

Review Article

Contents lists available at ScienceDirect

Journal of CO2 Utilization



Emerging CO₂ utilization technologies for construction materials: A review

Ning Li^a, Liwu Mo^{b,c}, Cise Unluer^{a,*}

^a School of Engineering, University of Glasgow, Glasgow G12 8LT, United Kingdom ^b College of Materials Science and Engineering, Nanjing Tech University, Nanjing 211816, China

^c State Key Laboratory of Materials-Oriented Chemical Engineering, Nanjing 211816, China

ARTICLE INFO ABSTRACT Keywords: The construction industry is a major contributor of CO₂ emissions. Carbonation, involving the reaction of CO₂ Construction materials with alkaline reactants, immobilizes CO₂ into thermodynamically stable carbonates used in construction mate-Carbonation technologies rials such as concrete and aggregates. The utilization of CO2 in construction materials is considered as one of the CO₂ utilization most promising routes for carbon sequestration, with a \$400 billion market opportunity and a potential to reduce Challenges annual CO_2 emissions by up to 3 Gt by 2030. This paper reviews the current status of the utilization of CO_2 in Recommendations construction materials from the perspective of scientific research and commercial applications. The explanation of the fundamental carbonation reaction mechanisms was extended to cover different binder systems involving Portland cement, non-hydraulic calcium silicate, industrial solid wastes and magnesium-based materials. Factors affecting the kinetics of the carbonation reaction and properties of the final products were reviewed. Furthermore, the current state of research and commercial initiatives involving the utilization of CO₂ in the production of various building components were presented. Finally, key issues regarding the challenges faced in the scaling up of CO2 utilization technologies from the perspective of academia, industry and relevant regulatory bodies

were highlighted. Recommendations to address the current utilization dilemma and promote large-scale application of CO₂ in the production and development of construction materials were provided.

1. Introduction

With more than 30 billion tonnes of carbon dioxide (CO₂) emissions into the atmosphere every year, global climate change has led to various environmental problems that need immediate attention. Carbon capture, utilization and storage (CCUS) technologies will play a key role in mitigating these adverse effects and achieving the upper temperature rise target of 1.5 °C as a part of the Glasgow Climate Pact set at the 2021 United Nations Climate Change Conference (COP26) [1,2]. As a subsequent step to its capture, the utilization of CO₂ can involve its conversion into high-value products (synthetic fuels, polymers, etc.) or thermodynamically stable products (carbonate minerals) for long-term storage through thermochemical, electrochemical, photochemical or biological processes. Converting CO2 into solid minerals can permanently store CO₂, thereby preventing its release into the atmosphere. While geological and marine storage have been ongoing for the last few decades [3], mineral sequestration has also shown great potential, particularly by accelerating the carbonation of hardened cement and concrete.

Carbonation can be applied to fresh and hardened Portland cement

(PC) concrete as a part of the curing process (i.e. within the first hours after casting and long-term curing). Accelerated carbonation reaction is a strongly exothermic and diffusion-controlled process and is influenced by material properties (e.g. physical properties and chemical composition) and carbonation conditions (e.g. CO₂ concentration, pressure and temperature) [4]. The chemical reaction that occurs between CO₂ and calcium-based compounds in cement is exothermic, presenting advantages such as: (i) rapid strength enhancement, (ii) enhanced durability, and (iii) permanent sequestration of CO₂ [5,6]. Furthermore, carbonation can be used to enhance the quality of recycled concrete aggregates (RCAs) and promote the strength development of artificial aggregates (AAs) [7-9]. These outcomes can enable the building industry to significantly reduce CO₂ emissions in the long-term, reducing its impact on global warming and the environment in general [10]. However, the commercial utilization of CO₂ in construction materials and associated technologies is still in the early development stage [11], currently being limited to prefabricated products, such as concrete blocks [10]. These advances need to be supported by further research initiatives to overcome the barriers to large-scale implementation [12].

https://doi.org/10.1016/j.jcou.2022.102237

Received 26 July 2022; Received in revised form 14 September 2022; Accepted 17 September 2022 Available online 1 October 2022 2212-9820/© 2022 The Author(s). Published by Elsevier Ltd. This is an open access article under the CC BY license (http://creativecommons.org/licenses/by/4.0/).

^{*} Corresponding author. E-mail address: Cise.Unluer@glasgow.ac.uk (C. Unluer).

With over 25 billion tons of concrete [13] and 40 billion tons of mineral aggregates [14] produced each year, the sheer size of the building materials market has led to mineral carbonation being recognized as one of the largest and most energy efficient ways to utilize CO₂. The growing demand for green building materials is expected to further drive the market for the production of construction materials through mineral carbonation using waste CO₂. In recent years, numerous studies, reviews and books have been published to demonstrate the mechanism of carbonation and optimize the reaction process for a wide range of alkaline solids [15–18]. However, the utilization of CO₂ in construction materials within commercial applications has not been recently presented in an extensive review. This paper reviews the recent advances in academic and commercial initiatives involving the accelerated carbonation of construction materials including concrete, aggregates, blocks and their derivatives. The fundamental carbonation mechanisms and processes, and factors affecting carbonation kinetics and environmental and economic impact are also discussed in detail. This work aims to elucidate the technical and industrial benefits of accelerated carbonation and its application as a means of reducing CO₂ emissions in different construction materials. The discussion also highlights the main advances made in recent years, the challenges faced and potential solutions to enable the large-scale application of carbonation technologies. Ultimately, a number of recommendations were provided to address the current issues associated with the commercial utilization of this technology in the building sector, with the goal of paving the way for the direction of future research in this area and providing guidance on the application of CO₂ sequestration in the production of construction materials with the desired sustainability and cost.

2. Fundamental carbonation mechanisms and processes

Carbonation usually refers to the reaction of alkaline solids, such as a Ca- or Mg-rich materials, with CO_2 in a fluid state or solution. These reactions can occur at ambient temperatures and pressures. The basic element dissolves from the precursor solid, CO_2 dissolves into the liquid phase (e.g. water), and the carbonate precipitates from the supersaturated solution. In a Ca-based system, a variety of calcium carbonate (CaCO₃) crystals are formed, such as amorphous, vaterite, aragonite, and calcite. Of these, calcite is the polycrystal that forms in equilibrium the most frequently [19], although the formation of one or more polycrystals can be directed by controlling the reaction conditions [20,21]. Magnesite and hydrated magnesium carbonate phases can also form in the presence of Mg, albeit the conditions for their formation are often less favorable than the formation of their Ca-bearing variants [22].

This method of producing CaCO₃/MgCO₃ is a fundamental approach that is used in the carbonation of concretes and aggregates. Reactants such as PC, non-hydraulic calcium silicates, industrial solid wastes, and magnesium-based cementitious materials can be used to carbonate. Within these systems, the accessibility and mobility of Mg or Ca (i.e. avoiding surface passivation), other than their abundance in the matrix, are crucial factors in determining the suitability of a given substrate for carbonation treatment and the final carbonation degree [23]. Focusing on different factors that control carbonation, this section reviews the fundamental carbonation reaction mechanisms and processes in these different systems.

2.1. Portland cement (PC)

The content of calcium silicates (C_3S and β - C_2S) in cement clinker exceeds 60%. The unhydrated C_3S and β - C_2S and their hydrate products (e.g. portlandite/calcium hydroxide (Ca(OH)₂) and calcium silicate hydrate (C-S-H) gel) can react with CO₂ and water to generate CaCO₃, as shown in Eqs. (1)–(4). Even in the presence of water, there is a competition between hydration and carbonation, with hydration being the preferred pathway [24].

(4)

$$Ca_3SiO_5 + 3CO_2 + nH_2O \rightarrow 3CaCO_3 + SiO_2 \bullet nH_2O \tag{1}$$

$$\beta - Ca_2SiO_4 + 2CO_2 + nH_2O \rightarrow 2CaCO_3 + SiO_2 \bullet nH_2O$$
⁽²⁾

$$Ca(OH)_2 + CO_2 + H_2O \rightarrow CaCO_3 + 2H_2O \tag{3}$$

$$Ca_{x}Si_{y}H_{z}O_{(x+2y+z/2)} + xCO_{2} \rightarrow xCaCO_{3} + y(SiO_{2} \bullet tH_{2}O) + (z/2 - yt)H_{2}O$$

During the hydration of cement, the anhydrous cement component dissolves in water and forms a solid hydrate phase. As a result, the volume of the solid material increases, leading to a reduction in porosity and the formation of hardened material [25]. Carbonation reaction in a cementitious matrix is more complex than hydration because it requires the transport of CO₂ and interaction with existing cementitious phases. During carbonation, gaseous CO2 diffuses into concrete, dissolves into the pore solution and reacts with the dissolved cementitious phases, mainly forming CaCO₃ and silica gel [26], as shown in Fig. 1. Similar to hydration, these processes lead to an increase in solid volume and thus a reduction in porosity and the development of mechanical properties. However, because of water saturation and fine porosity of hydrated PC, the carbonation of PC-based systems progresses slowly under ambient conditions due to CO₂ diffusion being hampered [27]. Despite this slow rate, if given enough time, the surfaces of concrete structures exposed to CO₂ can act as a CO₂ sink [28–30]. Furthermore, concrete construction and demolition waste that is crushed in the form of fine particles can achieve significant, albeit unintentional, CO₂ uptakes.

