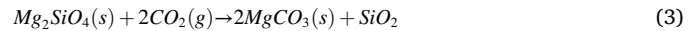


CO<sub>2</sub> storage by mineral carbonation is more expensive than geological storage due to pretreatment requirements, including extracting Ca and Mg from alkaline waste [127]. Standard Gibbs free energy ( $\Delta G$ ) of carbonate is much lower than CO<sub>2</sub>, which means CO<sub>2</sub> storage through mineral carbonation is safe [128].

Most previous research focused on carbonating natural silicates such as serpentine, olivine, limestone, wollastonite, and forsterite because these minerals are rich in Ca and Mg. The carbonation of these minerals is sufficient to store CO<sub>2</sub> emitted from the combustion of fossil fuels and the cement industry. But, utilization of these minerals for carbonation is not cost-effective because of a slow rate of carbonation reaction and large-scale mining operation [129,130]. As a result, there has been an increase in the use of alkaline solid wastes such as cement waste, steel slag, and coal fly ash for CO<sub>2</sub> sequestration. Alkaline solid wastes are more suitable for mineral carbonation because of their faster reaction rate, energy input is meager, and carbonate conversion efficiency is higher than natural minerals. Mineral carbonation of solid waste offers the double benefits of waste management and reduction of CO<sub>2</sub> gas [131]. Mineral carbonation is classified into direct and indirect carbonation; pathways of mineral carbonation are shown in Fig. 5.

#### 3.1.1. Direct carbonation

**3.1.1.1. Gas-solid carbonation.** It was first studied by Lackner and co-workers, which is the most straightforward reaction between the Ca/Mg-rich solid with the CO<sub>2</sub> gas [132]. Gas-Solid Carbonation of olivine shown in Eq. 3.



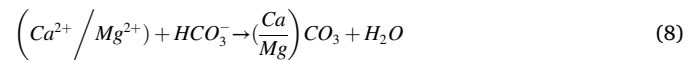
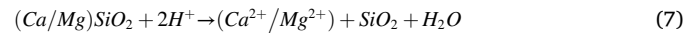
The reaction rate of Gas-Solid Carbonation is very slow or insufficient, even at elevated temperatures and pressure. Forsterite, serpentine, and wollastonite have Gibbs free energies ( $\Delta G$ ) of 44.6 kJ/mol, 43.0 kJ/mol, and 16.9 kJ/mol, respectively. These values indicate that the reaction is spontaneous, but the reaction rate is very slow. Therefore, It is necessary to maintain a high temperature (100–500 °C) and a pressure of 100–150 bar CO<sub>2</sub> to achieve a reasonable reaction rate [129,130]. In their study, DaCosta and coworkers passed the flue through fine-grained silicate rocks (2.5–60 µm). They reported that using 5 g of olivine and 10 % CO<sub>2</sub> at a temperature range of 100–500 °C, 0.12 g CO<sub>2</sub> per gram of olivine can be stored. The capacity for capturing CO<sub>2</sub> was increased to 18 % at a concentration of 15 % [133].

**3.1.1.2. Direct aqueous carbonation.** The addition of water in the two-phase reaction (gas-solid reaction) converts into the multiple-phase reaction (gas-liquid-solid reaction), which increases the carbonation reaction rate.

In this reaction, firstly, CO<sub>2</sub> dissolves into the water to form carbonic acid, then carbonic acid is further ionized into the H<sup>+</sup> and CO<sub>3</sub><sup>2-</sup> Eqs. 4, 5 and 6 [134].



The aqueous solution becomes acidic due to the formation of H<sup>+</sup> ions. Due to the acidic environment, Ca/Mg present in the matrix of the minerals leaches out and reacts with CO<sub>2</sub> to form calcium/magnesium carbonate (Eqs. 7 and 8) [134].



Direct carbonation of recycled concrete aggregate (RCA), in which CO<sub>2</sub> penetrated the RCA through the pores and cracks and dissolved in water present in the pores producing carbonic acid. Calcium ions, which

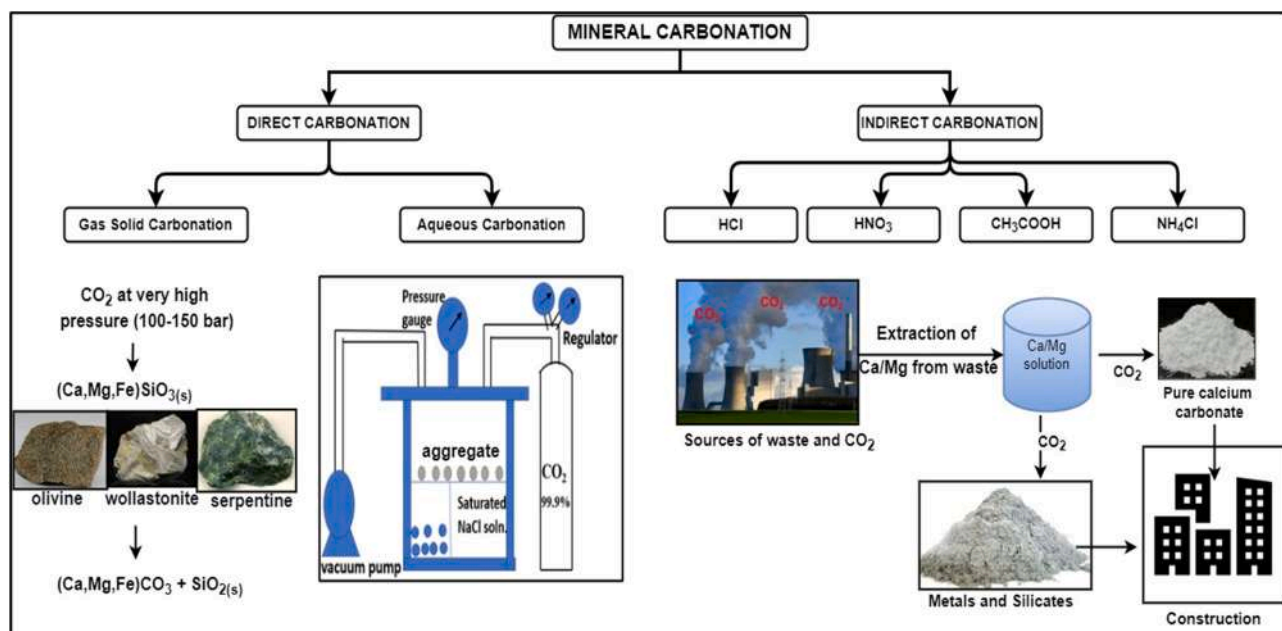


Fig. 5. Pathways of mineral carbonation [134].

leached from the Calcium hydroxide, calcium silicate hydrate, C<sub>3</sub>S, C<sub>2</sub>S, ettringite, calcium sulfoaluminate hydrates, etc., react with carbonate ions to form calcium carbonate and silica gel. Calcium carbonate is precipitated as calcium carbonate polymorphs like calcite, aragonite, and vaterite in the pores. Thus, the cracks of RCA are filled, decreasing the porosity [135,136].

### 3.1.2. Indirect carbonation

Compared with direct carbonation, indirect carbonation proceeds in two or more successive steps (leaching step and carbonation step), where the carbonation step is the rate-determining step [137,138].

1. Leaching step- Extracting reactive components (Mg/Ca) from the minerals using acid or other solvents (Table 5).
2. Carbonation step- Reaction of CO<sub>2</sub> with Ca/Mg in alkaline conditions.

Unlike direct carbonation, indirect carbonation depends more on the carbonation conditions, such as temperature, pressure, and pH [139]. Extraction of Ca and Mg using acid (HCl and HNO<sub>3</sub>) is accessible from the alkaline solid waste/silicate matrix. Lackner et al. first proposed extraction through HCl in 1995 [140]. The main limitation of this route is the cost of HCl, the energy consumption during the evaporation of the aqueous solution and the estimated cost of more than \$157/ton of CO<sub>2</sub> [141]. Extraction of magnesium from serpentinite using HNO<sub>3</sub> and carbonation of magnesium salt was carried out by Teis et al. [142,143]. In both cases, excess acid is required to efficiently extract calcium and magnesium from the alkaline solid waste, which increases the pH of the solution causing a decrease in carbonation. Therefore, it is necessary to increase the pH of solid waste before the carbonation reaction by approximately 8–9 pH by using caustic reagents [142,143].

Adding organic acids such as EDTA increases the extraction of Ca and Mg in mildly acidic conditions. As a result, less caustic reagent would be required during the carbonation process. However, extraction through organic acid is not economically beneficial because of the higher cost

NH<sub>4</sub>Cl and caustic reagents are not required during the carbonation steps. It is the most promising method, which has the potential for the recovery of reagent and reuse [147].

Cement kiln dust (CKD) is a byproduct of Portland cement manufacture in high-temperature rotary kilns [148]. CKD is classified as hazardous waste because it has a caustic nature and the composition of CKD is 38–48% CaO and 1.5–2.1 % MgO [137,149]. The CKD is already carbonated and contains 38–45 % CaCO<sub>3</sub> [150]. Cement bypass dust (CBD) has lower carbonated content than CKD and has more potential to sequester CO<sub>2</sub> than CKD by indirect carbonation [151]. CKD has sequestered up to 42 Mt of CO<sub>2</sub> annually or stores approximately 0.1 % of CO<sub>2</sub> emission globally from the combustion of fuels [152]. The CKD and CBD have the potential to store 0.08–0.025 t CO<sub>2</sub>/t CKD and CBD at ambient pressure and temperature in a column reactor [150,151].

