

Review article

Carbon dioxide sequestration through steel slag carbonation: Review of mechanisms, process parameters, and cleaner upcycling pathways



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ABSTRACT

The direct carbonation of steel slag has emerged as a promising approach for carbon dioxide (CO₂) utilization and sequestration, holding potential for advancing sustainable steel production. Despite considerably high expectations for these cleaner upcycling pathways, their maturity level remains relatively low and large-scale direct carbonation of steel slag is largely untested. To facilitate steel slag carbonation on a scale necessary for a zero-carbon future economy, this article provides a comprehensive review of fundamental carbonation mechanisms and critical parameters governing the reaction process, including temperature, pressure, reaction time, liquid-to-solid ratio, and CO₂ partial pressure. The study critically examines the unique interactions among these process parameters, which can either limit or enhance the process optimization. The spectrum of scientific challenges associated with this pathway, including reaction rate limitations and the carbonated product valorization, particularly as a binder or aggregate in the construction sector, are identified and addressed. These insights aim to enhance the carbonation potential of steel slag for possible cleaner upcycling implementation pathways, ultimately facilitating the development of more efficient and sustainable carbon capture utilization and sequestration (CCUS) technologies. The proposed improvements are expected to be instrumental in promoting sustainable practices, not only to foster the decarbonization of the steelmaking industry but also in aiding other hard-to-abate sectors, such as the cement and concrete industry, in achieving their own decarbonization goals.

1. Introduction

Steel stands as the most widely utilized and important metallic material, both in terms of production volume and its vast array of versatile applications. Despite its unrivaled significance, the iron and steelmaking process heavily relies on fossil fuels to meet energy demands and facilitate the primary extraction and refinement of ores [1,2]. Globally, the iron and steel industry represents one of the largest contributors to greenhouse gas (GHG) emissions, annually releasing 3.7 gigatons (Gt) of CO₂ [3]. This level of emissions averages around 2 tonnes of CO₂ for every tonne of steel produced, constituting approximately 7% of global CO₂ emissions [4]. Global steel production has increased drastically in recent decades, a trend that is projected to continue, given the strong correlation between steel market demand, population growth, and economic expansion [5]. Therefore, to address the growing issue of GHG

emissions, steelmakers currently face the challenge of reducing their CO₂ emissions by developing new technologies for sustainable production.

The two well-established methods predominantly used in steelmaking are the blast furnace-basic oxygen furnace (BF-BOF) and the electric arc furnace (EAF), with the former currently accounting for the majority (70.7%) of the global steel production [6]. However, the decarbonization potential of EAF steelmaking has garnered interest in recent years, aligning with the growing focus on green growth within the steel industry [7]. In both the BOF and EAF steelmaking processes, a by-product is generated known as *steel slag*, composed of various metallic oxides resulting from the removal of impurities from the molten steel. Typically, these processes yield 0.12 – 0.20 tonnes of steel slag for every tonne of hot metal produced. In contrast to the widespread reutilization of BF slag, the reutilization of steel slag is not as ubiquitous

Abbreviations: BF-BOF, blast furnace-basic oxygen furnace; BOF, basic oxygen furnace; CCUS, carbon capture, utilization, and sequestration; CRW, cold-rolling wastewater; C-S-H, calcium silicate hydrate; DRI, direct reduced iron; EAF, electric arc furnace; f-CaO, free calcium oxide; f-MgO, free magnesium oxide; GGBFS, ground granulated blast furnace slag; L/S, liquid-to-solid ratio; LF, ladle furnace.

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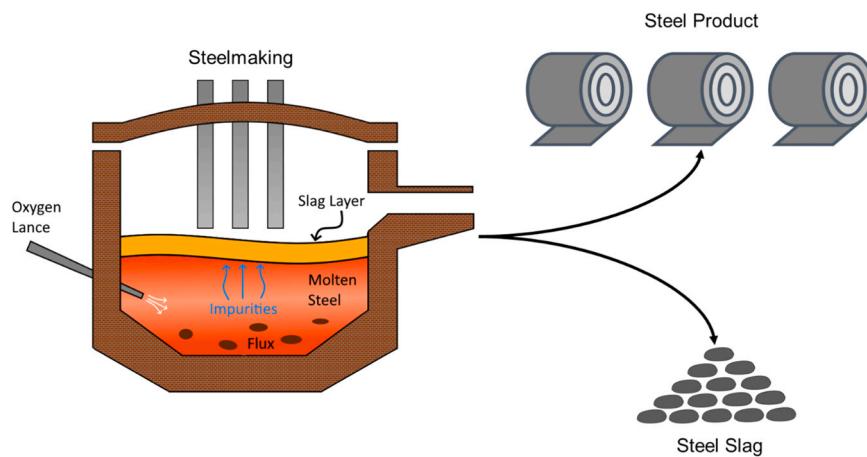


Fig. 1. Schematic showing the creation of the slag layer in a simplified EAF process.

[8]. Several obstacles contribute to this disparity, including difficulties in achieving desired granulometry due to high iron oxide content, heavy metal leaching potentials, and volume instability stemming from the elevated content of free lime ($f\text{-CaO}$) and free magnesia ($f\text{-MgO}$) [9,10]. While several studies have extensively investigated the utilization of steel slag in construction, agriculture, marine engineering, ceramics or water treatment applications [11–15,8], carbon sequestration is one area that has gained considerable attention in the past decade.

Several ongoing efforts aimed at decarbonizing the iron and steel industry are focused on increased usage of EAF from scrap and direct reduced iron (DRI) melting, as well as improvements to the BF-BOF route with hydrogen or biofuel substitutes. An effective strategy for CO₂ reduction that can significantly benefit both approaches is sequestering captured CO₂ into steel slag. The alkaline nature of these slags makes them excellent candidates for CO₂ sequestration by slag carbonation, contributing to the valorization of the industrial waste. The fundamental concept underpinning slag carbonation mirrors the geological weathering process [16], which naturally regulates Earth's climate by coordinating the reaction of Ca and Mg silicates with atmospheric CO₂ to form carbonate minerals [17]. This process is considered as the most stable CO₂ trapping mechanism that offers a secure and permanent storage for CO₂ [18]. However, natural weathering occurs at extremely slow rates for all feedstocks, including steel slag [19,20]. To address this challenge, accelerated carbonation and its associated process parameters have been the subject of extensive investigation in various *in-situ* and *ex-situ* processes [21–23].