2.2. Non-hydraulic calcium silicates

Compared to highly hydraulic C_3S , the hydration rate of nonhydraulic calcium silicates (e.g. γ -C₂S, C_3S_2 and CS, with a molar ratio of CaO/SiO₂ \leq 2) is relatively slow [32,33], involving the formation of lower amounts of portlandite. In the presence of water and high CO₂ concentrations, the formation of carbonates is observed, following Eqs. (2), (5) and (6). This reaction, which usually takes place under conditions similar to an autoclave, can be utilized to carbonate a variety of non-hydraulic calcium silicates, such as belite (2CaO·SiO₂), rankinite (3CaO·2SiO₂) and wollastonite (CaO·SiO₂). When non-hydraulic silicates are carbonated in this way, a mixture of CaCO₃ and amorphous silica is formed, which can act as binding agents [34,35]. However, in order to achieve good reaction kinetics, the carbonation of these systems needs to be performed by concentrated CO₂; otherwise, the kinetics are usually too slow for practical development.

$$\gamma - Ca_2SiO_4 + 2CO_2 + nH_2O \rightarrow 2CaCO_3 + SiO_2 \bullet nH_2O$$
(5)



Fig. 1. Schematic diagram showing the carbonation curing of cementitious materials [31].

$$CaSiO_3 + CO_2 + nH_2O \rightarrow CaCO_3 + SiO_2 \bullet nH_2O \tag{6}$$

Jang and Lee [36] cured belite cement mortars (β -C₂S in the range of 16–48%) at a low CO₂ concentration of 5% for 28 days and found that the cement with the highest β -C₂S content revealed the highest CO₂ sequestration of 16.9%. Carbonate precipitation not only occurs within the capillaries containing pore water, but also promotes the densification of the interfacial transition zone (ITZ) between hydrated C-S-H and β -C₂S particles. Mu et al. [37] investigated the carbonation curing of γ -C₂S as a building material and reported that the mechanical strength and surface hardness of carbonated γ -C₂S correlated highly with the degree of carbonation, and that the carbonation products of γ -C₂S were dominated by calcite with aragonite as a secondary product.

The hydration inert calcium silicate (CaSiO₃, CS) is considered to be an extremely promising raw material for carbonation due to its excellent reactivity and fast reaction in high temperature (100–250 °C) liquid phase carbonation processes [38]. However, purely hydrated inert materials are unable to develop strength at an early hydration stage and therefore require specific compression molding equipment with corresponding limitations in transport and pre-curing processes. Accordingly, the use of mixtures of CS with other hydraulic silicate minerals (especially C₃S and β -C₂S) are preferred in terms of carbonation. However, differences in reaction rates, crystalline phases and product structures of different components may have an impact on the overall carbonation process, and thereby the microstructural and mechanical development of these mixes. Therefore, the interaction mechanism of various calcium silicate components in a carbonation environment needs further research.

2.3. Industrial solid wastes

Industrial solid wastes such as steel slag, mine tailings, fly ash, cement kiln dust, and air pollution control residues constitute byproducts of metal processing and mining, coal combustion, PC production, and waste incineration, respectively. Each of these industrial wastes contains an intrinsic alkalinity (e.g. Ca, Mg, Al, Si, and alkali) that can be carbonated by reacting with CO₂. Fig. 2 shows the mechanism of the accelerated carbonation of these materials, indicating that carbonation reactions occur in four pathways: (i) transport-controlled mechanisms such as CO_2 and Ca^{2+} diffusion to/from reaction sites; (ii) boundary layer effects (diffusion of particles over precipitation coatings); (iii) dissolution of $Ca(OH)_2$ on the particle surface; (iv) pore plugging; and (v) precipitation coatings. The major challenge hindering



Fig. 2. Proposed mechanism of the accelerated carbonation reaction of alkaline solid wastes [39].

the large-scale use of industrial wastes for carbonation is their slow conversion rate associated with the heterogeneity of these wastes (i.e. most of the Ca and Mg are not present in pure form and are combined with silicates or another complex oxide phase). Therefore, most research in this field has focused on identifying reaction pathways by bench-scale experiments for determining reaction rates, as well as characterization of the mineralogy of the reactants and products.

The maximum CO₂ fixation capacity of industrial solid wastes depends on their alkalinity content and different operating parameters associated with the carbonation process [40]. Assuming all the Ca and Mg present in industrial solid wastes can react with CO₂, Table 1 summaries the main industrial wastes suitable for mineral carbonation and their theoretical CO₂ fixation capacity. The Ca-rich fly ash (Class C FA) carbonates quickly, is strong enough to be used for structural construction [23], and therefore has a higher potential for CO₂ sequestration than Ca-poor fly ash (Class F FA). The CaO content of steel slag is relatively high (23-60%), depending on the steelmaking process [41]. Mo et al. [42] reported that the compressive strength of steel slag pastes carbonated at 0.1 MPa CO₂ for only 1 day (22.4 MPa) was significantly higher than that of steel slag pastes cured under ambient conditions for 28 days (7.8 MPa). The CO₂ uptake of these carbonated steel slag mixes was in a range of 11.2–17.6% [42]. Alternatively, magnesia slag is a by-product of magnesium metal production and consists mainly of different forms of calcium silicate and periclase [43]. The compressive strength of magnesia slag pastes after 14 days of CO2 curing was 119.5 MPa, which was 6 times higher than the strength before CO₂ curing (19.8 MPa) [43]. The corresponding CO₂ uptake after 14 days of CO₂ curing was 15.6% [43].

Municipal solid waste (MSW) incineration process generates large amounts of waste incineration bottom ash (MSWIBA) [44]. Zhang et al. [45] developed an eco-cement, which was synthesized from 94% MSWIBA and 6% Ca(OH)₂ at an incineration temperature of 1100 °C. The eco-cement paste showed a significant increase in early strength (47.3 MPa at 1 day) under carbonation curing (99.5% pure CO₂ gas under a pressure of 0.069 MPa for 10 h). Sharma et al. [46] found that the compressive strength of cement mortars with 20% cement kiln ash admixture after 12 h of carbonation curing and 3–28 days of subsequent hydration was 17–35 MPa, which increased the early strength by 30%. Within these systems, the longer periods of subsequent hydration may reduce the production efficiency of concrete and increase energy consumption during the curing, and therefore be optimized depending on the application.

In addition to the materials mentioned above, other solid wastes such as contaminated sediment [52], upcycling wood waste [53], and recycled glass [54] are also used in the context of carbonation curing. Recent studies involve the combination of different materials in the production of various building components. For instance, Wang et al. [52] used PC and MgO to convert contaminated sediments into eco-friendly sediment blocks through CO₂ curing. Hossain et al. [53] produced cement-bonded particleboard (CBPs) by CO2 curing recycled wood aggregates and light-burned MgO cement. Cabral et al. [55] also manufactured cement-bonded balsa particleboards involving the use of balsa wood under acceleration carbonation (24 h in a CO₂ concentration of 15%), resulting in a product that met the ISO 8335 requirements. Guo et al. [54] prepared cement mortars using 100% recycled glass fine aggregates cured under flow-through CO₂ curing (100% purity at a flow rate of 1 \pm 0.1 L/min). The 9-day compressive and flexural strengths of these samples were 174.4% and 118.8% of those subjected to water curing, respectively.