Waste cement is a byproduct of concrete, where aggregate is separated from the waste cement. According to Bobicki et al., waste cement generated from the EU, China and the USA was approximately 1100 Mt and could sequester about 61 Mt CO<sub>2</sub> [137]. Most waste cement is already used in construction by mineral carbonation [148]. During indirect carbonation of the waste cement, extraction of calcium in the leaching step from the cement waste is achieved by HCl, CH<sub>3</sub>COOH, and NH<sub>4</sub>Cl (Table 2). When the concentration of CH<sub>3</sub>COOH and HCl is 0.5 M, the extraction of calcium is approximately 13,220 mg/L and 13,670 mg/L, respectively, whereas the extraction efficiency of 0.5 M NH<sub>4</sub>Cl is around 6733 mg/L. During carbonation, calcium extraction by NH<sub>4</sub>Cl in the leaching step sequestered approximately 0.1 kg CO<sub>2</sub>/kg cement, whereas extraction by HCl and CH<sub>3</sub>COOH could sequester 1.4Kg CO<sub>2</sub>/Kg cement [127]. Mineral carbonation in waste cement using the extracting agent NH<sub>4</sub>Cl is more economically beneficial than HCl and CH<sub>3</sub>COOH because of the fully recycling of solvent and the need for a caustic reagent to increase pH during the carbonation step.

The following Eq. 9 measures the efficiency of indirect carbonation [124].

$$\eta_{\text{(Carbonation)}} \% = \frac{\text{quantity of Mg or Ca converted into carbonate}}{\text{quantity of Mg or Ca available in the minerals}} \times 100 \quad (9)$$

and recycling of acid in the leaching steps [144]. Extraction using acetic acid is another approach and the advantage of this approach is the recovery of acetic acid in the carbonation step and recyclability [145,146]. Another extraction process for calcium and magnesium developed by Kodama et al. used NH<sub>4</sub>Cl, which has the potential for fully recycling the

Efficiency depends on the Ca or Mg content in the minerals, not the quantity of the minerals used for the carbonation.

### 3.2. Cement-based materials

In the past, cement carbonation was viewed as a negative factor as it deteriorates the hydration products with time, called weathering carbonation. The main component of cement is calcium-silicate-hydrate, which, when exposed to atmospheric CO<sub>2</sub>, decalcifies and eventually transforms into silica gel, losing its binding properties and durability [153]. Additionally, the high alkalinity of the cement product protects the steel reinforcement from corrosion. However, in recent years, studies have shown positive effects from the carbonation of cement hydrates, specifically in early-age carbonation. CO<sub>2</sub> curing of cement-based materials has gained more attention since the 1990s due to the increase in global warming. Carbonation of cement-based materials has been suggested recently for more active sequestration of CO<sub>2</sub> and numerous research are conducted in this field [154–156]. The carbonation reaction between the cement-based materials with CO<sub>2</sub> formed a thermodynamically stable carbonate [157,158].

It has been suggested that accelerated carbonation at an early age accelerates cement's hardening process in which CO<sub>2</sub> gas reacts with Ca (OH)<sub>2</sub>, cement clinkers (C<sub>2</sub>S and C<sub>3</sub>S) and C-S-H. The reaction of CO<sub>2</sub>

**Table 5**  
Extraction of reactive elements by different reagents.

Extracting reagent	Chemical reactions involved in the extraction	Ref.
HCl/HNO <sub>3</sub>	<ul style="list-style-type: none"> <li>• <math>\text{Mg}_3\text{Si}_2\text{O}_5(\text{OH})_{4(s)} + 6(\text{HNO}_3/\text{HCl})_{(aq)} \rightarrow 3\text{Mg}_{(aq)}^{2+} + 6(\text{NO}_3/\text{Cl})_{(aq)} + 2\text{SiO}_{2(s)} + 5\text{H}_2\text{O}_{(l)}</math> [140, 141]</li> <li>• <math>\text{Mg}_{(aq)}^{2+} + 2(\text{NO}_3/\text{Cl})_{(aq)} + x\text{H}_2\text{O}_{(l)} + y(\text{HNO}_3/\text{HCl})_{(aq)} \rightarrow \text{Mg}(\text{NO}_3)_2/\text{Cl}_2 \cdot 6\text{H}_2\text{O}_{(s)} + (x-6)\text{H}_2\text{O}_{(g)} + y(\text{HNO}_3/\text{HCl})_{(g)}</math></li> <li>• <math>5\text{Mg}((\text{NO}_3)_2/\text{Cl}_2)_{(aq)} + 10\text{NaOH}_{(aq)} + 4\text{CO}_{2(g)} \rightarrow 10\text{Na}(\text{NO}_3/\text{Cl})_{(aq)} + \text{Mg}_5(\text{OH})_2(\text{CO}_3)_4 \cdot 0.4\text{H}_2\text{O}_{(s)}</math></li> </ul>	
CH <sub>3</sub> COOH	<ul style="list-style-type: none"> <li>• <math>\text{CaSiO}_3 + \text{CH}_3\text{COOH} \rightarrow \text{Ca}^{2+} + \text{SiO}_2 + 2\text{CH}_3\text{COO}^- + \text{H}_2\text{O}</math> [145, 146]</li> <li>• <math>\text{Ca}^{2+} + 2\text{CH}_3\text{COO}^- + \text{H}_2\text{O} + \text{CO}_2 \rightarrow \text{CaCO}_3 + 2\text{CH}_3\text{COOH}</math></li> </ul>	
NH <sub>4</sub> Cl	<ul style="list-style-type: none"> <li>• <math>4\text{NH}_4\text{Cl} + 2\text{CaSiO}_3 \rightarrow 2\text{CaCl}_2 + 2\text{SiO}_2 + 4\text{NH}_3 + 2\text{H}_2\text{O}</math> [147]</li> <li>• <math>2\text{CaCl}_2 + 2\text{CO}_2 + 4\text{NH}_3 + 2\text{H}_2\text{O} \rightarrow 2\text{CaCO}_3 + 4\text{NH}_4\text{Cl}</math></li> </ul>	



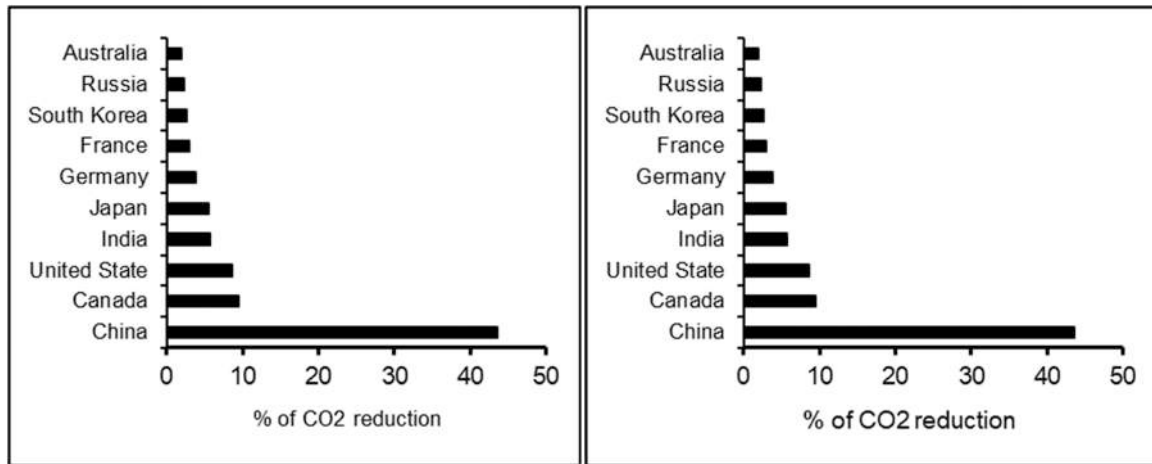
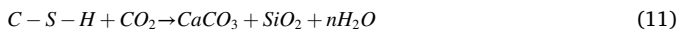


Fig. 6. . (a) % CO<sub>2</sub> reduction due to mineralization (b) worldwide CO<sub>2</sub> reduction through mineralization of alkaline solid waste [131].

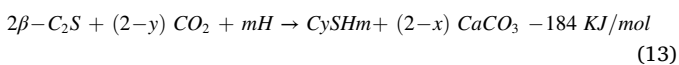
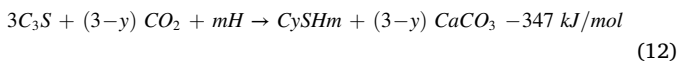
with cement paste mainly gives CaCO<sub>3</sub> and silica gel. This early-age carbonation improves the microstructural compactness of cement-based materials, resulting in greater strength and performance at an early age. Carbonation of Ca(OH)<sub>2</sub> occurs in different stages, i.e., dissolution of Ca(OH)<sub>2</sub> to form calcium ions, CO<sub>2</sub> gas reacts with water to form carbonate ions which further reacts with the calcium ions to form calcium carbonate polymorphs (calcite, vaterite and aragonite) (Fig. 6) [159].



Similarly, carbonation of C-S-H forms calcium carbonate and amorphous silica gel (Eq. 11), which promotes the removal of Ca<sup>2+</sup> from the C-S-H, leading to a reduced C/S ratio and a decline in C-S-H stability [160]. Both Ca(OH)<sub>2</sub> and C-S-H carbonate sequentially; however, Glasser et al. [114] reported that thermodynamically carbonation of Ca(OH)<sub>2</sub> took precedence over C-S-H. Morandau et al. [161] contradicted this by reporting that both Ca(OH)<sub>2</sub> and C-S-H carbonation occur simultaneously. Thermogravimetric analysis (TGA) concluded that initial carbonation rates for Ca(OH)<sub>2</sub> and C-S-H appeared to be comparable. However, carbonation of Ca(OH)<sub>2</sub> reached a relatively sufficient level with steady utilization of Ca(OH)<sub>2</sub>, and C-S-H gel continued to carbonate.