Several recent reviews have explored various aspects of CO₂ sequestration using steel slag as the storage medium. Humbert and Castro-Gomes [24] reviewed the current status of carbonated

construction materials. Their review largely focused on the mechanical performance, production process, and environmental benefits of carbonated steel slag. Li et al. [25] provided a life cycle analysis and studied the environmental benefits of steel slag reuse, detailing environmental benefits and drawbacks of carbonation. Their study, however, does not explicitly give a thorough investigation of the carbonation process itself. Teo et al. [15] assessed the recycling options for EAF steel slag, identifying multiple uses in construction materials but did not consider CO₂ sequestration. Similarly, Naidu et al. [26] reviewed potential uses of BOF slag and determined various applications, but provided limited insight into the CO₂ sequestration and BOF slag carbonation process. While several reviews [27–31,8] have explored the carbonation mechanisms of cement materials (due to their similar alkaline nature), the examination of these mechanisms in the context of steel slag has remained mostly unexplored. Therefore, advancing steel slag as a CO₂ sequestration material requires a detailed explanation of the reaction process, as undertaken in the current review.

Given the attractive CO₂ sequestration potential of steel slag in decarbonizing the steelmaking industry amid a dearth of relevant research, the current study aims to provide a comprehensive review of the available research pertaining to the cleaner upcycling of steel slag. The primary focus is on the carbonation process and the associated influential parameters, as well as the potential utilization of carbonated steel slag as a value-added material in the construction sector. This work, for the first time, intricately describes the role of each carbonation parameter on the reaction kinetics while systematically evaluating the impact of carbonation process parameters on CO₂ uptake. The insights thus provided from our analysis, spanning from fundamental principles of the carbonation process to practical applications of carbonated steel slag, will serve as a guide for designing steel slag carbonation systems and determining specific areas necessitating further research attention.

Table 1
Typical EAF and BOF oxide compositions and mineralogical phases, according to [32,35,41].

Oxides	EAF Slag	BOF Slag
CaO	20% – 40%	30% – 50%
SiO ₂	10% – 20%	10% – 15%
Al ₂ O ₃	2% – 10%	1% – 3%
MgO	5% – 15%	5% – 15%
FeO _x	20% – 40%	10% – 30%
MnO	2.5% – 5%	1% – 5%
TiO ₂	0.5% – 1%	0.5% – 2%
P ₂ O ₅	0.5% – 2%	1% – 3%
Typical mineral phases	<i>Major Phases:</i> $\beta\text{-Ca}_2\text{SiO}_4$, $\gamma\text{-Ca}_2\text{SiO}_4$, Ca_3SiO_5 , $f\text{-CaO}$, $f\text{-MgO}$, FeO, and the RO phase (a solid solution of CaO-FeO-MnO-MgO) <i>Minor Phases:</i> Merwinite ($3\text{CaO}\text{-MgO}\text{-}2\text{SiO}_2$), Olivine ($2\text{MgO}\text{-}2\text{FeO}\text{-}\text{SiO}_2$), Gehlenite ($\text{Ca}_2\text{Al}[\text{AlSiO}_7]$), $2\text{CaO}\text{-}\text{Fe}_2\text{O}_3$	

2. Distinctive steel slag characteristics

Steel slag broadly encompasses slag by-products from various steelmaking processes, including EAF, BOF, and ladle furnace (LF) slag. However, given the significantly lower production volume of LF slag, this review will primarily focus on EAF and BOF slags, which are generated in a similar manner [13,32]. In the steelmaking process, oxygen is injected into the melt for carbon combustion heating, but also to oxidize metallic impurities in the molten steel. Lime or dolomite fluxes are added to control slag chemistry and maintain slag basicity, which can combine with the resulting metallic oxides to form the slag. The slag layer has several key functions, including absorbing oxidation products from the steel. Furthermore, specifically in EAF steelmaking, the slag serves as a protective barrier, shielding the furnace lining from the direct impact of the arc. This not only prolongs the lifespan of the refractory

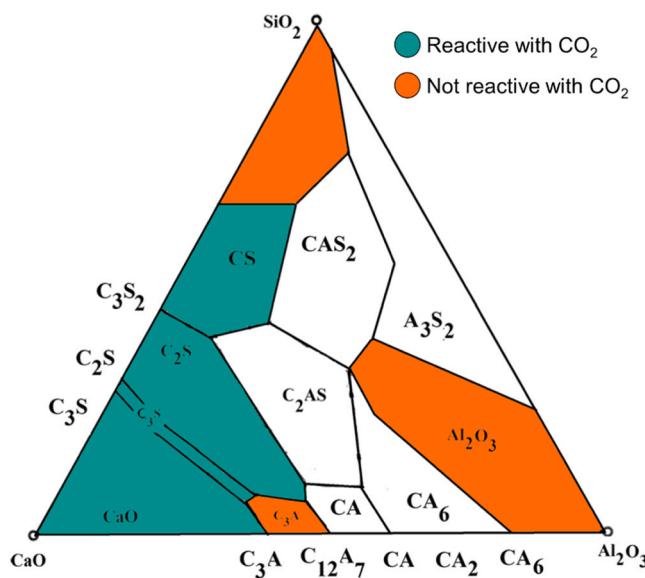


Fig. 2. The ternary phase diagram for the $\text{CaO}-\text{SiO}_2-\text{Al}_2\text{O}_3$ with CO_2 indicated (adapted from [46]).

lining but also contributes to the overall thermal efficiency of the process. The role of slag in the steelmaking process and its generation as a by-product are illustrated in Fig. 1. In both EAF and BOF steelmaking processes, the steel slag, comprising of oxide residues, is tapped off separately.

Although EAF and BOF slag share some similarities in their formation and main oxide constituents [32], the specific proportions of their chemical compositions can vary based on factors such as the steelmaking process, quantity of scrap steel used, the grade of steel being produced, and the condition of the furnace refractory lining [32–34]. In BOF slag, the main constituents are CaO , FeO , SiO_2 , and MgO , while EAF slag typically contains CaO , FeO , SiO_2 , MgO and Al_2O_3 [32,35]. Typical ranges of oxide composition and other minor oxide constituents are listed in Table 1. When cooled, steel slag emerges as a mixture of various mineral phases, among which the calcium silicate phases stand out as the most prevalent and crucial for carbonation [36]. Numerous mineral phases reported in steel slag, as listed in Table 1, include tricalcium silicate (Ca_3SiO_5 or C_3S), β -dicalcium silicate ($\beta\text{-Ca}_2\text{SiO}_4$ or $\beta\text{-C}_2\text{S}$), and γ -dicalcium silicate ($\gamma\text{-Ca}_2\text{SiO}_4$ or $\gamma\text{-C}_2\text{S}$). Due to the typically slow cooling of steel slag and a high basicity ratio (CaO/SiO_2), tricalcium and β -dicalcium silicate are not easily formed [36]. Other reported phases include Mg- and Fe-containing silicates, $f\text{-CaO}$, $f\text{-MgO}$, FeO compounds,

and a solid solution of $\text{CaO}\text{-FeO}\text{-MnO}\text{-MgO}$ (referred to as the RO phase), and less commonly wollastonite (CaSiO_3 or CS) [37–40].