2.4. Mg-based cementitious materials

There has been a rising interest in the use of MgO as an alternative binder for the carbonation curing of cementitious composites. Reactive MgO has a high CO_2 sequestration capacity and a low calcination temperature (700–1000°C for MgCO₃ versus 1450°C for CaCO₃) [56], with

Table 1

Main indus	trial wastes	used for mineral	carbonation a	nd their CO ₂	fixation efficiency.
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Waste	Alkaline content (%)		Major compounds for carbonation	Theoretical CO_2 fixation capacity (t	
	CaO	MgO		CO ₂ /t waste)	
GGBS [47]	38–42	1–13	gehlenite (Ca ₂ Al ₂ SiO ₇), akermanite (Ca ₂ MgSi ₂ O ₇)	0.31–0.45	
Class F FA [47]	1.5-5	0.6-2.0	lime (CaO), portlandite (Ca(OH) ₂), ettringite (Ca ₆ Al ₂ OH ₁₂ (SO ₄) ₃ ·26H ₂ O)	0.02-0.07	
Class C FA [48]	15-50	4.0-10	lime (CaO), portlandite (Ca(OH) ₂), ettringite (Ca ₆ Al ₂ OH ₁₂ (SO ₄) ₃ ·26H ₂ O)	0.06-0.21	
Steel slag [47]	40-60	5–15	larnite (Ca ₂ SiO ₄), brownmillerite (Ca ₂ FeAlO ₅), lime (CaO), ettringite	0.37-0.64	
			(Ca ₆ Al ₂ OH ₁₂ (SO ₄) ₃ ·26H ₂ O), portlandite (Ca(OH) ₂)		
Magnesium slag	42.5-54.5	4.4-17.2	beta-larnite (Ca ₂ SiO ₄), hatrurite (Ca ₂ SiO ₅), γ-dicalcium silicate (Ca ₂ SiO ₄), periclase	0.13-0.36	
[43,49]			(MgO), lime (CaO)		
MSWIBA [50]	16.3	2.6	gehlenite (Ca ₂ Al(AlSiO ₇)), portlandite (Ca(OH) ₂), ettringite (Ca ₆ Al ₂ OH ₁₂ (SO ₄) ₃ ·26H ₂ O)	0.005-0.01	
Cement kiln dust	34.5-46.2	1.1 - 2.6	lime (CaO), portlandite (Ca(OH) ₂), calcium silicates, gehlenite (Ca ₂ Al(AlSiO ₇))	0.09-0.12	
[51]					

the potential to be obtained from alternative routes such as seawater or reject brine [57,58]. The manufacturing process of carbonated reactive magnesia cement-based components is based on the carbonation curing of a Mg-based binder, where the carbonation process is preceded by the hydration of MgO to form magnesium hydroxide, also known as brucite (Mg(OH)₂). Because of the volume expansion associated with the production of Mg(OH)₂, the hydration of MgO in cement-based mixes reduces pore volume. Unlike PC-based systems, the hydrate phase, brucite, have a porous structure and does not contribute much to strength development. However, its subsequent carbonation leads to the formation of a range of hydrated magnesium carbonates (HMCs) such as nesquehonite (MgCO₃0.3H₂O), artinite (Mg₂CO₃(OH)₂0.3H₂O), hydromagnesite (Mg₅(CO₃)₄(OH)₂0.4H₂O), dypingite and $(Mg_5(CO_3)_4(OH)_20.5H_2O)$, as shown in Eqs. (7)–(11).

$$MgO + H_2O \rightarrow Mg(OH)_2 \tag{7}$$

$$Mg(OH)_2 + CO_2 + 2H_2O \rightarrow MgCO_3 \bullet 3H_2O \tag{8}$$

$$2Mg(OH)_2 + CO_2 + 2H_2O \rightarrow Mg_2CO_3(OH)_2 \bullet 3H_2O$$
(9)

$$5Mg(OH)_2 + 4CO_2 \rightarrow Mg_5(CO_3)_4(OH)_2 \bullet 4H_2O \tag{10}$$

$$5Mg(OH)_2 + 4CO_2 + H_2O \rightarrow Mg_5(CO_3)_4(OH)_2 \bullet 5H_2O \tag{11}$$

3. Factors affecting the kinetics of the carbonation reaction

The reaction kinetics and the total CO₂ uptake of these mineral carbonation techniques are influenced by a number of factors. As in typical fluid-solid reactions, the main influencing factors involve precursor properties and exposure conditions, as detailed in Table 2. The initial stages of the carbonation process are determined by the surface area and particle size of the reactants, which affects the reaction kinetics. Although fine particles may accelerate reaction kinetics, the need for grinding can make their manufacturing energy intensive. Another factor to consider is the solubility of CO2, which decreases with increasing temperature, thereby reducing the concentration of CO2 in the liquid. However, high temperatures can promote CO2 diffusivity and accelerate the leaching of Ca from solid particles. Hence, the temperature at which carbonation reaction is carried out needs to be considered and controlled. Finally, the carbonation reaction is sensitive to the presence and state of moisture (e.g. liquid or vapor), which may be present or freed during carbonation. This is because, whereas water is needed to accelerate carbonation processes on mineral surfaces, it slows down CO2 diffusive transport (and thus carbonation kinetics) when it condenses within the pores. Therefore, it is critical to control liquid water saturation levels within cementitious microstructures, such as when carbonation is performed in precast components.

4. Carbonation products, process and performance

The utilization of CO2 in construction materials is considered as a

Table 2

Summary	of	factors	influe	ncing	mineral	carbonation

Influencing factor		Findings
Exposure conditions	CO ₂ environment Temperature	 Carbonation can be carried out at low and high pressures, under vacuum and supercritical (scCO₂) conditions. Higher CO₂ concentrations and pressures can lead to higher carbonation rates. However, excess CO₂ concentration/ pressure does not necessarily lead to improved mechanical performance [34]. Higher temperatures are beneficial for the
		 leaching of Ca from solid particles, but slow down CO₂ solubility. The uptake of CO₂ increases with increasing temperature up to 60 °C (at atmospheric pressure) [59]. More stable polymorph (calcite) is formed at low temperatures (0–10 °C) [60].
	Relative humidity (RH)	 Lower RH inhibits carbonation kinetics, while higher RH slows down CO₂ diffusion. RH values in the range of 50–70% could be appropriate for the carbonation of PC-based mixes [61,62].
Precursor properties	Surface area	 Higher surface areas lead to greater extent of carbonation, but can decrease the overall energy efficiency.
	Composition	 Different materials of hydraulic, pozzolanic or lime-bearing nature, or other CO₂-reactive calcium or magnesium-containing material heavy metals can be used. Higher concentrations of Ca/Mg in the composition could lead to increased carbonation degrees [58,63]. The presence of ferrite/C₃A phases is important for the formation of ettringite, which in the presence of CO₂ decomposes to form gypsum, calcium carbonate and alumina gel [64].
	Porosity	 Low porosity impedes CO₂ diffusion between particles [65] and is often related to high degrees of compaction.
	Free water content	 Some water is necessary for the carbonation reaction, while higher water contents inhibits the diffusion of CO₂ [66]. Precuring can control the moisture content and subsequently enhance carbonation [67].

promising carbon utilization initiative [68]. Bobeck et al. [69] reported that there would be a \$550 billion market opportunity in this area by 2030 (\$400 billion for concrete and \$100 billion for aggregates), which has a great potential to reduce annual CO₂ emissions by up to 5 billion tons (1.4 billion for concrete and 3.6 billion for aggregates). Mineral carbonation can be used in the production of aggregates and cementitious materials, which can replace natural and existing synthetic sources constituting important components of common building materials. This

section reviews the production process and properties of concrete and aggregates obtained through the carbonation route.

4.1. Concrete

Concrete is one of the most widely used industrial products. It is estimated that around 11.5 billion tonnes of concrete are consumed worldwide each year, which is expected to reach 18 billion tonnes per year by 2050 [70]. Hence, carbonate bonded concrete is regarded as one of the main CO_2 utilization routes within the building industry.

The introduction of CO_2 into cement-based formulations can be achieved by either injecting CO_2 into fresh concrete over a short period of time, or exposing prefabricated components to gaseous CO_2 in a reactor, in dilute or concentrated form, over a period of several hours or days. Due to the variations in the application process, these pathways lead to different levels of CO_2 uptake. For instance, the CO_2 uptake of fresh concrete subjected to CO_2 injection is limited by the overall reaction rate of injected CO_2 with Ca and Mg, which also depends on the solubility of CO_2 in the alkaline aqueous solution (i.e. $\leq 0.2\%$ by weight of cement [71]). On the other side, due to the nature of the reactants used and the geometry of the body, exposure to preformed components can limit CO_2 uptake. These factors can result in restrictions in CO_2 transport to the point, where CO_2 uptake typically ranges between 5% and 50% by weight of cement [30,72].

Although several scientific studies have looked into developing new methods to pursue a higher carbonation efficiency in cementitious mixes, CO₂ concentration and pressure, temperature and relative humidity (RH), along with the chemical and physical properties of the body to be carbonated, strongly influence the diffusivity of CO₂, which is highly associated with carbonation efficiency. Studies have shown that increase of CO2 concentration from 1% to 20% nearly doubled the carbonation degree of dry-mix pressed cement pastes from 12.8% to 22.8% [73]. Rising gas pressure from 0.01 to 0.05 MPa also led to an increase in the carbonation degree of concrete blocks by 4.4% [74]. While ambient temperatures are generally adopted for carbonation curing, use of higher temperatures (e.g. 40–60 $^\circ\text{C})$ can significantly improve the carbonation degree of cement pastes and concrete by promoting CO_2 diffusion and leaching of reacting ions [26,75]. It was demonstrated by most studies that the carbonation efficiency of PC-based mixes is favored at a RH of 50-65% [76].