C<sub>3</sub>S, C<sub>2</sub>S, tricalcium aluminate (C<sub>3</sub>A), and tetracalcium aluminoferrite (C<sub>4</sub>AF) are the main mineral compositions of C-S-H based cement clinker. According to Liu et al., the carbonation of cement clinker is in the order of C<sub>3</sub>S > C<sub>2</sub>S > C<sub>3</sub>A > C<sub>4</sub>AF [162]. C<sub>2</sub>S and C<sub>3</sub>S are the dominant carbonatable reactants compared to C<sub>3</sub>A and C<sub>4</sub>AF, as they are in small quantities [163]. In the presence of water, C<sub>2</sub>S and C<sub>3</sub>S carbonated to form CaCO<sub>3</sub> and silica gel rather than Ca(OH)<sub>2</sub>. Carbonation of C<sub>3</sub>S, β-C<sub>2</sub>S and γ-C<sub>2</sub>S was generally faster than the hydration [164]. The mechanism of early-age carbonation was different from the weathering carbonation. The carbonation reaction of anhydrous alite C<sub>3</sub>S and belite (C<sub>2</sub>S) is given in Eqs. 12 and 13 [165] and these reactions are spontaneous and exothermic.



In Eqs. 12 and 13, C<sub>y</sub>SH<sub>m</sub> represents the (CaO)<sub>y</sub>(SiO<sub>2</sub>)(H<sub>2</sub>O)<sub>m</sub>, which can be described simply by C-S-H. Formation of C-S-H gel on

carbonation was also supported by the result of Berger et al. [166], in which β-C<sub>2</sub>S and C<sub>3</sub>S powders are cured by CO<sub>2</sub> gas and the heat release is 347 kJ/mol and 184 kJ/mol for C<sub>3</sub>S and β-C<sub>2</sub>S, respectively and the result also proved that compressive strength of CO<sub>2</sub> cured mortar increased and further strength is gained by subsequent water curing. The same result is also reported in the other literature [167,168]. C<sub>2</sub>S has five distinct polymorphs and can be activated by CO<sub>2</sub> for strength gain. Among the five polymorphs, γ-C<sub>2</sub>S is non-hydraulic and the rests of them are hydraulic and this CO<sub>2</sub> curing of γ-C<sub>2</sub>S was studied by Bukowski and Bergen in 1972 [169] and later complemented by FTIR and NMR [170, 171]. CO<sub>2</sub> uptake by γ-C<sub>2</sub>S is higher than β-C<sub>2</sub>S. They form different CaCO<sub>3</sub> polymorphs [170] and provide more mechanical strength [171]. Tricalcium aluminate (C<sub>3</sub>A) had minimal to no reactivity with CO<sub>2</sub> [172]. Ettringite is another cement phase produced by C<sub>3</sub>A and C<sub>4</sub>AF as a reaction product of hydration [173], which is prone to CO<sub>2</sub> reaction and decomposes to produce gypsum, alumina gel, calcite, amorphous gel and water [174,175]. No evidence exists in any literature on the carbonation of aluminoferrite (C<sub>4</sub>AF) [176].

A study of the percentage of CO<sub>2</sub> uptake by different cement-based materials is presented in Table 6. Overall, the CO<sub>2</sub> uptake was significantly increased by an increase in CO<sub>2</sub> concentration and pressure. Even though a higher temperature of up to 100 °C is beneficial for CO<sub>2</sub> sequestration, room temperature (about 20 °C) is usually preferred to avoid more energy consumption, which causes extra CO<sub>2</sub> emissions [177]. In the RH range of 50–70 %, the carbonation of cement paste is the highest [177]. Water content also plays a significant role in this process, so the pre-curing of samples is done before carbonation. A three-step curing procedure is usually used to achieve more effective carbonation, starting with pre-curing followed by standard carbonation and subsequent carbonation [178,179].

Due to CaCO<sub>3</sub> precipitation, the cement pastes' chemical composition changed, which changed its microstructure. Shi et al. [180] studied

Table 6  
CO<sub>2</sub> uptake % by cement-based materials.

Cement-based materials	Mineral composition %		CO <sub>2</sub> uptake %	Ref.
	CaO	MgO		
Lightweight concrete masonry unit	–	–	22.0–24.0	[268]
Waste hydrated cement paste	61.0	2.0	15.0–24.0	[269]
Belite-rich Portland cement	62.5	–	16.9	[270]
Precast concrete	–	–	12.7	[190]
Calcium silicate concrete	63.1–63.9	2.0–3.5	8.1–14.0	[271]
Early curing of cement paste subject	63.1	2.0	7.5–19.2	[272]

carbonated concrete microstructure and showed the filling effect, which decreased the porosity of the concrete. These changes remarkably improved the mechanical and durability of samples. The pores with 50–100 nm is largely filled during carbonation, whereas 10–50 nm pores size prevails throughout the volume size for the uncarbonated cement paste. Early carbonation occurred due to the rapid dissolution of cement and subsequent carbonation reaction in the pore solution [181]. Carbonation results in a low Ca/Si ratio in cement paste, promoting early-age hydration due to a higher degree of polymerization [182]. A study by Zhan et al. [183] found that CO<sub>2</sub> cured cement paste had a compressive strength twice that of water-cured cement paste after 24 h.

### 3.3. Recycled aggregates

Recycling construction and demolished (C&D) waste into a recycled aggregate (RA) is an important method for waste disposal worldwide. Natural disasters, rapid industrialization and urbanization, mainly in developing countries, generated a vast amount of C&D waste and will remain for the next few decades [184,185]. China produced around 1.65 billion and 1.85 billion tons of C&D waste in 2016 and 2017, respectively and was the largest C&D producer in the world [186,187]. Countries such as Germany, Japan and The Netherlands have recycled more than 80% of C&D waste. The rate of recycled waste in other developed countries is almost 20–40 % [188] and the recovery rate of developing countries is almost negligible [84,85]. Replacement of virgin aggregate with recycled aggregate (100 %) reduced the compressive strength of concrete by almost 30–40 % [189,190]. Accelerated carbonation improved the RA properties and captured CO<sub>2</sub> permanently as a stable CaCO<sub>3</sub>. RA carbonation is a very slow process, and to overcome it, CO<sub>2</sub> concentrations, temperatures, and relative humidity are increased, known as accelerated carbonation and the reaction occurs in a few hrs. [136]. According to the different types of carbonation methods (Table 7), standard and pressurized carbonation are the most commonly used methods for carbonating RA [187].

CO<sub>2</sub> uptake by RA is distinct from the concrete structure and generally. Both rate and amount of CO<sub>2</sub> uptake by RA increase with a decrease in particle size due to the more surface area exposed to the CO<sub>2</sub>. The carbonation of concrete structures takes a minimal amount of CO<sub>2</sub> over a long period due to the minimum surface area exposed to CO<sub>2</sub> gas [135,191]. The amount of CO<sub>2</sub> uptake by RA with particles size 5–10 mm is almost 50 % higher than that of 14–20 mm [192] and the CO<sub>2</sub> uptake of 1.18 mm is almost 100 % higher than 7.5 mm [193]. CO<sub>2</sub> uptake is further increased by pressured carbonation and pre-soaking treatment [194,195]. There are two types of RA, a new type RA and an old type of RA. The CO<sub>2</sub> uptake by the old type RA is lower than the new type because the natural carbonation already occurred in the old

type RA during storage and demolition [136]. Rapid carbonation is occurred in the RA after demolition during 1–2 years due to exposed surface area [135,196]. New RA's with particles between 5 mm and 20 mm sequester almost 7.9 kg CO<sub>2</sub> per ton [136]. During the carbonation of RA, thermodynamically stable CaCO<sub>3</sub> is formed, increasing the solid phase volume by ~11.9 % [197]. Pores and cracks in RA are satisfactorily filled by the CaCO<sub>3</sub> and silica gel (formed in the reaction between C-S-H and CO<sub>2</sub>) which enhances the property of RA. Carbonation of RA improved the compressive strength and the elastic modulus. Compressive strength increased with increased RA content and the compressive strength reached almost equal to the natural aggregate concrete at 28 days [198]. The elastic modulus of RA is increased by almost 13 % [198] and 8–27 % [199] on carbonation. An increase in the compressive strength and elastic modulus (3.1–27.0 %) of concrete with CRA is reported in different literature [198–203].

### 3.4. Solid wastes

In recent decades, solid waste generation has expanded dramatically worldwide. In 2020, 12 billion tons of solid waste were generated globally, which is anticipated to rise to 19 billion by 2050 [204]. Around 4.4 billion tons of trash are generated in Asia, with 48 million tons (6 %) annually in India alone [205], causing many economic, environmental, and social issues. Therefore, solid waste management in an effective way is necessary for the development of sustainable and habitable cities. Using ACT, various solid wastes were treated with CO<sub>2</sub> gas and converted into value-added products to address this problem. ACT changes the chemistry of the materials by stabilizing the contaminants [206, 207].