The reported $f\text{-CaO}$ and calcium silicate phases (and to some extent Mg-silicate phases) make steel slag a potential candidate for CO_2 sequestration through the formation of carbonates (e.g., CaCO_3) [42, 43]. The ternary phase diagram depicted in Fig. 2 represents the $\text{CaO}\text{-SiO}_2\text{-Al}_2\text{O}_3$ system, and highlights phases that are reactive with CO_2 in green, not reactive phases in orange, and unstudied phases in white. Compositions or phases closer to the CaO corner are particularly notable, as they signify phases well-suited for carbonation and are typically observed in steel slag (as shown in Table 1). This potential has been substantiated through extensive carbonation research on calcium silicate phases in cement [44,45].

3. The carbonation process

3.1. Carbonation process methods

Carbonation can occur in two distinct forms: directly or indirectly. The latter is a two-step reaction process that involves the use of an extracting agent to leach certain metal elements from the steel slag, followed by the dissolution of injected CO_2 gas into the aqueous solution to precipitate carbonates. In contrast, direct carbonation is a one-step process where CO_2 directly reacts with the steel slag, forming carbonates. Since direct carbonation does not require an extraction agent, it is a less expensive process than indirect carbonation. Hence, this review will focus on direct carbonation, which can be further divided into the following.

3.1.1. Gas-solid carbonation

Gas-solid carbonation involves two phases (gas and solid) where the gas, typically CO_2 , reacts with the solid material (oxides or minerals), resulting in the sequestration of CO_2 as a stable carbonate. In this case, reaction kinetics is well understood [47] and can be enhanced to achieve accelerated carbonation by raising the reaction temperature, pressure or the CO_2 concentration. The reaction proceeds by a rapid surface reaction-controlled step, followed by a slower second stage product (carbonate) layer diffusion-controlled process. Since the process is eventually diffusion controlled, parameters such as temperature, pressure, and CO_2 volume percentage accelerate the carbonation process [48]. The experimental procedure is typically carried out in an environmentally controlled chamber, as shown in Fig. 3(a). The carbonation reactions of Ca oxides are rapid and therefore occur over a time scale of hours. Mg oxides and hydroxides can be carbonated as well, but their reactions do not proceed as rapidly as that of their Ca counterparts [19]. The Ca/Mg oxide and hydroxide carbonation reactions are presented in

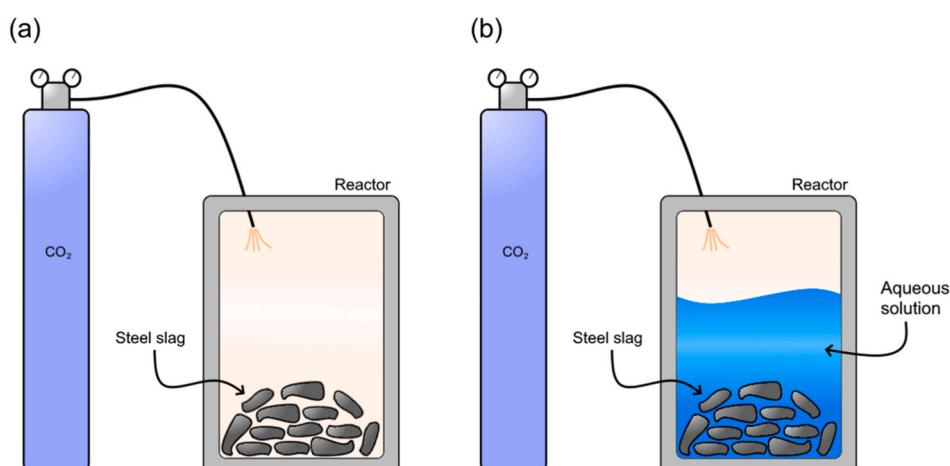


Fig. 3. Schematic representation of an experimental for (a) direct gas-solid carbonation setup (b) aqueous carbonation setup.

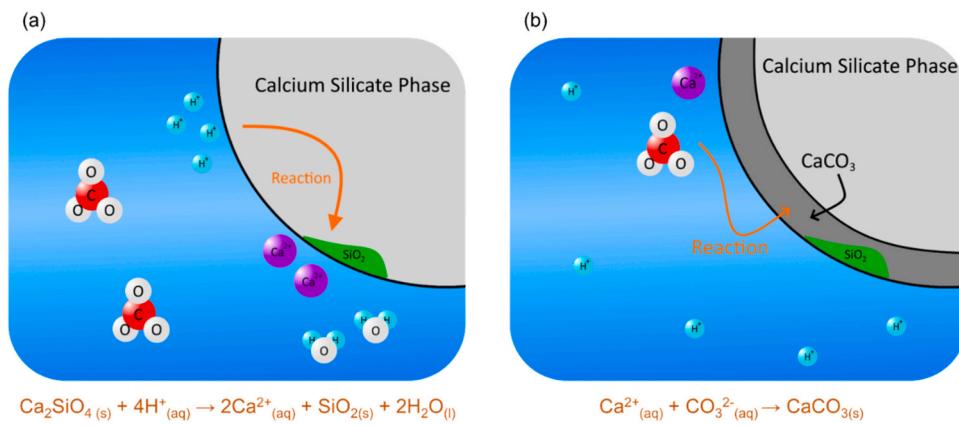
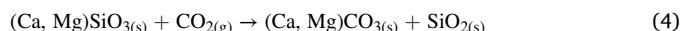
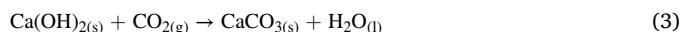


Fig. 4. Illustration of the (a) Ca leaching reaction (b) CaCO_3 formation reaction.