Shi et al. [77] investigated the effects of CO₂ pressure, curing time, water-to-cement (w/c) ratio and continuous curing after CO2 curing on the carbonation degree and strength of PC-based concrete. Results revealed that the reaction between PC and CO₂ occurred mainly in the first 15 min, accompanied by a rapid exothermic process and a temperature rise, regardless of curing conditions. Among these factors, pre-conditioning conditions, including the w/c ratio of the sample before CO₂ curing, have the greatest influence on the amount of CO₂ sequestered and strength development, leading to an optimum w/c ratio range of 0.36-0.43 [77]. After 4 h of CO₂ curing (100% CO₂ concentration and 60 psi pressure), the maximum CO₂ sequestration degree was recorded as 17%, along with a compressive strength of 7.5-12 MPa. To reduce plastic shrinkage cracking due to rapid water loss, Shi et al. [78] further proposed to control the rate of water loss during pre-conditioning, with an optimum moisture loss of ~4.5%. Compared to conventional steam curing of concrete (18-24 h), it was concluded that CO₂ curing only required 4–8 h to achieve the same mechanical strength, thereby potentially reducing energy consumption during the process, while trapping CO_2 in a stable form [78].

To further improve carbonation efficiency, Kashef-Haghighi et al. [79] proposed a flow-through reactor, and achieved a CO_2 sequestration of 8–10% in 1–2 h at atmospheric pressure, with a final CO_2 sequestration degree of 16–20%. This presented a promising development in carbonation technology as lower amounts of energy is required in a flow reactor than in a static CO_2 pressure chamber. Junior et al. [80] reported that after CO_2 curing cement pastes for 1 h, followed by subsequent

hydration for 28 days, the compressive strength could be improved from 32.2 to 35.5 MPa. This increase in strength was associated with the role CO_2 curing played in increasing the contents of silica and hydrated alumina and reducing the chemically bound water, thus contributing to the subsequent hydration reaction [81].

The use of various inert or fibrous materials that are not directly involved in hydration or carbonation reactions in PC-mixes can also enhance the reaction with CO₂ and improve the properties of the matrix after CO₂ curing, while reducing the use of PC. The carbonation reaction of PC mixed with inert materials is generally not significantly different from that of pure PC as inert minerals mainly act as structural modifying components and fillers at specific interfaces to enhance performance. Limestone can be used as a supplementary cementitious material (SCM) to accelerate the hydration process in PC. Tu et al. [82] found that using limestone powder to replace 5-20% of cement could significantly increase the carbonation degree of the cement pastes from 17.5% to 30.2%. Limestone powder with a maximum particle size of 20 µm could serve as a nucleus for the precipitation of reaction products (e.g. calcium carbonates) and induce the consumption of calcium silicates (e.g. C₃S and C₂S) [83]. Incorporating limestone powder in PC mixes can also generate more carbonation products and reduce the porosity of the mix [83]. Tonoli et al. [84] investigated the carbonation curing of fiber-reinforced cement composites and found that the precipitation of calcium carbonate reduced the diffusion of silica, sulfur and aluminum ions during the early stages of hydration, leading to pore densification and improved fiber-matrix interface, therefore reducing porosity and dry shrinkage. Pizzol et al. [85] also reported that the precipitation of carbonated products improved the interfacial transition zone between the cellulose fibers and the cement matrix, contributing to the long-term properties of these mixes.

4.2. Aggregates

Aggregates typically make up 60-80% of the volumetric composition of concrete. Natural resources such as sand, gravel, and crushed stone make up the vast majority of aggregates utilized today. Depletion of natural resources has led to an increased focus on the use of recycled aggregates and artificial aggregates. Today in Europe, about 10% of the aggregate mass fraction is composed of recycled or artificial materials [86]. In recent years, secondary and manufactured aggregates have experienced an increased demand due to restrictions on mining activities and the depletion of high-quality aggregate sources close to urban areas. The production of aggregates by mineral carbonation is considered a promising production route, provided that the synthesized product is economical and meets the relevant quality and performance criteria. The evolution of carbonate bonded aggregates for engineering applications is also facilitated by limited landfill capacities and high landfill usage fees [87,88], leading to a rising demand in the use of alternative sources.

4.2.1. Recycled concrete aggregates

Recycled concrete aggregates (RCAs) are produced from construction and demolition wastes with an annual production exceeding 3 billion tons [89]. In recent years, the use of accelerated carbonation to enhance the properties of RCAs has revealed an effective method to improve the quality of RCAs and permanently sequester CO_2 as for every tonne of cement attached to RCAs, the absorption of about 0.5 tonnes of CO_2 could be theoretically expected [90]. Furthermore, the water absorption of RCAs with particle sizes of 5–10 mm and 10–20 mm could be reduced by 30% and 22% after accelerated carbonation, along with a 4.8% and 3.2% increase in apparent density, respectively [7]. The compressive and flexural strengths of concrete samples do not decrease significantly until the replacement rate of carbonated RCAs for natural aggregates (NAs) reaches over 40% [91]. However, the addition of carbonated RCAs may increase the risk of reinforcement corrosion and carbonation of reinforced concrete due to the reduction of the alkali

content [92,93].

Accelerated carbonation of RCAs consists of the following key steps shown in Fig. 3. First, CO₂ penetrates through the pores or cracks into the loosely attached mortar around the RCAs and dissolves in the pore water, generating carbonic acid and calcium ion, induced by the decomposition of phases such as Ca(OH)₂, C-S-H, C₃S, C₂S and ettringite (AFt). Further reaction of calcium with carbonate ions leads to the formation of CaCO₃ and silica gel [63,94]. CaCO₃ precipitates in the pores and cracks of RCAs in the form of calcite, vaterite and aragonite [95]. As the carbonation reaction continues, the surface of RCAs is gradually carbonated, while the interior is generally unchanged because the initial precipitation of CaCO₃ on the surface hinders the further penetration of CO₂. The precipitation of CaCO₃ results in the reduction of porosity and filling of cracks, associated with the higher hardness and solid volume of CaCO₃ than those of Ca(OH)₂ and C-S-H, thereby improving the quality of RCAs [96].

Accelerated carbonation techniques have been developed for RCAs to achieve rapid carbonation within hours [98]. Generally, the optimal conditions for carbonation involve a CO₂ concentration of 40–60% [99, 100], humidity of 40–70% [74,99,101] and temperature of 20–30 °C [7, 98,102,103]. To further improve the carbonation efficiency of RCAs, pressurized carbonation technologies involving high CO₂ pressures are also employed [104,105], necessitating the determination of an optimal gas pressure range [100]. Another method involves the use of flow-through CO₂ curing, during which a mixture of CO₂ gas and air is injected on one side of the chamber and discharged on the other to improve the carbonation efficiency of RCAs [79,99,106]. As a part of this process, saturated magnesium nitrate (Mg(NO₃)₂) solution can be used to control the indoor RH at 50 \pm 5%, with a recommended flow rate and CO₂ concentration of 5 L/min and 10%, respectively [99,107]. Compared with conventional carbonation, flow-through CO₂ curing presents a higher carbonation efficiency as it shortens the treatment duration.

An alternative carbonation technique involves the liquid-solid (wet) carbonation treatment of RCAs, which exhibits a better carbonation efficiency than gas-solid (dry) carbonation. In a water-CO₂ environment, CO₂ has a higher solubility and therefore the reacts more completely with Ca²⁺ than CO₂ gas in a dry environment [108,109]. Therefore, this method is preferrable for the carbonation-based curing of RCAs and recycled concrete powders (RCPs) derived from construction and demolition waste [110–113]. To enable their wet carbonation, RCAs are first placed in water in a chamber, followed by the injection of CO₂ gas into the water. An additional improvement was proposed via the use of CO₂ nanobubbles mixed with water, which contributed to the improvement of the carbonation efficiency [109]. The incorporation of

these treatments in the production of carbonated RCA (CRCA) results in the improvement of several key properties (i.e. density, mechanical and durability properties) [97]. These improvements, summarized in Table 3, are associated with the pore filling effect of CaCO₃ and silica gel generated by carbonation, which also reduces the severity of the cracks present in the old mortar around the RCA.

4.2.2. Artificial aggregates

Artificial aggregates (AAs) are usually prepared by pelletizing techniques that combine fine moisturizing particles into larger ones. The fresh manufactured pellets can be turned into a solid composite aggregate by accelerated carbonation. The strength development of pellets can be accelerated at high CO₂ concentrations [8]. For example, the strength of basic oxygen furnace slag (BOFS) aggregates increased to 3.2 times that of normally cured aggregates when the fresh pellets were subjected to 4 days of carbonation curing under 20% CO₂, 65% RH and 20°C [128]. Carbonated AAs can be made entirely from low-value wastes such waste cement, steel slag, and incineration ashes like wood ash and

Table 3

Improvements in the properties of CRCA.