Regarding fundamental research, engineering applications, and economic evaluation [208], Xie et al. recently published a comprehensive review of CO<sub>2</sub> mineralization methods for natural ores and industrial solid wastes. The researchers predicted that CO<sub>2</sub> mineralization and natural resource extraction would be combined with industrial solid waste treatment in the future. Around the world, alkaline solid wastes include iron/steel slags, coal and fuel combustion products, mining/mineral processing wastes, incinerator residues, cement and concrete wastes, and pulp and paper mill waste for CO<sub>2</sub> mineralization. It is reported that ~310 Mt CO<sub>2</sub> can directly reduce CO<sub>2</sub> through mineral carbonation of alkaline solid waste across the globe, wherein 43% CO<sub>2</sub> reduction is achieved by carbonation of steel slag only. Further, it is reported that the total amount of CO<sub>2</sub> reduction by mineral carbonation in China is more than four times (~45 %) that of any country (Fig. 6) [131]. Gunning et al. [151] described the production of light-weight carbonated aggregates with individual pellet compressive strengths exceeding 0.10 MPa. Further, they expanded this study by discussing the

**Table 7**  
Carbonation of RA using various methods.

Method	Condition	Procedure	Conclusion	Ref.
Standard carbonation	RH= 70 ± 5 % Temp.= 20 ± 2 CO <sub>2</sub> conc.= 20 ± 3	<ul style="list-style-type: none"> <li>Carbonation proceeded in standard Chinese GB50082–2009.</li> <li>RA is placed in a chamber.</li> <li>CO<sub>2</sub> is maintained at ambient pressure.</li> </ul>	Carbonation was low and the time of CO <sub>2</sub> curing was also higher.	[197, 273]
Pressurized carbonation	RH= 50 ± 5 Temp.= 25 ± 3	<ul style="list-style-type: none"> <li>RA was first dried in a drying chamber.</li> <li>Vacuumed the chamber to – 0.6 Bar.</li> <li>CO<sub>2</sub> pressure is controlled until a required constant is reached.</li> </ul>	Carbonation was higher than standard.	[190, 193]
Flow-through CO <sub>2</sub> curing	RH= 50 ± 5 CO <sub>2</sub> conc.= 10 %	<ul style="list-style-type: none"> <li>Exposure to RA with CO<sub>2</sub> gas</li> <li>CO<sub>2</sub> is injected from one side and discharged through the opposite side.</li> <li>RH was maintained by using a saturated Mg(NO<sub>3</sub>)<sub>2</sub> solution.</li> <li>The flow rate of CO<sub>2</sub> was almost 5 L/min.</li> </ul>	Low energy consumption and higher efficiency than standard.	[157, 181]
Water CO <sub>2</sub> cooperative		<ul style="list-style-type: none"> <li>RA was first placed in the chamber, which contained water.</li> <li>A mixture of CO<sub>2</sub>, N<sub>2</sub> and O<sub>2</sub> gas was injected into the water.</li> <li>The CO<sub>2</sub> nano bubble was also mixed with the water to increase efficiency.</li> </ul>	Higher efficiency condition for CO <sub>2</sub> curing.	[274, 275]

**Table 8**  
pH changes before and after carbonation [206].

	Biomass ash	MSWI-BA	APC	CKD	PS-AI	WA
Before carbonation	10.6	12.4	11.1	13.2	12.7–13.0	13.4
After carbonation	9.9	9.1	8.4	11.0	9.9–11.4	11.0

\*PSIA (paper sludge incineration ash), MSW-BA (municipal solid waste incineration bottom ash), WA (wood ash), CKD (cement kiln dust), APC (Air pollution control)

commercial possibilities of aggregates formed from municipal solid waste [209].

Fernandez-Bertos et al. [154] examined waste streams for CO<sub>2</sub> gas reactivity. It is reported that due to the formation of CaCO<sub>3</sub>, there is an increase in volume and reduction in porosity, which helps in the retention of contaminants and also reduces the pH of waste below a threshold value (11.5) (Table 8), making them less hazardous [206]. CO<sub>2</sub> uptake percentage is dependent on the Ca and Mg content of the waste and reactivity of CO<sub>2</sub> increases with the Ca content, but the phase of the mineral is also important in which calcium is bonded because CWIA (clinical waste incineration ash) and SSA (sewage sludge ash) contain the significant amount of calcium 28.1 (wt%), 14.8 (wt%) respectively and is expected the carbonation reaction. Still, the reaction was almost negligible because calcium in these solid wastes bounded in a different phase [194,195,197]. CO<sub>2</sub> uptake percentage also depends on the carbonation methods, as some waste requires pre-treatment with different additives. Table 9 represents the Ca and Mg content and the CO<sub>2</sub> uptake% of different solid wastes and Table 10 represents the different carbonation methods of solid wastes.

#### 4. Commercialized approaches

Accelerated carbonation of industrial waste may be utilized to produce engineered products. In a study by Gunning et al. [151], artificial aggregates were produced from alkaline solid wastes such as cement kiln dust, wood ash, and paper ash etc. The process was licensed in 2012 for using MSW air pollution control residue to produce artificial aggregates for construction purposes [209]. Morone et al. [210] produced pilot-scale carbonate bonded aggregates from BOF steel slags using mineral carbonation. After 28 days of curing (pure CO<sub>2</sub> at 140 °C and 20 bar), the products captured up to 10 % CO<sub>2</sub> (v/v). According to Quaghebeur et al. [211], monolithic compacts made from stainless steel waste had a compressive strength of 55 MPa. A commercially available pre-cast building block made from moist carbonated steel slag was introduced in 2017 [212]. Natural sand and finely ground steel slag are used in the products. Since 2009, there has been a continuous increase in the percentage of commercialized CO<sub>2</sub> usage technologies. Fig. 7 shows the number of CO<sub>2</sub> usage patents filed each year from 2009 to 2021.

Several commercial processes have been developed to sequester CO<sub>2</sub> from the atmosphere in response to an increased interest in waste mineralization as a technology for CCU. Some of these are summarized

**Table 9**  
Ca and Mg content (wt%) of different waste and CO<sub>2</sub> uptake %.

Waste	Chemical composition (wt%)			CO <sub>2</sub> uptake (wt%)	Ref.
	CaO	MgO	Ref.		
PS-IA (paper sludge incineration ash)	45–69	1.3–5.3	[206]	17	[206]
MSWI-BA (municipal solid waste incineration bottom ash)	32–53	2.8	[206,276,277]	3–14	[278]
MSWI-FA/APC (municipal solid waste incineration fly ash)	50–60	8	[278–280]	7–25	[281]
RCA (recycled concrete aggregate)	15–24	2–3	[282,283]	7.5, 16.5	[281,284]
WA (wood ash)	24–46	8–9	[206]	8	[206]
CKD (cement kiln dust)	34.5–46.2	1.5–2.1	[149,206]	10	[206]
BF slag (blast furnace slag)	15–41	8–11	[146,285,286]	7, 22.7	[73,283]
Steel slag	25–47	4–15	[146,287]	12–18	[280,288]
EAF (electric arc furnace slag)					
BOF (basic oxygen furnace slag)	34–55	1.5–10	[146,287]	21	[289]

**Table 10**  
Method for carbonation of different solid waste.

Waste	Condition	Method	Ref.
RCA	RH= 70 % CO <sub>2</sub> conc.= 100 % Pressure= 2 bar	<ul style="list-style-type: none"> <li>Waste was treated with pure CO<sub>2</sub> in a pressured vessel.</li> <li>A Saturated NaCl solution was used to maintain the humidity.</li> </ul>	[290]
BFS	CO <sub>2</sub> conc. = 100 % Temp.= 70 °C Pressure= 40 bar Acidic sol.= 20%	<ul style="list-style-type: none"> <li>Reaction proceeded in a batch reactor.</li> <li>Calcium was extracted by an acidic solution.</li> <li>Precipitated by stirring 600–700 rpm for almost 2 hrs. in the presence of CO<sub>2</sub> gas and NaOH solution.</li> </ul>	[286]
EAF	CO <sub>2</sub> conc. = 100 % Temp.= 50 °C Pressure= 10 bar RH = 75 %	<ul style="list-style-type: none"> <li>Carbonation proceeded in the stainless-steel reactor.</li> <li>A Saturated NaCl solution was used to maintain RH</li> <li>Maintain liquid to solid ratio of 0.4 L/kg</li> </ul>	[280]
MSW-BA	Temp = 30 °C CO <sub>2</sub> conc.= 100 % Pressure = 10 bar	<ul style="list-style-type: none"> <li>The reaction proceeded in stainless steel pressurized reactor</li> <li>100% CO<sub>2</sub> and 30 °C temperature for almost 24 hrs.</li> <li>Maintain liquid to solid ratio of 0.3 L/kg</li> </ul>	[279]
APC/MSW-FA	Temp.= 200–500 °C CO <sub>2</sub> conc.= 100 %	<ul style="list-style-type: none"> <li>Carbonation proceeded in a muffle furnace</li> <li>Constant CO<sub>2</sub> 100% flowed for 6 hrs.</li> <li>Ranging the temperature from 200 to 500°C</li> </ul>	[291]
BM	RH = 75 % CO <sub>2</sub> conc.= 100 % Pressure = 2 bar	<ul style="list-style-type: none"> <li>The reaction proceeded in a reactor vessel</li> <li>100%CO<sub>2</sub> atm and 2 bar pressure were maintained.</li> </ul>	[206]

in Table 11 and discussed below.

##### 4.1. Carbon8

The University of Greenwich founded Carbon8 Systems Ltd. (UK) in 2006 to commercialize Accelerated Carbonation Technology (ACT) [213,214]. ACT produces artificial limestone by combining CO<sub>2</sub> with various industrial wastes (thermal wastes from cement manufacturing sites, waste energy, steel slags, etc.). The alkaline solid waste is mixed with liquid CO<sub>2</sub> and water in a pretreatment mixer. Binders and fillers are added to carbonated waste during batch mixing. To produce rounded aggregates, the slurry passes through a pelletizer where gaseous CO<sub>2</sub> is introduced to speed up cementation (Fig. 8) [215].

In 2012, ACT was commercialized and licensed as Carbon8 Aggregates, supported with an investment by Grondon Waste Management and producing > 150,000 tons of product per year. It can permanently absorb over 1 million tons of CO<sub>2</sub> using only 20 % of the waste available in Europe [216]. Furthermore, for every ton of Carbon8 aggregate, 1.4 tons of natural aggregate are preserved, and 0.5 tons of waste are

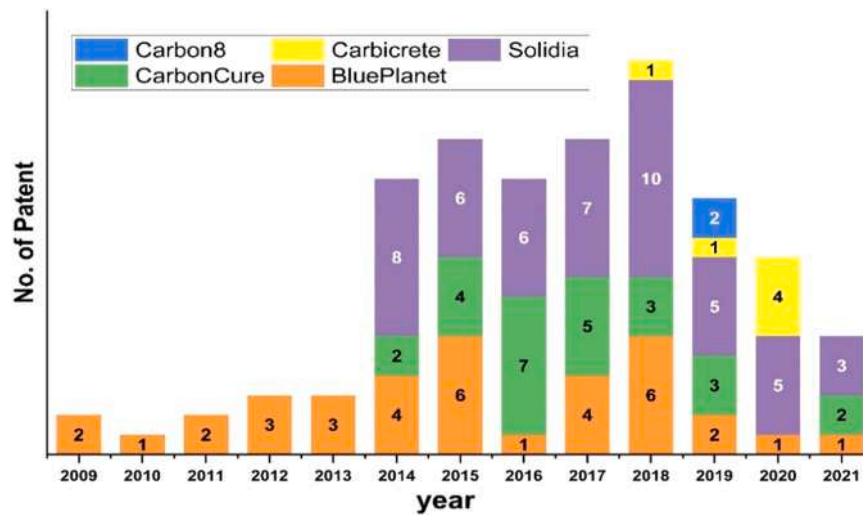


Fig. 7. No. of patents published by different companies with respect to the year [306].