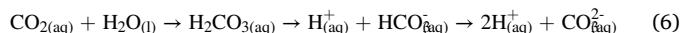
Eqs. 1–3, with carbonates as the resultant products. Zhang et al. [49] found that gas-solid carbonation is also applicable to the calcium silicate and magnesium silicate phases present in steel slag. The general reaction is presented in Eq. 4. Huijgen et al. [50,51] also discovered that calcium silicate phases in steel slags were efficient materials for CO_2 sequestration, but required a high CO_2 partial pressure to maintain an appreciable reaction rate.



3.1.2. Gas-liquid-solid carbonation

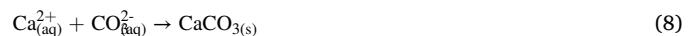
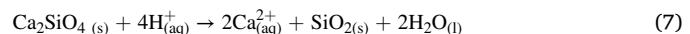
Gas-liquid-solid carbonation involves the reaction of CO_2 with a solid material in an aqueous or water-based medium. It is more complex than gas-solid carbonation since it involves three phases (gas-liquid-solid). The experimental set-up is like that of gas-solid carbonation, but the steel slag is mixed with water, which introduces an additional process parameter of liquid-to-solid ratio (L/S). A schematic of the experimental set-up is shown in Fig. 3(b). The aqueous solution allows for the dissolution of CO_2 and the leaching of Ca ions from the steel slag, which increases the reaction rate [52]. Additionally, it does not require elevated temperatures and pressure, as is the case with gas-solid carbonation, making it a more energy-efficient process, yet still maintaining a carbonation timeframe on the scale of hours [53]. This method has received widespread attention in the literature [54,55].

The case of aqueous carbonation presents a more complex reaction sequence, as demonstrated below through non-hydraulic phase – dicalcium silicate ($\gamma\text{-Ca}_2\text{SiO}_4$) – reactions. The first step is the dissolution of CO_2 gas into the aqueous solution (Eq. 5). The CO_2 then reacts with water to form carbonic acid (H_2CO_3), which is a slow exothermic step. Almost instantaneously, the H_2CO_3 ionizes into H^+ and bicarbonate (HCO_3^-). In high pH environments, the final product is carbonate (CO_3^{2-}) [56]. This sequence of reactions is shown in Eq. 6.

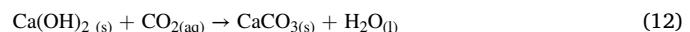
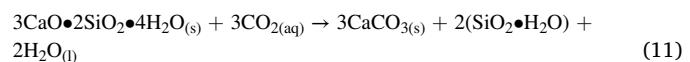


The H^+ ions leach the Ca^{2+} ions from the $\gamma\text{-Ca}_2\text{SiO}_4$ matrix into the aqueous solution. Additional reaction products are silica gel (SiO_2) and H_2O (Eq. 7). The free Ca^{2+} ion reacts with the CO_3^{2-} ion to form CaCO_3 , thus storing CO_2 (Eq. 8). A schematic illustration of the chemical reaction is shown in Fig. 4 and the overall carbonation reaction is given in Eq. 9. As shown in Fig. 4, the CaCO_3 (and SiO_2) forms as a product layer

on the calcium silicate phase [57–61]. This is a critical feature of the carbonation reaction, as it can act as an inhibitor to further carbonation [47,62,63].



In the case of hydraulic calcium silicate phases (Ca_3SiO_5), they undergo a hydration reaction upon contact with water to form calcium silicate hydrate ($\text{CaO} \bullet \text{SiO}_2 \bullet \text{H}_2\text{O}$ or C-S-H) and calcium hydroxide [Ca(OH)_2 or CH]. The carbonation and hydration reactions do not exactly occur simultaneously and depend on the CO_2 concentration in the aqueous solution [64]. The overall carbonation reaction for Ca_3SiO_5 , as an example, is given in Eq. 10. The hydrated products can be further carbonated, as shown by Eqs. 11 and 12.



Although Ca(OH)_2 has a higher initial reaction rate, its rate is eventually surpassed by that of calcium silicate hydrate [65,66]. Once Ca(OH)_2 is consumed, the calcium silicate hydrate will continue to carbonate [67,68]. Whether the calcium silicate is a hydraulic or non-hydraulic phase, the reactions have similar fundamental steps, relying on the dissolution of CO_2 into CO_3^{2-} and the leaching of Ca^{2+} from the calcium silicate phases. A layer of CaCO_3 forms on the calcium silicate particle (also called the ash layer), through which the CO_3^{2-} must diffuse to continue the reaction process. However, one distinction is the role of water. For the non-hydraulic phases, CaCO_3 is directly formed from a reaction with the CO_3^{2-} without the formation of hydration products. Therefore, during the carbonation of non-hydraulic calcium silicate phases, water serves as a catalyst and plays no direct role in the carbonation reaction sequence.

Generally, the RO phase in steel slag (a solid solution of CaO-FeO-MnO-MgO) is known to have poor hydration activity [69]. In fact, it has been determined that the RO phase with a high ratio of Fe/Mn can also be detrimental to the hydration activity of Ca_2SiO_4 [70]. However, the carbonation of the RO phase has been found to increase its hydration activity. Li et. al [71] found that carbonation promotes the RO phase hydration due to the increase in Mg-leaching induced by CO_2 dissolution. This underscores an additional advantage of carbonating steel slag, as it can enhance the utilization of the conventionally inert RO phase in applications requiring elevated hydration activity. However, the exact carbonation reaction and associated mechanisms still lack clarity and

Table 2
Thermodynamic parameters of select carbonation reactions [72].

Reaction	ΔH (kJ/mol)	ΔG (kJ/mol)
Ca(OH) ₂ (s) + CO _{2(g)} → CaCO _{3(s)} + H ₂ O _l	-68	-73.04
Mg(OH) ₂ (s) + CO _{2(g)} → MgCO _{3(s)} + H ₂ O _l	-37	-38.06
Ca ₂ SiO _{4(s)} + 2CO _{2(g)} → 2CaCO _{3(s)} + SiO _{2(s)}	-81	-64.02
Ca ₃ SiO _{5(s)} + 3CO _{2(g)} → 3CaCO _{3(s)} + SiO _{2(s)}	-148	-91.86

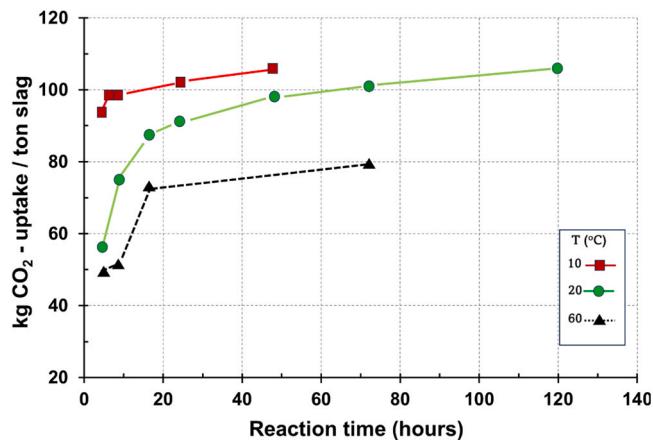


Fig. 5. Effect of reaction time on the CO₂ uptake of steel slag adapted from [77] and reproduced [in colors] for improved readability.

would be a useful topic for future research.