	Property	Change (compared to RCA)	Ref.
Properties of CRCA	Apparent	+ 2.9 - 7.8%	[101,114,
	density		115]
	Water absorption	- 22.6–28.3%	[98,114,116]
	Crushing value	- 25.9–44%	[98,100,117]
Mechanical properties	Compressive	+ 13.1 - 32.9%	[7, 114, 115,
of concrete with	strength		118-120]
CRCA	Elastic modulus	+ 5.8 - 27%	[117,
			119-121]
	Tensile strength	+ 9.2 - 37.1%	[104,122,
			123]
	Flexural	+ 16.2 - 50.8%	[98,124]
	strength		
Durability properties of	Gas	- 43.5–43.6%	[7,117]
concrete with CRCA	permeability		
	Water	- 20.7%	[7,117]
	absorption		
	Carbonation	- 19.7%	[125,126]
	depth		
	Chloride	- 33–64%	[115,117,
	permeability		127]
	Drying	- 8–25.2%	[104,114,
	shrinkage		117]
	Autogenous	+ 126%	[114]
	shrinkage		



Fig. 3. Reaction processes involved in the accelerated carbonation treatment of RCAs [97].

paper ash [128–130]. Fig. 4 depicts a typical schematic diagram of the carbonated AAs production process, in which the binder is premixed with the waste powder to assure fresh granule quality. To thoroughly carbonate the fresh pellets, CO_2 is used in both the pelletizing and post-curing processes in this approach.

In most cases, the carbonation process contributes to the formation of $CaCO_3$ in the outer layer, resulting in a relatively dense carbonated shell layer. The relatively loose microstructure of carbonated AAs is the main reason for their high water absorption. Table 4 summarizes the physico-mechanical properties of carbonated AAs.

5. Commercialization of carbonation technology: products, process and applications

In line with the rising need for the development of sustainable solutions, the cement industry has been promoting the production of building materials with various CO₂ utilization technologies. Fig. 5 shows the flow chart of commercial CO₂ utilization technologies for construction materials and typical industrial units. Generally undertaken by start-up companies and pre-commercial entities, most of the initiatives in this area fall into four main categories: (1) Production of concrete via CO2 curing of cement-based materials (e.g. CarbonCure, Solidia Technologies and CO₂-SUOCOM), (2) manufacturing of blocks from industrial wastes such as steel slag (e.g. CarbiCrete and Carbstone Innovation); (3) preparation of synthetic aggregates (e.g. O.C.O technology and Blue Planet); and (4) preparation of concrete with carbonated bonded mineral (e.g. Calera Corporation). In most of these approaches, existing industrial equipment is usually sufficient, without necessitating major alterations. However, as a part of the CarbonCure technology, the installation of an additional CO₂ injection system is needed (Fig. 5(a)). Alternatively, for the carbonation curing of concrete and blocks, only a CO₂ curing tank is required (Fig. 5(b)), which can easily be retrofitted from an existing steam curing system. For aggregates, pelletizers are already common in existing aggregate production facilities (Fig. 5(c)). This section summarizes the different methods of utilizing CO2 in cement-based materials that are currently being developed or utilized in the industry. For each technology, the production, characteristics and potential applications of carbonated products are presented, a summary of which is shown in Table 5.

5.1. Carbonate bonded concrete/blocks based on cement-based materials

There are three main commercial application technologies in carbonate bonded concretes: (1) CO₂ cured PC, an example of which is CarbonCure that involves the injection of CO₂ into PC concrete mix; (2) CO₂ cured PC with mineral additives, examples of which are CO₂-SUI-COM that involves the reduction of the cement content by γ -CS₂ and fly ash, and CarbonBuilt where the cement is replaced by portlandite and fly ash; and (3) CO₂ cured non-hydraulic cement, an example of which is Solidia Technologies, whose mixes are composed primarily of wollastonite (CS) and rankinite (C₃S₂).

Table 4

Physico-mechanical	properties	of	carbonated	AAs.
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Raw materials	Bulk density (kg/m ³)	Water absorption (%)	Crushing strength (MPa)
Air pollution control residues [131]	950	19.6	-
Cement kiln dust [132]	1200	25.4	1.15
Basic oxygen furnace slag [128]	1280	12.5	5.23
Paper ash and quarry fines [129]	850	27.8	0.17
Concrete waste powder, PC, slag and Ca(OH) ₂ [133]	926	6.3	2.98
Quarry fines, cement kiln dust, slag and FA [134]	1000	19	-

5.1.1. CO₂ bonded concrete based on Portland cement

CarbonCure Technologies (Canada) developed a CO₂ curing concrete technology, in which liquid CO₂ transported from pressurized tanks is injected into fresh concrete during mixing [139]. The amount of CO₂ injected is limited to up to 1.5% by weight of cement (i.e. about 5 kg/m³ of concrete). Established gas suppliers collect, purify and distribute the CO₂ sourced from industrial emitters. Once injected into the wet concrete mixture, the Ca²⁺ from cement reacts with the CO₂ to form nanoscale CaCO₃, which becomes embedded in the concrete, thereby increasing the strength of concrete.

The CO₂ absorption efficiency of concrete is estimated to be around 50–80%, with the rest escaping into the atmosphere [139,140]. The cement content in the concrete mixture can also be reduced by 5% due to the strengthening effect of CO₂ [141]. Taking into account net emissions reductions from raw material transportation and the replacement of carbon-intensive plasticizing additives with CO₂, CarbonCure's process ultimately reduces net CO₂ emissions from concrete manufacturing by 18 kg/m³ when compared to that of PC concrete [142]. The process flow diagram for the utilization of CO₂ in the production of ready-mixed concrete by CarbonCure's technology is shown in Fig. 6. The cost is reduced due to reduced curing time, especially when compared to the use of non-chloride accelerators [139]. Nevertheless, the use of liquid CO₂ in the curing process offsets the cost savings to some extent.

CarbonCure's technology has been adopted by several concrete manufacturers, with 2.5 million truckloads (about 13.92 million tons) of concrete and 180,000 tonnes of CO₂ saved (relative to the equivalent amount of PC concrete produced) [135]. A 12-story building in Atlanta (Fig. 7(a)) was completed in 2018 by using CarbonCure concrete (367, 000 m³), enabling the sequestration of 680 tonnes of CO₂. Another example is the distribution center shown in Fig. 7(b), which was completed in 2021. This structure involves the consumption of 6308 m³ of CarbonCure concrete, resulting in the sequestration of 63,503 kg of CO₂ [135].

Another company, Carboclave (Canada), offers a retrofit that strengthens prefabricated cement products by using CO_2 instead of steam or mist, which makes use of existing autoclave curing equipment [143]. The technology is suitable for a variety of dry-cast and precast



Fig. 4. Schematic diagram of carbonated AAs' production process [8,129].



Fig. 5. Flow chart of commercial CO₂ utilization technologies for construction materials and typical industrial units, showing: (a) CO₂ injection system [135], (b) carbonation curing tank [136], and (c) pellet machine [8].

Table 5

Different initiatives inv	volving the use of	CO ₂ in the prod	uction of constru	ction products.
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Product/Area	Company/ Technology	Approach	CO ₂ source and curing parameters	Final product
Carbonate bonded concrete/blocks based on cement-based materials	CarbonCure	Injection of CO ₂ into fresh concrete mix	Pure CO ₂ , atmospheric pressure and room temperature	Concrete
	CO2-SUICOM	Reduce cement by γ -C ₂ S and fly ash	Flue gas, CO ₂ gas 15–20%, 40°C	Concrete
	Solidia	Low lime, low kiln burning temperature cement and CO ₂ cured concrete	Flue gas, ambient gas pressures and moderate temperatures (10% CO ₂ , 30°C [137])	Cement and concrete
Carbonate bonded blocks involving	Carbicrete	Carbonation of steel slag	Pure CO ₂ , CO ₂ partial pressure of 1.5 bar	Carbonated blocks
industrial wastes	Orbix (Carbstone innovation)	Carbonation of steel slag	Flue gas, autoclave under high pressure and temperature	Carbonated blocks, roofing tiles
Carbonate bonded aggregates	0.C.0	Manufactured limestone aggregate from industrial waste	Flue gas (100% CO ₂ , 2 bar [138])	Aggregates and fill for blocks, concrete and screed
	Blue planet	Carbonate coating over an alkaline substrate	Dissolved CO_2 with a sufficient pH	Aggregates
Concrete with carbonated bonded minerals	Calera	Carbonate mineralization by aqueous precipitation	Circulating flue gas throughout brine	Carbonate precipitates

concrete products. The CO_2 used in this process can be used as an accelerating and strengthening curing agent to make concrete harden quickly and increase strength. The reaction is highly exothermic and greatly accelerates the hydration of cement [143]. In turn, the CO_2 gas is permanently trapped as tightly bound calcium carbonate crystals in the cement slurry matrix.

5.1.2. CO_2 bonded concrete/blocks based on Portland cement with mineral additives

CO₂-Storage Under Infrastructure by Concrete Materials (CO₂-SUI-COM, Japan) developed a CO₂ curing concrete technology, in which flue CO₂ is fed into the concrete curing chamber [144,145]. CO₂ comes directly from waste flue gases produced by industrial sources such as steel plants, power plants and cement plants. In addition, supplementary cementitious materials with low CO₂ emissions (e.g. coal ash and blast furnace slag) are used in large quantities, making CO₂-SUICOM carbon-negative. The γ -C₂S involved in CO₂-SUICOM can also absorb CO₂ and contribute to the formation of CaCO₃. This not only reduces the amount of cement significantly, but also accelerates the absorption of CO₂ within the concrete, as outlined in Fig. 8. A typical mixture for CO₂-SUICOM consists of 161 kg/m³ of water, 101 kg/m³ of cement, 82 kg/m³ of GBFS, 53 kg/m³ of FA, 32 kg/m³ of γ -C₂S, and 844 kg/m³ and 965 kg/m³ fine and coarse aggregates, respectively [145]. The prepared concrete is cured for approximately 2 weeks under a CO₂ concentration of 15–20% at 40 °C [144].