Table 11

Status of selected Commercialized technologies.

Technology	CO <sub>2</sub> sequestration method	TRL	CO <sub>2</sub> uptake	Location	Patent filed & year	Ref.
Carbon8	Permanent capture of CO <sub>2</sub> through ACT	9	100–200 kg CO <sub>2</sub> /ton of aggregate	UK	U.S.Patent No. 10,343,199, 2019 U.S. Patent No. 20190119158, 2019	[213,214]
Carbon Cure	Injection of CO <sub>2</sub> in the concrete mix.	8–9	161713 MT CO <sub>2</sub> save till now	UK, Canada	U.S.Patent No. 8845,940, 2014 U.S. Patent No. 9492,945, 2016 U.S.Patent No. 9376,345, 2016 U.S.Patent No. 10,570,064, 2020	[228–230, 292]
Solidia	CO <sub>2</sub> curing cement concrete	8	1.5 gigatons of CO <sub>2</sub> sequestered/year & 250–300 kg CO <sub>2</sub> /ton of cement.	USA, Canada	U.S.Patent No. 9221,027, 2015 U.S.Patent No. 10,016,739, 2018 U.S.Patent No. 10,668,443, 2020	[293–295]
Carbstone	Developed high-quality materials by adding CO <sub>2</sub> from flue gas to steel slag	9	180–200 g CO <sub>2</sub> /kg of steel slag	Belgium	US Patent No. 8709,151, 2014 U.S. Patent Application 14/354,024, 2014	[296,297]
BluePlanet	Capture CO <sub>2</sub> is coated over the calcium rich substrate to form aggregate	6–7	440 kg CO <sub>2</sub> /ton of aggregate	California USA	U.S. Patent No. 9993,799, 2018 U.S. Patent No. 10,766,015, 2020 U.S. Patent No. 9714,406, 2017 U.S. Patent 10,197,747, 2019	[298–301]
Carbicrete	Carbonation of steel slag to replace cement (cement free concrete)	6–7	200 kg CO <sub>2</sub> /day	Canada	U.S. Patent No. 10,112,871, 2018 U.S. Patent No. 10,633,288, 2020	[302,303]
Calera corporation	Carbonated precipitates from CO <sub>2</sub> in water/ brines	8–9	3.4MT CO <sub>2</sub> /year & 460 kg CO <sub>2</sub> /ton	USA	U.S.Patent No. 7887,694, 2011 U.S. Patent No. 8333,944, 2012 U.S.Patent No. 7771,684, 2010 U.S. Patent 8006,446, 2011	[222,223,304, 305]

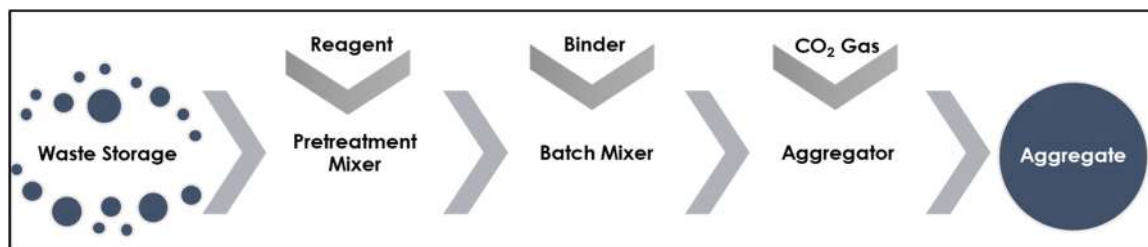


Fig. 8. Flow Chart of Carbon8 aggregate production [215].

diverted from landfills [151,206,217]. A total of 350,000 tons of aggregate are now produced in the United Kingdom each year using this technology, and this number will grow to over 750,000 tons in the next five years [218]. In April 2017, Carbon8 Systems received the UK's highest corporate honor award (Queen's Award) for its breakthrough technology [219].

Carbon8 demonstrated the first CO<sub>2</sub>ntainer project in 2018 at a

cement factory owned by Cement Roadstone Holdings (CRH) in Mississauga, Ontario. The project demonstrated direct CO<sub>2</sub> capture from flue gases and the potential to produce two commercially viable products, i.e., mineral-rich fertilizers and lightweight aggregates [220]. In the CO<sub>2</sub>ntainer, the volume of trash that will be processed corresponds to a fixed place's trash output (8000–12,000 Mt/year), eliminating the transportation of waste materials and the emissions of CO<sub>2</sub>. In addition



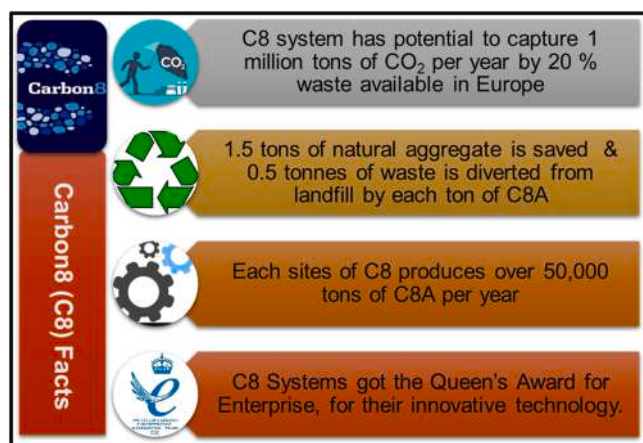


Fig. 9. Facts of Carbon8 [213,214].

to precast concrete blocks, pipe bedding, and road sub-base, the process's outputs can be used in high-value goods like lightweight flooring and green roofing substrates or as fertilizers according to the source of the industrial waste. The Carbon8 company announced its first global licensing agreement with a significant French cement company in 2019 [220].

Both Carbon8 and CEMEX Systems recognize mineralization's potential as a carbon sink, and they have partnered to develop a line of low-carbon building products. This technology can be applied to the production of byproducts as well as to the production of alternative aggregates and cementitious materials [221]. Fig. 9 illustrates some carbon8 facts.

#### 4.2. Calera corporation

Calera is a Los Gatos, CA-based startup company which develops a new carbon capture system wherein mineral carbonate is converted into a stable carbonate slurry that can be used to make cement-based materials or building materials [222,223]. The flue gas reacts with alkaline solutions rich in Ca or Mg to form a thermodynamically stable CaCO<sub>3</sub> solid. They further used carbonate as a cement building material and Supplementary Cementitious Materials (SCM) (Fig. 10) [134]. This process has offset carbon emissions compared with traditional cement production and generated positive revenue. In a cost-analysis of carbon capture and storage, Charles Kolstad found that the Calera process would be the cheapest way to retrofit an existing coal-fired power plant to capture CO<sub>2</sub> [224,225]. A research team at Calera has identified regions in the United States and China that may have sufficient alkalinity and calcium for the process. A series of projects are now being developed in these areas. Calera's technology might prove useful in developing nations, such as China and other countries where the cement and power industries are growing rapidly [226].

A demonstration facility developed by Calera in Moss Landing, California, can capture 30,000 tons of CO<sub>2</sub> per year, equivalent to the effluent of a 10-megawatt natural gas power plant. Using flue gas from a

Table 12

A comparison of different concrete mixes illustrates different formulations in lb. CO<sub>2</sub>/yd<sup>3</sup> concrete to reduce the carbon footprint in the concrete product via offsetting and CO<sub>2</sub> sequestration [226].

Ordinary Portland concrete						
Ingredient	Ordinary Portland cement	Water	Fine aggregate	Coarse aggregate	Total	
lb. CO <sub>2</sub> / yd <sup>3</sup> concrete	494.1	2.8	16.9	23.4	537	
High volume fly ash concrete						
Ingredient	Ordinary Portland cement & Fly ash	Water	Fine aggregate	Coarse aggregate	Total	
lb. CO <sub>2</sub> / yd <sup>3</sup> concrete	247.0 & 12.7	2.7	15.9	23.4	302	
Carbon native concrete (first carbon native concrete)						
Ingredient	Ordinary Portland cement & fly ash	Water	Calera SCM	Calera fine aggregate	Calera coarse aggregate	Total
lb. CO <sub>2</sub> / yd <sup>3</sup> concrete	269.1 & 5.1	2.7	-50.9	-562.5	-810.0	-1146

100-megawatt output, the company claims that this facility can produce almost 550,000 tons of valuable construction materials annually from CO<sub>2</sub> [227]. According to the company, using Calera fine and coarse aggregate creates the most carbon-neutral concrete. Portland cement-based concrete has a 600 lb CO<sub>2</sub>/yd<sup>3</sup>. Reducing Portland cement with fly ash (50%) decreases its carbon footprint by 300 lb CO<sub>2</sub>/yd<sup>3</sup>. Replacing the natural fine and coarse aggregate with Calera fine and coarse aggregate reduces the carbon footprint by 1146 lb CO<sub>2</sub>/yd<sup>3</sup> (Table 12) (Fig. 11). In 2009, Calera SCM replaced 20% of OPC in the US, resulting in a decrease in cement output from 75 Mt/year to 60 Mt/year, reducing CO<sub>2</sub> emissions by roughly 13 Mt/year. With Calera SCM replacing 20% of OPC in China, cement output would be reduced from 1.4 Gt/year (2009) to 1.12 Gt/year, resulting in a 246 Mt/year CO<sub>2</sub> emissions reduction [226].