From a thermodynamic perspective, the carbonation reaction is exothermic and spontaneous [72]. Cheng and Wu [72] studied the thermodynamics of key carbonation reactions and their findings are summarized in Table 2. The negative enthalpy change (ΔH) indicates that all the studied carbonation reactions are exothermic, with the carbonation of Ca₃SiO_{5(s)} releasing the most heat. Similarly, the Gibbs free energy change (ΔG) for all cases is negative showing the carbonation reactions are spontaneous reactions. Given that the carbonation of Ca₃SiO_{5(s)} has the largest absolute value for ΔG, it is the most susceptible to carbonation. Similarly, Ca- is more carbonation prone than Mg- and thus more likely to be carbonated.

It is important to note that while carbonation reactions are thermodynamically viable at ambient conditions, the reactions kinetics must be taken into account. Potential rate-determining steps include CO₂ dissolution, CO₂ diffusion through the aqueous solution, CO₂ diffusion through the product layer, the dissolution of calcium or magnesium, and the precipitation of carbonates. These steps can be influenced by various factors, such as CO₂ pressure, water content, temperature, and the chemical composition of the slag, which can be optimized to enhance the overall rate of the reaction and increase the potential for CO₂ sequestration.

3.2. Process parameters

The carbonation process is influenced by several parameters, such as reaction time, reaction temperature, CO₂ pressure, CO₂ concentration, particle size, and slag composition. These parameters can significantly affect the kinetics and extent of carbonation, as well as the characteristics of the resulting carbonated materials. Therefore, it is crucial to carefully evaluate these parameters and determine their optimal utilization in enhancing the carbonation process, leading to the development of a stable and safe material suitable for various applications. This section will systematically examine the impact of various parameters on the carbonation process and overall CO₂ sequestration of steel slags.

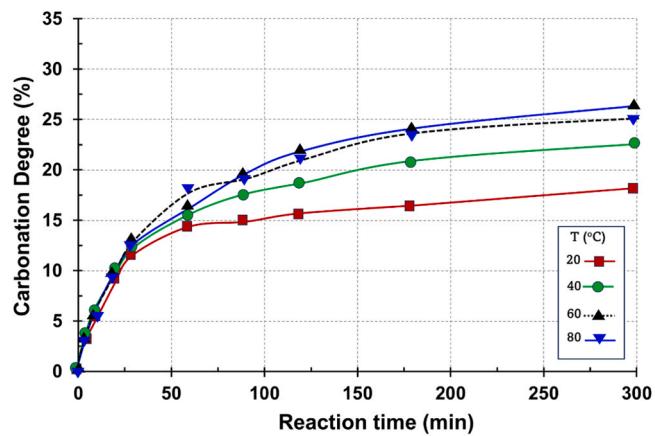


Fig. 6. A plot of CO₂ uptake at various temperatures showing that CO₂ uptake decreases after 60 °C, adapted from [83] and reproduced [in colors] for improved readability.

3.2.1. Reaction time

The reaction time provides the temporal window for carbonation to occur. It has widely been observed that the rate of steel slag carbonation is initially high but decreases as the reaction progresses, and eventually reaches a plateau [73,74] as shown in Fig. 5. There are several possible explanations for this phenomenon. The most widely cited explanation is that the product layer forms on the steel slag particle, gradually thickening as the reaction proceeds, making it increasingly challenging for the CO₂ to diffuse to the unreacted core [47,62,63,75]. The other theory is that the solution pH decreases as the carbonation reaction proceeds, due to the consumption of alkalinity and the release of H⁺ from CO₂ dissolution [76]. In either case, the reaction time should be optimized to maximize the extent of carbonation and minimize the amount of unreacted material.

3.2.2. Reaction temperature

The reaction temperature is a critical parameter that significantly influences steel slag carbonation. As previously highlighted, the carbonation reaction depends on two distinct mechanisms: the dissolution of CO₂ in the aqueous solution and the leaching of Ca ions. Raising the reaction temperature enhances the rate constants and thermodynamic equilibrium constants of the leaching reaction [77–79]. However, increasing the reaction temperature decreases the CO₂ solubility/dissolution process [80]. Interestingly, at higher pressures (> 100 bars), the solubility decrease with temperature reverses at elevated temperatures [81]. Hence, this could result in temperature having a positive and negative effect. Recent literature has shown that there is typically an optimal reaction temperature to balance these effects for maximum CO₂ sequestration. This has been observed experimentally by Chang et al. [82] and Tu et al. [83] (shown in Fig. 6), both of whom showed an optimal reaction temperature of 60 °C. However, the optimal reaction temperature may depend on other process parameters, such as pressure, and slag properties. Generally, reaction temperatures range from 20 – 100 °C for aqueous carbonation, although in some cases, the temperature was not observed to reach an optimal value [84,85]. In the case of gas-solid carbonation, elevated temperatures are required to accelerate the process. In fact, below 300 °C the carbonation was observed to be negligible [86]. However, there still exists an optimal temperature since, at very high temperatures (>600 °C), free CO₂ is more stable than carbonates [19,87]. Overall, reaction temperature is a critical but delicate parameter to optimize the carbonation reaction.