The strength of CO₂-SUICOM is comparable to that of PC-based concrete products, along with excellent abrasion resistance and reduced efflorescence. The main products are currently precast concrete components such as factory-made road blocks and paving blocks used in both public and private buildings. As a part of their carbon recovery technologies, Mitsubishi Corporation supports technology demonstration, commercialization and marketing of the technology to achieve a decarbonized society. Kajima Corporation is in charge of the development and design of concrete products, Chugoku Electric Power Company (thermal power plant) is responsible for production demonstration, Denka Corporation is involved in the development of special admixture, and Landes Corporation is handling the manufacture and sale of precast products. In February 2011, CO₂-SUICOM was used in the boundary blocks, foundation blocks, and paving blocks in the information facility at the Fukuyama Photovoltaic Power Station (Fig. 9). Field trials showed



Fig. 6. Process flow diagram for the utilization of CO₂ in the production of ready-mixed concrete proposed by CarbonCure [142].



(a) 725 Ponce

(b) Cedar Creek Distribution Center

Fig. 7. Engineering applications using CarbonCure's concrete technology [135].

that the performance of CO₂-SUICOM met the requirements for this application. In January 2012, CO₂-SUICOM was used in the terrace ceiling in Nakano Central Park Residence (Fig. 10), where it was estimated to enable the absorption of 100–200 kg/m³ of CO₂ during the hardening process.

Another CO_2 sequestration approach involves CarbonBuilt's ReversTM process, proposed by the University of California, Los Angeles (UCLA), enabling the incorporation of industrial CO_2 in both concrete mix design and the curing process. The introduction of portlandite (calcium hydroxide) leads to a reduction in the amount of traditional cement used and increases the use of waste products such as fly ash [146]. While concrete is prepared via the same process and equipment used today, waste CO_2 is applied to this process without the need for expensive capture, compression or purification. In 2020, with support from the U.S. Department of Energy and NRG COSIA Carbon XPRIZE, the technology was applied in the production of over 10,000 concrete blocks using CO_2 from a flue gas taken directly from a coal power plant at the Wyoming Integrated Test Center. In 2021, a second demonstration was successfully completed at the National Carbon Capture Center in Alabama, during which CO_2 in flue gases from both coal and natural gas power plants was used to produce more than 5000 blocks [146].

5.1.3. CO₂ bonded concrete based on non-hydraulic cement

Solidia Technologies (USA) has developed two products with lower net CO_2 emissions relative to traditional construction materials: Solidia CementTM and Solidia ConcreteTM (Fig. 11). Solidia CementTM is a nonhydraulic cement composed mainly of wollastonite (CS) and rankinite (C₃S₂) [147]. Due to the lower amount of CaO present in this binder, the calcination of CaCO₃ is greatly reduced, along with a lower production temperature (1200°C) than PC production (1450°C). Solidia concrete is manufactured by curing Solidia cement under CO₂, as shown in Eq. (6). The concrete ready-mix containing Solid cement is poured and vibrated for consolidation. After demoulding, the concrete is placed into the carbonation incubator. The CO₂ sourced from waste flue gas streams is captured and injected into the incubator, which is sealed until the curing process is complete [148,149]. Although the process requires a CO₂-rich



Fig. 8. CO₂-SUICOM concept used in the preparation of concrete [145].



Fig. 9. Application of CO₂-SUICOM to building elements in Fukuyama Photovoltaic Power Station [144].



Fig. 10. Application of CO₂-SUICOM to building elements in Nakano Central Park Residence [144].

environment, it can be performed under an ambient pressure and moderate temperature (~60 °C). A report by Ashraf et al. [137] declared that CS could be effectively carbonated at 30 °C and 10% CO₂ concentration in 145 h. Most manufacturers of precast concrete are capable of achieving these parameters.

According to the developer's claims [151], Solidia technology can reduce the carbon footprint by up to 70% and water consumption by 60–80%, while easing production and reducing costs by improving the performance of cement and concrete. The CO₂ emissions of Solidia cement could be reduced from ~810 kg/ton in PC clinker to ~565 kg/ton due to the low-lime content [149]. During the carbonation-based curing process, each tonne of Solidia cement can further absorb 220–236 kg of CO₂ [152]. The concrete reaches full strength within 24 h, compared to up to 28 days for conventional concrete products [148]. This curing process can be easily tailored to a range of concrete formulations, production processes, and standards while utilizing the batching/mixing equipment already in use by the producer. Lafarge Holcim and Air Liquide are currently working with Solidia Technologies for commercializing the developed technology in the form of concrete blocks and paving stones for now, as shown in Fig. 12. These initiatives are being followed by the development of a ready-mix product suitable for in-situ concrete applications.

5.2. Carbonate bonded blocks involving industrial wastes

Accelerated carbonation is investigated as an alternative sustainable



Fig. 11. Comparison of Solidia cement with PC [150].



(a) Solidia concrete paving stones

(b) Solidia concrete blocks

Fig. 12. Product applications involving the use of Solidia concrete [153].

curing technique to steam curing concrete blocks [154], not only to sequester CO_2 and reduce energy consumption, but also to limit the associated CO_2 production [136]. At present, commercial applications of this technology are mainly based on the recycling of steel slags in

high-quality construction blocks, as proposed by CarbiCrete and Carb-Stone [155–158]. This process combines slag recycling with the beneficial use of CO_2 at increased pressure and temperature and may provide a solution for the recycling of steel slag, which is currently difficult to



Fig. 13. Diagram showing the accelerated carbonation steel slag blocks [160].

reuse. The adopted technique usually involves three main steps, as shown in Fig. 13 [159]: (1) Pre-treatment of the steel slag including mixing and grinding; (2) compaction of steel slag into blocks; and (3) CO_2 curing with elevated CO_2 pressure (autoclave) or low CO_2 pressure (climate chamber).

CarbiCrete developed an accelerated carbonation process, as a part of which CO₂ is injected into wet steel slag mixtures [161]. The BOF/EAF slag mixture (obtained from a Canadian steel plant, originally destined for landfill), water and aggregates are cast into construction blocks using steel molds and then dried in front of a fan. The typical mixture consists of 1555 kg/m³ steel slag mixture, 930 kg/m³ of slag aggregates and 311 kg/m³ of water. The blocks are then carbonated for 24 h at 1.5 bar CO₂ partial pressure and stored in plastic bags for 35 days to allow for further hydration [162]. The procedure can be used in any concrete-producing plant with minimal disruption to the process flow [158]. Fig. 14 shows concrete blocks produced using CarbiCrete technology. It is estimated that the production of a standard size concrete block (referred to as an 18 kg cinder block) using this process would save 2 kg of CO₂ emissions and absorb 1 kg of CO₂ during the curing process.

CarbStone also uses CO2 cured steel slag-based concrete blocks (Fig. 15). The coarse slag is ground into fine filler particles using a grinding mill. The filler and slag sand are mixed together with water, during which the particle size of the raw materials and the amount of water are precisely controlled for optimum carbonation. The moist material is then pressed hydraulically into the desired shape, such as a large brick, which can be hollow or solid. Carbonation is performed in an autoclave under high pressure and temperature [157]. With this technology, the Carbstone blocks are able to sequester up to 150 g of CO₂ per kg of slag [160]. Moving on from a lab scale, this technology is further developed and validated by the optimization of process parameters, such as pressure, temperature, and the pre-treatment of the raw material feed in subsequently (semi-)industrial equipment and process conditions. Since 2013, a pilot plant along the Sambre River in Farciennes-Walloon Municipality has used a carbonation technology (patented by CarbStone Innovation NV) to treat slag from a variety of sources [157]. The objective of the plant is to produce high-value construction products from steel slag by carbonation [157].

5.3. Carbonate bonded aggregates

There are two main commercial applications of artificial carbonated aggregates: (1) carbonate bonded manufactured aggregates from industrial wastes such as blast furnace slag and steel slag, for which a representative company is O.C.O Technology (UK); and (2) carbonate bonded artificial pebbles from waste concrete or mineral aggregates, for which a representative company is Blue Planet (USA).