#### 4.3. CarbonCure

A process developed by CarbonCure (Nov. Scotia, Canada) accelerates concrete curing and enhances strength by directly injecting CO<sub>2</sub> into the mixture (Fig. 12) [228–230]. Injection of CO<sub>2</sub> accelerated the hydration and strength of the concrete mix without altering its properties. The initial and final setting times were accelerated by 95–118 min (25% time reduction) and 103–126 min (23% time reduction), respectively [231]. The reaction of mature concrete with CO<sub>2</sub> results in shrinkage, decreased pH, and corrosion induced by carbonation. Fresh concrete does not undergo the same effects as carbonated concrete. The strength, absorption, chloride permeability, and freeze-thaw performance of precast or masonry concrete improved after exposure to early-age carbonation [232]. It is possible to reduce cement without

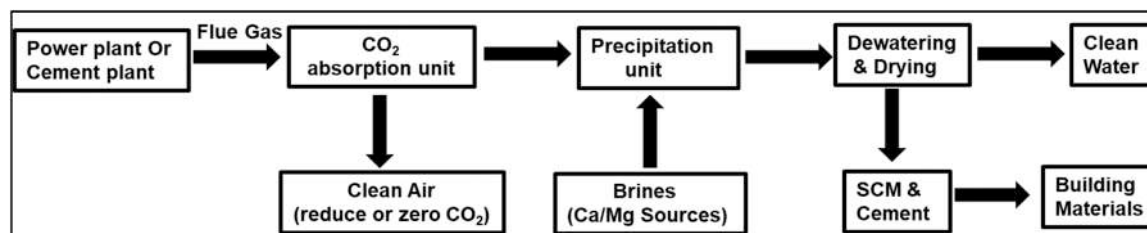


Fig. 10. Process flow diagram of Calera corporation technology [134].

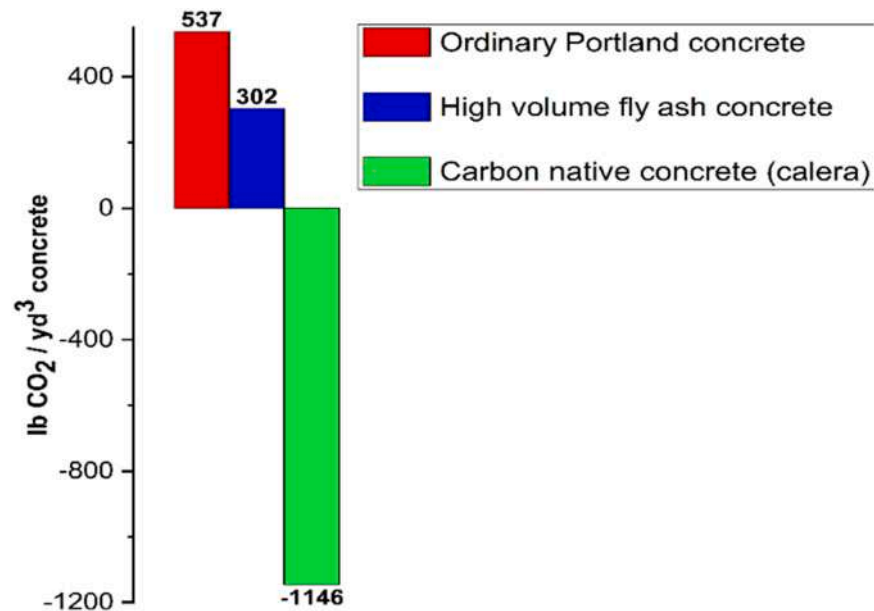


Fig. 11. Comparing concrete mixes lb. CO<sub>2</sub>/yd<sup>3</sup> of 100% OPC, 50% replacement by fly ash and 100% Calera aggregate to offset and sequester CO<sub>2</sub> [226].

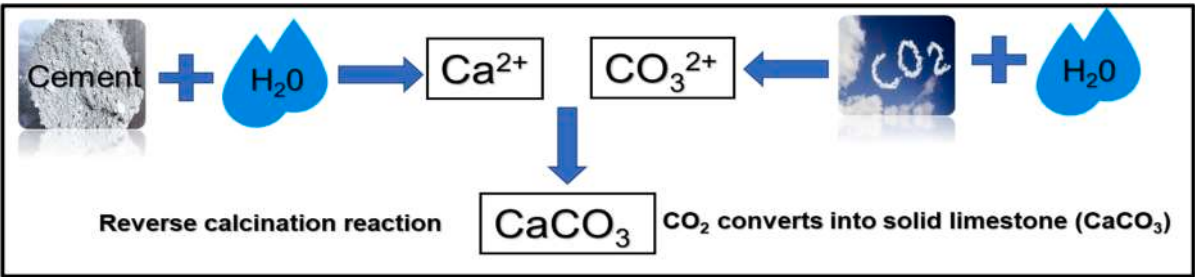


Fig. 12. Chemistry of CarbonCure process [228–230].

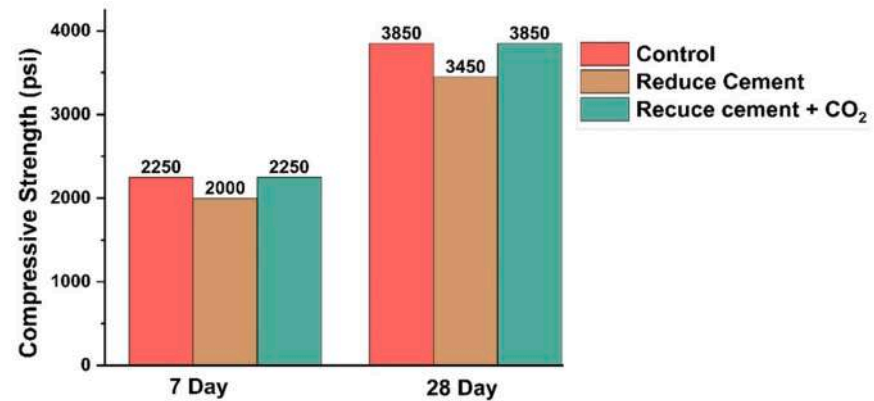


Fig. 13. Carbon Cure technology produces reduced cement content concrete without sacrificing the compressive strength [134].

Table 13  
CarbonCure mission by 2030 [233].

	CarbonCure for ready mix	CarbonCure for reclaimed water	CarbonCure for masonry & precast	CarbonCure for recycled aggregate	Total CO <sub>2</sub> reduced
CO <sub>2</sub> sequestered	4.2	65.3	0.4	95.8	166
Reduce cement impact	126.1	229.6	12.3	–	368
Net Impact	130.3	294.9	12.7	80.7	519

compromising its compressive strength through performance improvement by carbonation. Adding CO<sub>2</sub> to a concrete mix would restore compressive strength after a reduction of 7–8 % cement (Fig. 13), and the CO<sub>2</sub> emissions are also reduced by approximately 150 kg/m<sup>3</sup> [134]. Through CarbonCure, the concrete industry strives to reduce CO<sub>2</sub> emissions by 500 million tons per year by 2030 (Table 13) [233].

Several CarbonCure ready-mix concrete producers are located in Alberta; these plants produce 22 million m<sup>3</sup> of concrete with 8.3 million tons of CO<sub>2</sub> and save 354 million tons of CO<sub>2</sub> emissions. The Calgary Airport location, where 50.5 tons of CO<sub>2</sub> were utilized, produced 70,535 m<sup>3</sup> of concrete and avoided 425 tons of cement. Over 25 years, raised in concrete production to 55 million m<sup>3</sup>, the utilization of CO<sub>2</sub> was up to 21 million tons, and 884 million tons of CO<sub>2</sub> emissions were saved [234]. The life cycle assessment (LCA) of CarbonCure shows that this process reduces the net CO<sub>2</sub> emission of around 18 kg/m<sup>3</sup> of concrete (Table 14) [235].

#### 4.4. Solidia

In 2008, Solidia Technologies was founded in Piscataway, New Jersey, USA, a company offering green building materials that use CO<sub>2</sub> [236]. The company developed Solidia Cement and Solidia Concrete, which reduce net CO<sub>2</sub> emissions compared to traditional construction materials [237]. Solidia Cement is a nonhydraulic cement containing less lime minerals than ordinary cement, primarily composed of wollastonite (CaOSiO<sub>2</sub>) and rankinite (3CaO<sub>2</sub>SiO<sub>2</sub>). In contrast, ordinary cement is composed of alite (3CaOSiO<sub>2</sub>) and belite (2CaOSiO<sub>2</sub>) [238]. Since less lime is required per cement unit, less limestone must be calcinated, reducing emissions by approximately 30 % [239]. Solidia Cement clinker is manufactured at about 1200 °C, about 250 °C lower than Portland cement clinker, which is sintered at a temperature of 1600 °C. The lower calcination temperature reduces thermal fuel consumption, reducing emissions by 30 % [239]. Ordinary cement produces around 500 kg CO<sub>2</sub>/ton in process emissions and another 300 kg CO<sub>2</sub>/ton in thermal fuel combustion, totaling 800 kg CO<sub>2</sub>/ton. Solidia Cement, on the other hand, emits 550 kg CO<sub>2</sub>/ton (process & thermal) [240].

Concrete manufactured with Solidia Cement is characterized by its patented curing method and formula, consisting of fine and coarse aggregate, CO<sub>2</sub>, and Solidia Cement. Solidia cement reacts only with CO<sub>2</sub>, not with water, like ordinary cement. CO<sub>2</sub> interacts with Solidia Cement during curing to form thermodynamically stable calcium carbonate. Calcite polymorphs of CaCO<sub>3</sub> and silica gel are formed during the carbonation reaction and are crucial for developing strength within the concrete (Fig. 14) [238]. In contrast to ordinary concrete, Solidia Concrete cures in 24 h, saving cement and concrete producers time, money, and inventory space. During the curing process of Solidia concrete, it can sequester 290–310 kg of CO<sub>2</sub> per ton of Solidia cement used in Solidia concrete [240].