3.2.3. CO₂ pressure

Theoretically, increases in CO₂ pressure will result in increased CO₂ dissolution [81]. Henry's law states that the CO₂ concentration in

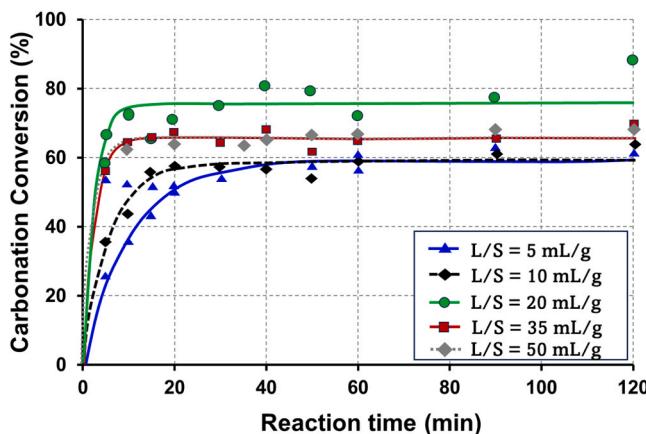


Fig. 7. The effect of L/S on carbonation showing high L/S can lead to a decrease in carbonation, adapted from [91] and reproduced [in colors] for improved readability.

solution is proportional to the CO₂ partial pressure above the solution. However, the literature gives conflicting reports on the effect of CO₂ pressure on the carbonation reaction. Rushendra et al. [53] initially found no effect from increases in CO₂ pressure, but further increases exceeding 6 bar showed a positive effect on CO₂ uptake, which eventually saturated. Omale et al. [63] observed similar behavior when increasing CO₂ pressure. While several studies reported a positive effect [88], others have observed an insignificant effect [89] or perhaps even a negative effect [90]. Increases in CO₂ pressure would only have a positive effect if the dissolution of CO₂ was the rate-determining step. In cases where it is not, such as when the leaching of Ca ions is limiting, the increase in CO₂ pressure would be insignificant, as observed in the previously mentioned studies. The negative effect of CO₂ pressure is more perplexing. One possible explanation is the decreased pH levels of the solution [90]. However more work is needed to understand these observations. A more detailed examination of the interaction between CO₂ pressure and other process parameters may explain the disagreements in the literature. Nonetheless, it can be understood that generally increased CO₂ pressure increases CO₂ uptake.

3.2.4. Liquid-to-solid ratio

Liquid-to-solid ratio (L/S) has been identified as a critical parameter in the aqueous carbonation of steel slag. It refers to the volume of liquid (typically deionized water) used per unit weight of the solid material (slag). The L/S determines the availability of water for the carbonation reaction and influences the reaction rate and extent of carbonation [91]. A higher L/S generally results in a higher reaction rate due to the increased availability of water for dissolution and the reaction of CO₂ with the slag [62,92,93]. Kurusta et al. [94] found that three times as much CO₂ can be sequestered in aqueous conditions versus dry. However, an excessively high liquid to solid ratio can lead to a lower concentration of dissolved CO₂ and lower concentration of leached Ca ions, which may result in a limited extent of carbonation [51,95]. Additionally, too much liquid can act as a mass transfer barrier, which will limit the reaction rate [96,97]. Obviously, insufficient L/S will result in a slower reaction rate and a limited extent of carbonation due to the limited availability of water to facilitate the reaction. Balancing the positive and negative effects creates an optimal value for L/S, similar to the discussion on reaction temperature in 3.2.2. Therefore, an optimal L/S should be chosen to maximize the extent of carbonation. This balance was observed by Rushendra et al. [53] and Cheng et al. [91]. The results from Cheng et al. are presented in Fig. 7. However, Kim et al. [98] showed that there is an interaction effect between L/S and temperature. The optimal ratio can depend on the other carbonation process parameters and should be determined through experimental studies. Generally, the optimal L/S has been found to be in the 10 – 20 range.

3.2.5. Particle size

Particle size has been determined to have the most significant effect on carbonation efficiency and CO₂ sequestration potential [16,62,83,84, 89,99–101]. In fact, Polettini et al. [99] found particle size to be more significant than slag chemistry. The effect of particle size on carbonation is mainly attributed to the increased surface area available for reaction with CO₂. Finer particle sizes generally result in a higher reaction rate and a higher degree of carbonation due to the increased surface area available for the reaction. This effect has been observed in numerous studies. For example, a kinetic study by Tu et al. [83] determined particle size had the largest effect on CO₂ uptake compared with reaction temperature, L/S, and CO₂ flow rate. A similar conclusion was reached by Su et al. [84] and Santos et al. [100]. In a study by Kim et al. [62] the design of experiments showed the strongest effect from particle size. Unlike the previously-mentioned factors, the reduction of particle size is universally agreed upon to increase CO₂ uptake. In terms of carbonation or CO₂ uptake, no negative effect from fine particle sizes has been reported. However, excessively fine particle sizes can result in practical difficulties such as handling and processing the slurry, as well as increased energy consumption during the carbonation process. The use of finer particles also increases the risk of clogging in the process equipment. Therefore, the optimal particle size distribution of slag should be determined to balance the advantages of increased reactivity with the disadvantages of handling difficulties and increased energy consumption.

3.2.6. CO₂ volume percentage

While the effect of CO₂ volume percentage on the carbonation of steel slag has not been extensively studied, there are a few studies that have provided pertinent results. Specifically, the interaction between CO₂ volume percentage and temperature in the case of gas-solid carbonation. Yu et al. [102] found that at high temperature (550 °C) Ca utilization and therefore carbonation was maximal at low CO₂ concentration (<10%) or high CO₂ concentration (>75%). The exact mechanism behind these observations is not explained in detail, but the authors postulate that at low concentrations other metallic compounds in the steel slag may have a catalytic effect. Similar results were observed by Prigobbe et al. [48] but no explanation was given.

Overall, while the effect of CO₂ volume percentage on the carbonation of steel slag may not have been extensively studied, the few studies that have been conducted provide interesting insights into its influence and the interactions with other parameters such as temperature, pressure, and particle size. Further research is needed to fully understand the role of CO₂ volume percentage in the carbonation of steel slag and its potential for carbon sequestration.

3.2.7. Slag composition

For carbonation to occur the steel slag must possess certain Ca or Mg phases. Therefore, slag composition is another factor that affects the carbonation reaction and CO₂ uptake. A few studies have examined the carbonation level of various calcium silicate phases. Zhang et al. [40] studied the carbonation conversion rates of four distinct mineral phases Ca₃Al₂O₆, CaSiO₃, Ca₂SiO₄, and f-CaO. The study found the conversion rates to be 40%, 42%, 51% and 100% respectively, showing the high carbonation of f-CaO and moderate carbonation of Ca₂SiO₄. Ashraf and Olek [103,104] studied the carbonation degree and activation energies of various calcium silicate phases. Hydraulic calcium silicate phases were found to have a much lower activation energy compared to non-hydraulic phases in the dissolution phase of the carbonation reaction. However, during the product layer diffusion-controlled portion of the reaction, the activation energies were found to be similar. Zhu et al. [105] found that γ-Ca₂SiO₄ generates more carbonates and had a higher carbonation activity compared to β-Ca₂SiO₄. However, the carbonation conditions were sufficient to cause equal activation energies. A similar conclusion was reached by Jang and Lee [106]. In terms of non-hydraulic calcium silicates, γ-Ca₂SiO₄ was found to have the highest