5.3.1. Manufactured LimeStone aggregate from industrial wastes

O.C.O Technology (UK), also called Carbon8 Systems in the past, developed an accelerated carbonation technology in a commercial



Fig. 14. Concrete blocks produced using CarbiCrete technology [158].

operation to produce Manufactured LimeStone (M-LS) aggregates. A wide range of thermal wastes such as cement dusts, steel slags, oil-shale ash, incinerator ash, paper ash and contaminated soils are collected and treated by captured CO₂ [163,164]. The flowchart of the M-LS production process, which employs multiple stages to convert wastes into aggregates, is shown in Fig. 16. The waste is mixed with precisely controlled quantities of liquid CO2 and water in a pre-treatment mixer for carbonation. Fillers and binders are then added to carbonated waste and sent to a batch mixer together. After mixing, the mixture is conveyed to the pelletizer and CO₂ gas is injected to accelerate the cementation process and form round aggregates. The process is completed with the final screening and storage of aggregates [162,165]. A variety of flue gases are captured and directly utilized in the O.C.O accelerated carbonation technology. Furthermore, rainwater is also collected and utilized, with no solid, liquid or gaseous waste being emitted. The overall treatment process is exothermic, requiring no additional heat input. The main electricity consumption is associated with the transportation and mixing of the materials, resulting in a potentially carbon-negative process.

At three UK-based commercial facilities located in Brandon (Suffolk), Avonmouth (Bristol) and Leeds, carbonated thermal wastes are blended with binders and fillers, and then pelletized to form rounded M-LS aggregates. Over 100,000 tonnes of waste are processed each year for the production of over 200,000 tonnes of sustainable aggregates [166]. M-LS aggregates manufactured by treating Air Pollution Control residues (APCR) from the Energy from Waste (EfW) sector with waste CO_2 gas were used for the first time in an asphalt mix, applied in a 2.5 km cycle path (Fig. 17). This approach was claimed to save up to 90% CO_2 when compared to the use of conventional hot AC20 asphalt mixes, resulting in the reduction of the overall CO_2 emissions by up to 70 tonnes throughout the construction process.

5.3.2. Artificial pebbles from waste concrete

Blue Planet (USA) developed carbonated rocks as aggregates, for which flue gas is converted into a solid carbonate coating on small pebbles made of demolition debris, as shown in Fig. 18. CO2 is first dissolved in water containing divalent cations (e.g. Ca^{2+} and Mg^{2+}) to form carbonates. In order for the dissolved CO2 to exist as a carbonate or bicarbonate, an excess of alkali is added to maintain the pH at 9.5 [167]. According to Blue Planet's patent [167], divalent cations can come from many different sources, depending on the availability at a particular location. These sources include industrial wastes, seawater, brine, hard water, minerals and any other suitable source. The resulting carbonate precipitates are filtered, washed, and dewatered. A binder (e.g. PC) can then be added to the carbonate minerals to bind the powdered material and attach it to the recycled aggregates [167]. Ultimately, aggregates of various shapes, surface textures, hardness, chemical resistance, density, porosity and reactivity can be produced. 1 m³ of Blue Planet aggregates typically contains 44% CO₂ [168].

In 2016, Blue Planet's limestone-coated light weight aggregates were used in the boarding area of San Francisco International Airport, as shown in Fig. 19. Concrete tests showed that Blue Planet's concrete met all necessary specifications, for which 1 m^3 of concrete contained 1.14 tons of CO₂ [168].

5.4. Concrete with carbonate bonded minerals

Calera Corporation (USA) developed a unique technology for carbon sequestration through the carbonate mineralization by aqueous precipitation method (CMAP) [169]. The basic principle of the CMAP method is to capture flue CO₂ gas from gas or coal-fired power plants, turn it into carbonates and finally react with calcium or magnesium. The CO₂ is dissolved in an aqueous solution with a high pH value and reacted with Ca²⁺ and Mg²⁺ to precipitate CaCO₃ and MgCO₃. The stable CaCO₃ and MgCO₃ can be used as supplementary cementitious materials or aggregates in concrete. Previous studies reported the production of



Fig. 15. Concrete blocks produced using CarbStone technology [157].



Fig. 16. Flowchart of the M-LS production process [164].



Fig. 17. Product applications involving the use of O.C.O technology [166].

Calera products by adding a NaOH solution to a brine of seawater and CaCl₂, then circulating CO₂ throughout the brine to produce CaCO₃ and MgCO₃ precipitate [169]. The precipitated CaCO₃ and MgCO₃ was spray-dried after being rinsed in a slurry. Then 10 g of Calera product was mixed with 40 g of PC with a water-to-solid ratio of 0.8. In capturing CO₂ from raw flue gases, Calera's carbonate mineralization technology utilizes alkaline solid and liquid sources, such as fly ash, wastewaters, and brines [170].

Calera has constructed a demonstration plant at Moss Landing (California), capable of capturing 30,000 tons of CO_2 per year. According to the company, this plant could be expanded to capture 300,000 tons of CO_2 and convert it into more than 550,000 tons of valuable building materials per year [170]. The flue gas originates from Dynegy's Moss Landing natural gas power plant, while the alkalinity for the CO_2 capture process comes from onsite $Mg(OH)_2$ or Calera's

electrochemistry process [170]. Another related initiative, the Calera Yallourn project in Victoria's Latrobe Valley (Australia), is expected to collect more than 300,000 tons of CO_2 and create more than one million tons of building materials every year and two million gallons of fresh water every day. Alkalinity for the CO_2 capture process could come from brines and fly ash [170].

6. Future challenges and solutions

The commercial use of CO_2 in building materials is still in its early stages. While the industrial trials reported above show that CO_2 can be used in construction components, the scalability and market feasibility are still influenced by a number of factors. These include the potential cost of mineral carbonation technologies, the carbonation process, the technical challenges faced by carbonation methods and the various



Fig. 18. Process flow involving the use of CO₂ to manufacture Blue Planet aggregates, showing: (a) general flow chart and (b) process of converting flue gas into solid carbonate coating on small pebbles made of demolition debris [168].



Fig. 19. Placement of Blue Planet's concrete at the San Francisco International Airport [168].

economic and social factors linked to market penetration.

6.1. Challenges

6.1.1. Availability of raw materials and cost of carbonated products

PC is currently produced at a global level of 4.1 billion tonnes, while reactants suitable for carbonation, including portlandite and wollastonite, are currently produced globally at around 350 million tonnes and 800,000 tonnes, respectively [171]. Therefore, depending on volume ratios and raw material availability, it is possible to produce portlandite and wollastonite at the same cost as PC and with a much lower CO₂

intensity due to the lower synthesis temperatures required during their production [172] and lower CaO content. In addition, the costs of CO_2 capture, operation and transportation need to be considered. To sequester 1 ton of CO_2 within cement-based materials is estimated to cost 25–56 USD [173]. Current carbonation technologies are constrained by the location of the material and waste gas streams, making it challenging for them to be implemented efficiently in large and centralized operations. The production of carbonated aggregates is unlikely to be cost efficient when compared to the quarrying industry in the near future. Therefore, without suitable incentives, carbonated binders could be more market viable than carbonated aggregates.

6.1.2. Source, availability and suitability of CO₂

Most studies on carbonation utilization involve the use of pure CO₂ gas. However, high purification and pressurization is required in the treatment of CO2, which could limit its applications. However, the construction materials industry presents opportunities and markets for the use of waste CO₂, which can be obtained from different sources (e.g. flue gases from coal-fired power plants or waste incineration plants), albeit their low concentrations ($\leq 25\%$ CO₂ v/v) [174,175]. Large point sources of CO2 include large fossil fuel or biomass energy facilities, major CO2-emitting industries, natural gas production, synthetic fuel plants and fossil fuel-based hydrogen production plants. Table 6 summarizes the CO₂ emissions of various industrial activities on a global level, whereas Table 7 lists the CO2 content, presence of major and minor impurities and temperature in flue gas streams. Therefore, when using CO₂ to cure building materials on a commercial scale, low-cost CO₂ capture systems or the ability to handle large volumes of gas streams containing components other than CO2 is required. A more important issue is that these waste CO₂ sources usually originate from energy intensive enterprises such as power plants, which are far from the main markets [171]. This may require the co-location of production facilities for carbonated materials with CO₂ emission sites, which may also provide waste heat that facilitates the advancement of chemical reactions. Alternatively, the transport of CO2 to production and consumption centers would need to be considered, which will result in additional costs and safety precautions.

6.1.3. Carbonation reaction and processing

Carbonation usually proceeds readily under certain conditions, which is often owing to favorable thermodynamics rather than design. Table 8 summarizes the key barriers in carbonation reaction with different precursors. However, carbonation is usually hindered by three technical factors: (1) competition between hydration and carbonation (i. e. highly hydratable Ca-rich solids typically show poor potential for carbonation); (2) carbonate phases covering the precursor particle surface, thereby preventing further contact between the precursor and CO₂; and (3) the role of water, whose adsorption on the surface can favor carbonation [184], whereas condensation in the pores of microporous or mesoporous solids can inhibit carbonation [185,186]. Accordingly, even when CO₂ is solubilized into the pore liquid, the trapping of water prevents the transport of vapor-phase CO2 into the microstructure [187]. Therefore, it is necessary to develop additives or seeds to promote carbonation by influencing the rate of reaction control, either kinetically or thermodynamically. This could involve increasing growth rates (e.g. by epitaxial matching [188]), promoting surface dehydration to alleviate ion attachment or other interactions with adsorbed water [189], and surface complexation to facilitate the subsequent adsorption and non-homogeneous growth [28].