In comparison with the Solidia, carbonation of natural concrete over 40 years sequesters up to 48% of the process emissions from the cement used in the concrete (non-energy) (Fig. 15) [241]. As previously stated, 500 kg of CO<sub>2</sub> emissions per ton of ordinary cement, implying that natural sequestration will eventually sequester 240 kg of CO<sub>2</sub> per ton of cement used in the concrete, approximately the same amount as Solidia

during the curing process today (Fig. 15). [241]. The Solidia technology has shown it can save 1.5 Gt of CO<sub>2</sub>, save 3 trillion liters of fresh water every year, reduce the cement industry's energy consumption by 67 million tons of coal, and remove 100 million tons of landfills each year [237].

LafargeHolcim and Solidia Technologies signed a Joint Development Agreement in August 2013 and a Commercial Agreement in January 2015 to bring Solidia cement and concrete solutions to market. With a novel CO<sub>2</sub> curing method, they can reduce the overall carbon footprint of concrete by up to 70 % compared to conventional Portland cement concrete [242]. The LafargeHolcim group's Whitehall (USA) and Pecs (Europe) facilities conducted the first two non-hydraulic cement manufacturing campaigns. The first precast (in the United States) was manufactured using Solidia cement and concrete solutions. Durability testing and characterization are proceeding in compliance with PC requirements in the United States and the European Union [242].

#### 4.5. Blue planet

Blue Planet has developed and commercialized a scalable, economically and technically sustainable climate change mitigation solution. Blue Planet manufactures artificial fine and coarse aggregate by mineral carbonation, similar to the natural formation of ooids [243,244]. Demolished concrete, cement kiln dust, steel slag, fly ash, bauxite residue, and silicate rocks are calcium-rich geomass sources, which produce a carbon-sequestering coating aggregate that is 44% by mass CO<sub>2</sub> (Fig. 16) [245]. Blue Planet concrete's aggregate absorbs so much CO<sub>2</sub> in concrete that the whole structure becomes carbon-negative [245]. Each ton of CO<sub>2</sub>-sequestered limestone prevents the accumulation of 440 kg of CO<sub>2</sub>. The Blue Planet process reduces CO<sub>2</sub> emissions by 100 kg per ton of concrete. This equates to about 220 kg CO<sub>2</sub>/m<sup>3</sup> of concrete. Considering a cement content of 320 kg/m<sup>3</sup> of concrete, this results in a reduction of about 0.65 tons of CO<sub>2</sub> per ton of Portland cement, and would therefore compensate for the raw material CO<sub>2</sub> emissions of the cement production [246].

The company says employing this aggregate is the most efficient method of providing carbon-neutral concrete. A cubic yard of standard Portland cement-based concrete has a carbon footprint of around 600 pounds of embodied CO<sub>2</sub>. Reducing the cement component of a mixture with SCM (supplementary cementitious material, such as fly ash) reduces the carbon footprint by just 300 lb/yard<sup>3</sup>. By substituting traditional fine and coarse aggregate with Blue Planet synthetic limestone aggregate, it is possible to offset the carbon footprint of Portland cement by 1320 lb/yard<sup>3</sup>. If the Portland cement in the mixture comes from a cement kiln where Blue Planet has collected CO<sub>2</sub>, the normal 600-pound carbon footprint of cement in the mix is excluded from the computation. The total CO<sub>2</sub> offset per cubic yard of concrete is 1320 pounds of industrial or atmospheric CO<sub>2</sub> sequestered in aggregate and 600 pounds captured during Portland cement manufacturing, resulting in the total 1920 lb CO<sub>2</sub> offset per cubic yard of concrete (Fig. 17) [247,248].

San Francisco Bay Aggregates is planning and constructing the first commercial plant using Blue Planet Systems' patent-protected carbon mineralization process. The plant will gather CO<sub>2</sub> from the nearby Los Medanos Energy Center and permanently deposit it in synthetic limestone aggregate via mineralization. SF Bay Aggregates is at the forefront of developing this paradigm-shifting technology and will provide carbon-negative aggregate to Bay Area projects by 2022 [249]. Blue Planet technology was used to construct a temporary boarding area at San Francisco International Airport. MC and Blue Planet will perform a feasibility study on potential Silicon Valley use until the fiscal year 2021, following which the partners hope to make the technology commercially accessible [250]. Blue Planet has launched a pilot program and Sulzer to reduce CO<sub>2</sub> emissions and reshape the cement industry sustainably. Sulzer Chemtech, the industry leader in separation and mixing technology, is creating an efficient and effective carbon capture unit at Blue Planet's pilot plant, which is now under construction in Pittsburg,

**Table 14**

Life cycle assessment of concrete production by CarbonCure [235].

CO <sub>2</sub> emission	g CO <sub>2</sub> /m <sup>3</sup> concrete
Gas processing & transport	55.5
Equipment production & operation	9.3
Avoided from Materials transport	-123.6
CO <sub>2</sub> sequestered	-289.1
Avoided from cement reduction	-17584.8
<b>Total</b>	<b>-17932.7</b>

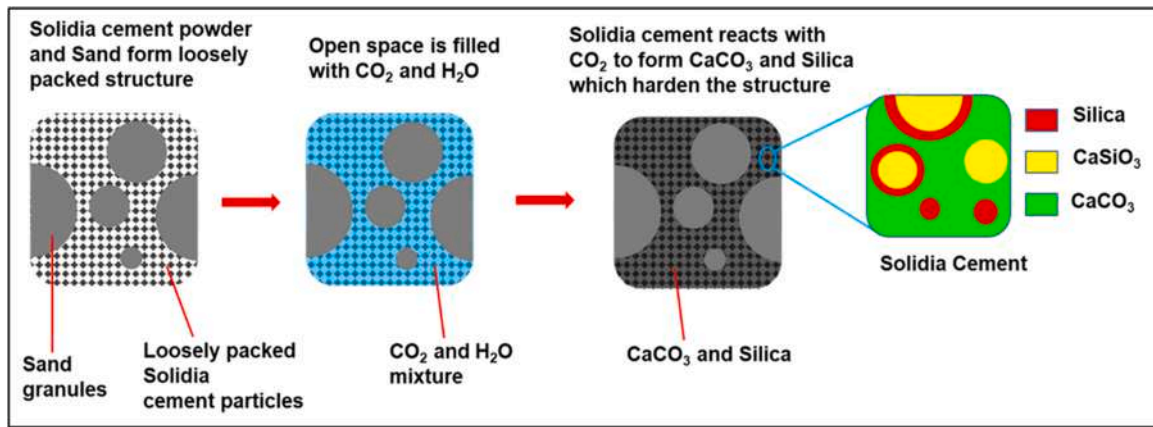


Fig. 14. Mechanism of carbonation in Solidia concrete [238].

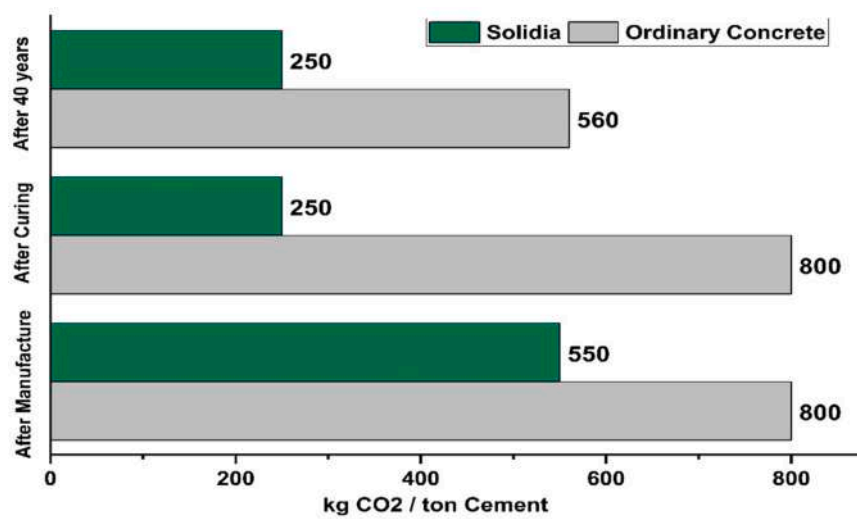
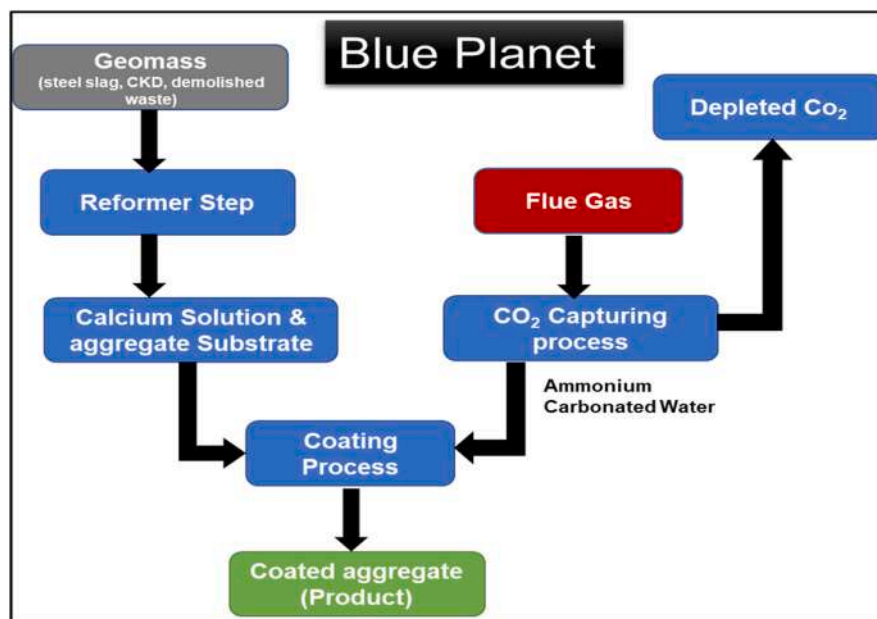
Fig. 15. Comparing CO<sub>2</sub> emissions from ordinary concrete and Solidia at different stages of concrete life (net thermal + process) [241].