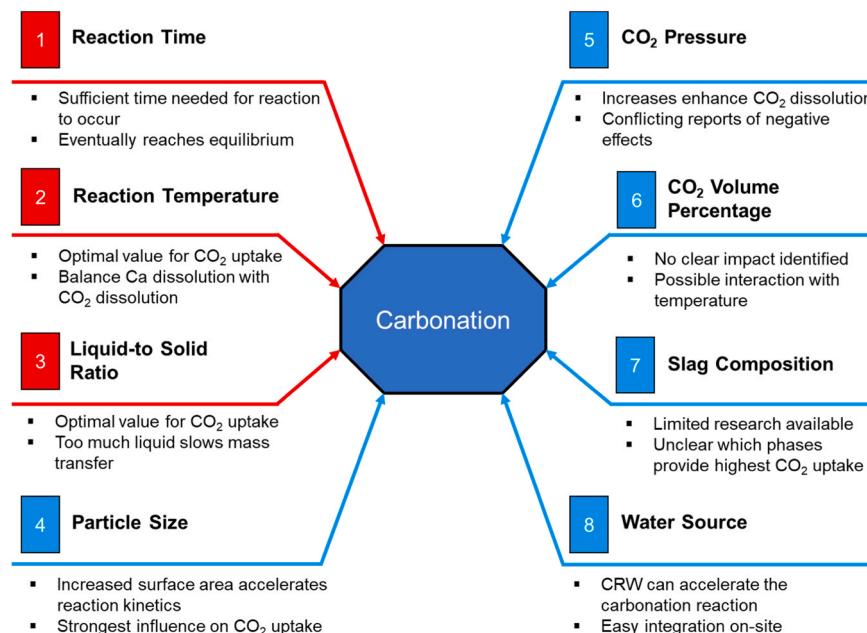


Fig. 8. Summary of the carbonation reaction parameters emphasizing their effect on the overall process with critical parameters in red.

CO₂ uptake capacity. Overall, both hydraulic and non-hydraulic calcium silicate phases can be well activated by CO₂ under the appropriate conditions.

In terms of overall steel slag chemical composition, a limited number of studies have suggested that higher concentrations of Ca and Mg in the slag may lead to increased carbonation [107,108]. Johnson [107] examined carbonation of materials with various Ca content (3.36% – 65.04%) and found a significant linear trend between percentage weight gain from carbonation and Ca weight percentage. These elements have the potential to enhance the reactivity of the slag with CO₂, thus promoting more efficient carbonation. However, the specific mechanisms and optimal concentrations of Ca and Mg for maximizing carbonation efficiency remain poorly understood. Studies on slag composition remain relatively scarce and detailed work is missing from the current literature due to the inherent challenges associated with controlling the composition of steel slag. Steel slag composition can vary widely depending on the steelmaking process, raw materials used, and operational conditions. This characteristic variability makes it difficult to conduct systematic studies investigating the direct relationship between slag chemical composition and carbonation efficiency. Consequently, research directly relating the concentration of Ca or Mg phases to CO₂ uptake is limited, highlighting the need for further investigations in this area.

Understanding how changes in the concentrations of essential phases, such as calcium silicates and magnesium silicates, affect carbonation will provide valuable insights into optimizing the composition of steel slag for enhanced carbonation performance. By delving deeper into this under-researched aspect, future studies can contribute to the development of more efficient carbonation processes and the utilization of steel slag as a valuable carbon capture and storage material.

3.2.8. Water source

The use of cold-rolling wastewater (CRW) as a liquid in the aqueous carbonation of steel slag has gained attention as a sustainable approach for both waste management and carbon capture. CRW, generated during the cold-rolling process in steel mills, exhibits a higher pH compared to other water sources, which has been found to facilitate CO₂ dissolution in the liquid phase [91,109]. This observation aligns with previous studies highlighting the positive correlation between pH and CO₂ dissolution [110]. Additionally, O'Connor et al. [111] demonstrated that

the presence of NaCl in the aqueous solution enhances the carbonation reaction; however, the exact mechanism behind this enhancement remains unclear. Interestingly, CRW is characterized by its high Na and Cl content, as reported by Cheng et al. [91], suggesting that these components may positively influence the carbonation of steel slag. Further supporting these findings, Cheng et al. [112] developed a kinetic model that analytically demonstrated the accelerated carbonation effect when CRW was used as the liquid medium in the carbonation process. Collectively, these findings suggest that utilizing CRW in the aqueous carbonation of steel slag holds promise due to its higher pH, Na and Cl content, and potential for enhanced CO₂ dissolution and carbonation efficiency, as demonstrated by both experimental and kinetic modeling studies. Furthermore, it offers a practical advantage as both CRW and steel slag are generated within the same steel mill facility, reducing transportation costs and environmental impact associated with sourcing external liquids for the carbonation process. Given the numerous advantages, the use of CRW should be further researched and trialed at industrial scale.

3.2.9. Summary of process parameters

The carbonation of steel slag involves several key process parameters that influence the efficiency and effectiveness of the carbonation reaction. These parameters include reaction time, reaction temperature, L/S, CO₂ pressure, particle size, and the composition of the slag. Optimizing these parameters is essential for achieving maximum CO₂ uptake and mineral conversion. Reaction time is often listed as a critical parameter. However, it does not accelerate the reaction kinetics. Therefore, it is only necessary to ensure there is sufficient time to reach the CO₂ uptake plateau. Conversely, temperature affects the reaction kinetics and has been shown to have a negative effect on CO₂ uptake at lower and higher temperatures. L/S was observed to have a similar effect with lower and higher values showing negative effects. The findings of this chapter identify temperature and L/S as critical parameters due to the delicate balance that needs to be struck for optimal conditions. Other parameters such as reduced particle size and CO₂ pressure are generally accepted to enhance the carbonation reactions and should therefore be maximized. However, it is important to note the economic and environmental restrictions on reduced particle size and CO₂ pressure. A similar approach applies to CO₂ volume percentage, but the impact of this factor has not been described in detail. The slag composition, including the presence of

reactive phases such as calcium and magnesium silicates, is crucial for facilitating carbonation. However, slag composition is difficult to control at an industrial-scale and the other parameters should be optimized to suit the available slag. In Fig. 8 the factors and their impact are summarized and categorized based on how critical they are to the carbonation process (critical parameters in red). Understanding and optimizing these process parameters are crucial for maximizing the carbonation efficiency of steel slag and enabling its utilization as a sustainable solution for CO₂ capture and storage.