6.1.4. Construction codes and standards

Carbonation cured building products may be comparable or even superior when compared to PC-based concrete in terms of performance, but they still have not been assigned the standardized performance indicators in terms of durability and structural engineering performance. The lack of relevant building standards and codes makes it difficult to

Table 6

CO ₂ emissions of	different industrial	activities	[176]
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Industry/activity	CO ₂ emissions (Mt/year)
Power	10,539
Cement production	932
Refineries	798
Iron and steel industry	646
Petrochemical industry	379
Oil and gas processing	50
Bioethanol and bioenergy	91
Other sources	33

Table 7

CO ₂ content,	, impurities and	temperature in	flue gas streams
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Flue gas sources	CO ₂ (%)	Major impurities	Minor impurities	Temperature (°C)
Gas fired [177] Coal-fired [178, 179]	7.4–7.7 12.5–15	H ₂ O, O ₂ , N ₂ H ₂ O, N ₂	CO, NOx CO, NOx, SO ₂	50–75
Power plant [180]	15-20	N ₂ , O ₂ , H ₂ O	NOx	
Steelmaking plant [181]	20	CO, N ₂	H ₂	50–75
Cement kiln plant [182,183]	14–33	H_2O, O_2, N_2	CO, NO, SO ₂	
Atmosphere	0.04	N ₂ , O ₂		Ambient

Table 8

Precursor	Carbonation reaction
Fresh PC-based concrete	Limited CO ₂ dosage in the mixture. At ambient conditions, the reaction rates are slow. Besides, the presence of moisture prevents carbonation
Mature PC-based concrete	Carbonation takes years to occur under diffusion- controlled ambient conditions
Non-hydraulic calcium silicates	To increase the reaction rate, high concentrations and pressures of CO_2 are required
Industrial wastes (e.g. fly ash, steel slags, etc.)	The reaction kinetics are strongly influenced by the heterogeneity of the wastes, and their CO ₂ uptake is significantly lower than that estimated from their bulk oxide composition
Mg-based cementitious materials	A high water content is required for magnesium hydration, however this limits CO ₂ diffusion

commercialize these products on a large scale. Another dilemma is that building standards tend to be prescriptive rather than performancebased, specifying the composition of the materials that can be used instead of the performance requirements. Furthermore, some wastes, as well as waste flue gases, are often restricted or subject to CO_2 taxes and fines, which is a substantial challenge that needs to be overcome by specifying appropriate regulations for the widespread use of carbonated materials within the construction industry. This necessitates a change to a unified system of performance-based standards, as well as new tools that can anticipate or simulate the long-term performance of these unique components.

6.2. Suggestions

The construction materials market offers significant potential for CO_2 utilization in the short to medium term. Although the process technology for carbonation-based curing of concrete is further developed than many chemical and biological utilization routes, further research is required to address key barriers in carbonation technology, processing, and market acceptance. From an academic perspective, the following aspects should be considered further:

- (1) Carbonation process is inherently complex, particularly with regards to the growth and arrangement of the carbonation products. Furthermore, the co-occurrence of hydration and carbonation reactions creates an additional challenge in understanding reaction mechanisms. Thus, the recognition of the chemical reactions that dominate the relative rates of carbonation and hydration, and the growth of different crystalline forms of reaction products can facilitate better selection of alkaline solids and reaction conditions to achieve controllable carbonation reactions.
- (2) Development of additives to enhance the solubility of CO_2 or structure-directing seeds to accelerate the growth of carbonate crystals for reducing the time cost in carbonation.

- (3) Optimization of the process design for industrial applications aimed at balancing ecology and efficiency.
- (4) Systematic evaluation of the durability of CO₂-cured concrete, especially its resistance to reinforcement corrosion, to increase its market acceptance.
- (5) Mastering the engineering structure performance to develop related standards for the large-scale application of CO₂-cured building components.
- (6) Development of new material formulations with novel properties and advancement of the use of additive manufacturing technologies to build components with higher strength-to-weight ratios, optimized topologies, and more complex geometries than existing manufacturing methods.
- (7) Modeling the carbonation process involved with gaseous CO₂ diffusion, dissolution, composition alternation, and carbonation reaction in the inner matrix of cement-based materials to enable their long-term monitoring.

Generally, products with productibility and large-scale applicability could attain sufficient market potential. Moreover, costs could be reduced if concentrated sources of flue gas and waste heat are close to mineral sources, such as mines or industries that generate highly alkaline wastes. Therefore, commercial initiatives and other financial investments should be directed towards:

- Enhanced competitiveness for producers, increased profitability and sales differentiation.
- (2) Rapid industrial upgrading with very low barriers.
- (3) Integrating and complementing existing low carbon solutions, regulations and supply chains.
- (4) Continuous innovation in low-carbon and recycling technologies.
- (5) Win-win partnerships between the construction value chain, governments and industry.

The demand for sustainable concrete from governments and related public and commercial bodies is increasing, despite certain obstacles. However, environmental protection policies are not fully in place in most countries, particularly in developing countries. When the use of new cementitious components is scaled up rapidly and combined with active policies, emission reductions can be achieved in all infrastructure projects. Currently, most of these initiatives involving the sequestration of CO_2 within construction components have not reached commercial viability yet. A series of coordinated schemes leading to large-scale demonstration, research and development is required, further supported by incentives for the public and private sectors to invest in and consume these products.

7. Conclusions

This paper provided an in-depth review of the recent advances in academic and commercial initiatives involving the accelerated carbonation of various construction materials. Along with a discussion on the fundamental carbonation mechanisms and processes, and factors affecting carbonation kinetics and environmental and economic impact; the information presented in this paper shed light on the technical and industrial benefits of accelerated carbonation and its applicability to different construction applications. The up-to-date presentation on the research and industrial advances made in recent years was supported by outlining the challenges faced and potential solutions to enable the large-scale application of carbonation technologies. Relevant recommendations were also provided to address the current issues associated with the utilization of carbonation in the building sector, with the goal of paving the way for the direction of future research in this area and providing guidance on the application of CO₂ sequestration in the production of construction materials with the desired sustainability and cost. The following conclusions were drawn from this review:

- (1) Carbonation can be accomplished by using various reactants including PC, non-hydraulic calcium silicate, industrial solid wastes, and magnesium-based cementitious materials. While academic and industrial R&D initiatives have focused on identifying methods to improve the CO₂ utilization of these binders, further effort is needed to reach the maximum theoretical values for carbonation within a certain duration. Therefore, there is still room for improvement in the conversion rates and reaction kinetics presented by existing carbonation technologies.
- (2) Many factors influence the carbonation process, including the characteristics of the precursor material, CO_2 concentration and pressure, curing temperature, water to binder ratio and precuring conditions. Therefore, future research efforts need to focus on optimizing reaction mechanisms and associated microstructural changes for the development of effective methods to enhance the carbonation reaction according to the characteristics of the precursor material.
- (3) Carbonation that leads to the production of carbonates such as $CaCO_3$ and $MgCO_3$ from Ca- and Mg-based precursors is a fundamental method that can be applied for the production of mineral aggregates and binders for concrete mixes. When compared to other industries, the utilization of CO_2 in the production of construction materials is considered as one of the most promising initiatives for CO_2 sequestration.
- (4) Recently developed commercial products involve the reuse of captured CO₂ in the curing, manufacture and modification of construction materials. Key utilization techniques include carbonate bonded blocks containing cement-based materials and industrial wastes, carbonate bonded aggregates, and concrete with carbonate bonded minerals. While these applications hold some promise in terms of CO₂ sequestration, they are still considered at demonstration or small-scale production level as they face various technical and market challenges.
- (5) Availability of raw materials and cost of carbonated products; source, availability and suitability of CO₂; carbonation reaction and processing; and construction codes and standards are the main factors limiting the use of CO₂ on a large scale in the development of construction materials. Combining industrial solid waste recycling with carbonation technology can help integrate solid and gaseous waste utilization processes in high emission industries such as steel, power and cement production. However, the structural stability and durability performance of CO₂-cured building components in actual use requires further long-term testing. On the basis of the development of new material technologies, the development of specific processes and devices for carbonation technology should be actively promoted.
- (6) Current gaps in literature and associated technology regarding the utilization of CO_2 in construction materials that need further investigation include the enhancement of chemical reactions, identification of suitable additives and seeds to promote hydration and carbonation, optimization of the process design, systematic evaluation of durability and mechanical performance, development of new formulations with novel properties, advancement of the use of additive manufacturing technologies and modeling of the carbonation process.

Declaration of Competing Interest

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

Data availability

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

Acknowledgment

The authors acknowledge the financial support from The Royal Society (project ref: $ICA\R1\201310$).

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