Fig. 16. A flow chart showing the process of manufacturing coated aggregates [245].



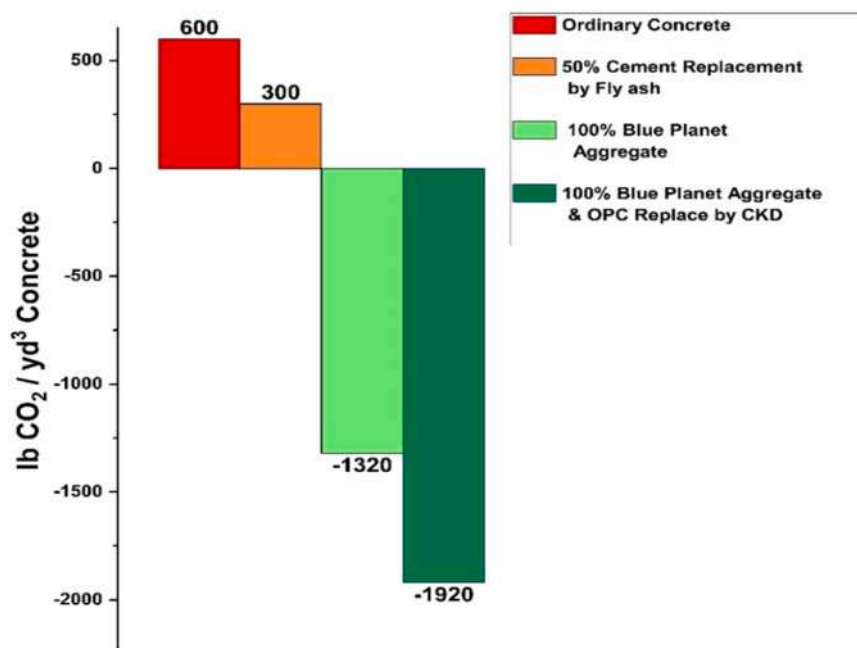


Fig. 17. Comparing four concrete mixes shows the consequences in lb. CO<sub>2</sub>/yd<sup>3</sup> of using different formulations to offset and sequester CO<sub>2</sub> [252,253].

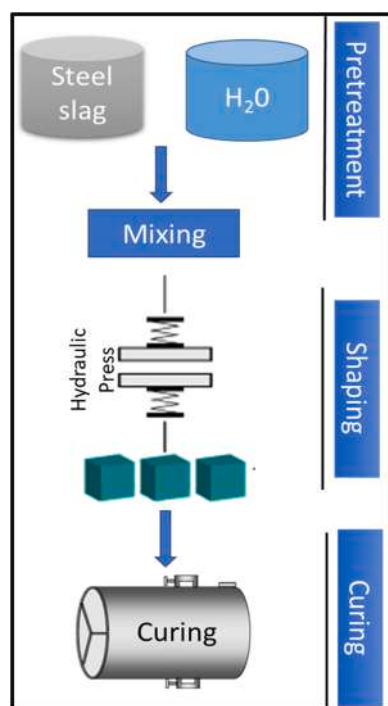


Fig. 18. Schematic representation of the accelerated carbonation of stainless-steel slag [252,253].

California, United States, and will capture emissions from a nearby natural gas-fired power plant. The plant will use Sulzer Chemtech's technology to achieve high CO<sub>2</sub> absorption performance with minimal energy usage [248,251].

#### 4.6. Carbstone

In the 1990s, the slag was dumped after being separated from the ore. Nowadays, it is converted into bricks by carbonation. Slag from steel mills is rich in magnesium and calcium silicates, which convert to

high-quality products when reacted with captured CO<sub>2</sub> (Fig. 18) [252, 253]. The steel industry in Belgium generates approximately 500,000 tons of steel slag per year [254]. The recycling company Orbix discovered in 2004 that CO<sub>2</sub> and H<sub>2</sub>O could harden the finest steel slag particles. Orbix, a company specializing in slag processing in the steel industry, recovers residual steel. Carbonation (Fig. 18) involves three steps: pre-handling of slag; forming the building blocks with a hydraulic press at a pressure between 75 and 609 kg/cm<sup>2</sup> and subsequent compacting to achieve the desired porosity; and diffusion of CO<sub>2</sub> into slag at high temperatures and pressures (0.5–10 MPa) in autoclaves [211,255, 256]. In this reaction, CO<sub>2</sub> is reacted with calcium silicate to form calcium carbonate, an effective substitute for cement (a binding material in the building block) that effectively sequestered CO<sub>2</sub> for a long time [257]. As an alternative to conventional concrete-making, this block has a negative carbon footprint (200 g CO<sub>2</sub>/kg slag less than conventional concrete-making) [258].

A patented process developed in partnership with the Flemish Institute for Technological Research (VITO) led to the founding of Carbstone Innovation in 2009. Although the carbonation process is straightforward, it still took us almost ten years to refine the basic technology. Towards the end of 2013, a pilot plant was built in Farciennes, near Charleroi, to produce large blocks. With this facility in Farciennes - Walloon Municipality, Slag from various sources will be processed and used to produce high-value construction products through carbonation [256]. In 2016, Carbstone Innovation signed its first deal with RuwBouwGroep CRH of the Netherlands, which produces the stones in its factories. Since the market for building materials is highly competitive, they are partnering with a company that can bring the carbstone to market in the most efficient possible way. In 2017, carbstone aimed to set up a production installation for the Belgian market. At the same time, it developed a license to commercialize the technology globally [256].

#### 5. Future challenges

Over the past 30 years, the technology of CO<sub>2</sub> sequestration by mineral carbonation has developed significantly. The fundamental characteristics of mineral carbonation have been discovered to acquire mineral carbonation. The carbonization efficiency can be improved by

varying various parameters, such as high CO<sub>2</sub> pressure, high temperature, fine particle size, and adding a catalyst. Despite the apparent advantages of mineral carbonation over other geological CO<sub>2</sub> storage technologies, the following research gaps need to be addressed:

1. Reducing the total emission of CO<sub>2</sub> is one of the most important goals that must be addressed broadly.
2. Additional energy consumption during mineral carbonation leads to direct or indirect emissions of CO<sub>2</sub>, which must be addressed while calculating the CO<sub>2</sub> sequestration in a particular process.
3. The life cycle assessment technique should be introduced to evaluate mineral carbonation and optimization.
4. The mineral carbonation mechanism has not yet been precisely established. Further study of mineral structural transitions and phases during CO<sub>2</sub> mineral carbonation is possible with the development of in-situ tools such as XRD and IR techniques.

## 6. Conclusions

The purpose of this paper was to review CO<sub>2</sub> capture technologies in the cement industry and the utilization of CO<sub>2</sub> in the construction industry. In addition, the number of commercialized CO<sub>2</sub> sequestration technologies was discussed. The following are the main conclusions:

1. Based on the current climate situation, the development of different methods for mitigation and adaptation is essential to meet the 1.5 °C targets by the end of the century.
2. CO<sub>2</sub> capture methods used in the cement plant included amine scrubbing with \$84.1/t CO<sub>2</sub> and oxy-combustion with \$44.1/t CO<sub>2</sub>, roughly half that of MEA scrubbing. Calcium looping in the tail end and integrated configurations were approximately \$54.6/t CO<sub>2</sub> and \$59.7/t CO<sub>2</sub>, respectively.
3. Mineral carbonation is one of the most promising technologies for reducing CO<sub>2</sub> emissions. The process has the double benefit of reducing CO<sub>2</sub> emission and utilizing solid wastes in the carbonation to produce certain value-added products.
4. Indirect carbonation using extracting agent NH<sub>4</sub>Cl is better for CO<sub>2</sub> sequestration than extracting agents such as HCl, HNO<sub>3</sub>, and CH<sub>3</sub>COOH. An advantage of using NH<sub>4</sub>Cl is that it is easily regenerated and does not need basic reagents during the precipitation of calcium carbonate.
5. CO<sub>2</sub> uptake by mineral carbonation of different cement-based materials was 7–25 %. Both hydration products (Ca(OH)<sub>2</sub> and C-S-H) and unhydrated cement clinker (C<sub>3</sub>S and C<sub>2</sub>S) react with CO<sub>2</sub>. The uptake of CO<sub>2</sub> by  $\gamma$ -C<sub>2</sub>S (non-hydrated) was higher than by  $\beta$ -C<sub>2</sub>S. On the other hand, C<sub>3</sub>A had a minimal reactivity with CO<sub>2</sub>, and the study of C<sub>4</sub>AF with CO<sub>2</sub> is still unavailable in the literature.
6. Carbonation of RA has significantly reduced water absorption, porosity and crushing value while increasing the apparent density. Concrete prepared with CRA has better compressive strength and elastic modulus, with increased solid phase volume by 11.8 % than untreated RA. RA has a particle size between 5 and 20 mm sequestrates, almost 7.9 Kg CO<sub>2</sub> per ton.
7. Carbonation is maximum in those solid wastes with a higher concentration of CaO content (but the phase of the mineral is also important in which calcium is bonded), more surface area and optimum relative humidity of 50–70 %. Carbonation decreased the pH of solid waste from the threshold value (11.5), making them less hazardous.
8. Currently, waste-derived construction products are cost-competitive on the market. Different companies have successfully produced construction materials from different solid wastes by mineral carbonation. Artificial aggregates completely replace the use of natural stone in a variety of construction applications.

## Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Mohd Hanifa reports financial support was provided by University Grants Commission. Usha sharma reports financial support was provided by Council for Scientific and Industrial Research.

## Data availability

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

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