4. Relation between process parameters and reaction kinetics

Each process parameter discussed in Section 3.2 has a specific effect on the carbonation reaction kinetics. To optimize the carbonation process, it is essential to understand the kinetic effect from each process parameter and how they interact with each other. The rate limiting steps, such as CO₂ dissolution, Ca leaching, or diffusion through the product layer, can then be described and addressed by the appropriate process parameters.

4.1. CO₂ dissolution

The first step in the carbonation reaction sequence is the dissolution of CO₂ into the aqueous solution, as described in Eqs. 5 and 6. According to Henry's law, as the CO₂ partial pressure increases, the dissolution of CO₂ into solution also increases. Therefore, in cases where the rate limiting step is CO₂ dissolution, CO₂ partial pressure will have a beneficial effect and conversely no effect when it is not. The use of high reaction temperatures was reported to be limited due to the negative impact on CO₂ dissolution. In this case, higher CO₂ pressure will counteract the negative impact of higher temperatures. This could allow the extended use of higher temperatures for increased Ca dissolution without the consequence of slowing CO₂ dissolution. This approach has been observed in some studies [84,85,92] where optimal CO₂ uptake was reported at high temperatures (100 °C). In each study a pressure >1 bar was used. It is unclear if the authors knew the higher pressures would enhance CO₂ dissolution and allow for higher temperatures, but the approach was taken, nonetheless. The higher pH levels of CRW were also stated to aid in the dissolution of CO₂ as it promoted the formation of CO₃²⁻ instead of HCO₃⁻. However the use of CRW alone will not enhance the initial dissolution to CO_{2(aq)} and will not allow the use of higher temperatures. Overall, increases in CO₂ pressure will be beneficial only if the rate limiting step is CO₂ dissolution and will allow for the use of higher reaction temperatures.

4.2. Ca dissolution

The dissolution or leaching of Ca ions is more commonly the rate limiting step in the initial stages of the carbonation reaction. This can be accelerated by increases in temperature. As previously mentioned, when temperatures are too high CO₂ becomes more stable in gaseous form and will not dissolve as easily, but this can potentially be remedied by using higher CO₂ pressure. However, increasing reaction temperature and CO₂ pressure can be economically and environmentally costly. Another approach is to increase the steel slag surface area by reducing the particle size. The decreased particle size creates more surface area to dissolve the Ca ions resulting in accelerated reaction kinetics. However, due to the high hardness, grinding steel slag to an appreciable particle size can also be energy intensive. A favorable slag composition (either chemical or phase) may accelerate the Ca dissolution reaction but from the current literature it is not clear.

4.3. Mass transfer

In order for the carbonation reaction to proceed, there is a mass transfer step that must occur. That is the movement of the H⁺ to the steel

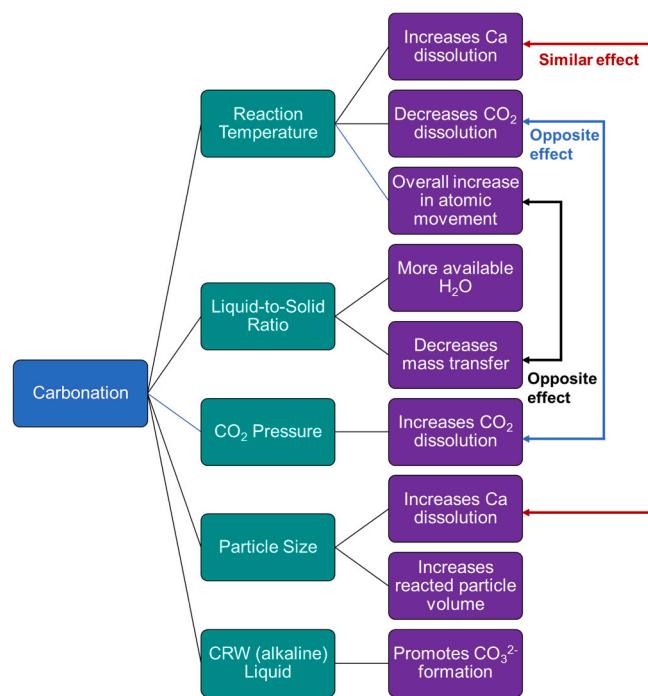


Fig. 9. Carbonation process parameters organized according to their kinetic effect on the chemical reactions with opposite and similar effects highlighted.

slag particle to leach the Ca²⁺ and the CO₃²⁻ to react with the Ca²⁺ to form CaCO₃. Excessive L/S will slow the mass transfer and lower the concentration of the reactants in solution. However, high L/S is beneficial since it provides more H₂O for CO₂ dissolution and the overall reaction sequence. The issue of slowed mass transfer can possibly be remedied by higher reaction temperatures as that will enhance the ion movement in solution. It is unclear at this point how the challenge of lower reactant concentration can be overcome by parameter optimization. Therefore, L/S should be monitored to ensure it is not hampering the reaction kinetics.

In the second stage of the carbonation reaction, the diffusion through the product layer becomes the rate limiting step. As the product layer grows, the diffusion distance increases and thus further slowing the reaction. This reaction step has been accurately modeled using the shrinking core model. The shrinking core model is typically used for the analysis of heterogeneous solid-fluid reactions. A primary assumption is that the reaction first occurs at the steel slag outer region and then gradually moves into the inner part, leaving behind completely reacted product. A kinetic study by Tu et al. [83] found the shrinking core model to be an accurate description of the carbonation process and therefore it is controlled by the diffusion through the product layer. At this point, the reaction proceeds very slowly and can only be accelerated by increases in temperature. None of the other process parameters, except for reaction time, will have an influence on the diffusion process. Interestingly, Kim et al. [62] discovered another solution to the product layer limitation. It was observed the product layer thickness was ~5 μm regardless of the particle size. Therefore, a smaller particle size would result in a larger consumption of the particle volume, given the product layer is always ~5 μm thick. This consistent product layer was observed experimentally, and particle size was observed to have the strongest influence on CO₂ uptake. Overall, there are some available process adjustments to overcome the slow reaction once the product layer forms such as increased temperature and decreased particle size. However, this step is still the largest hindrance towards complete and efficient CO₂ sequestration as indicated by the CO₂ uptake plateau observed with increased reaction times.

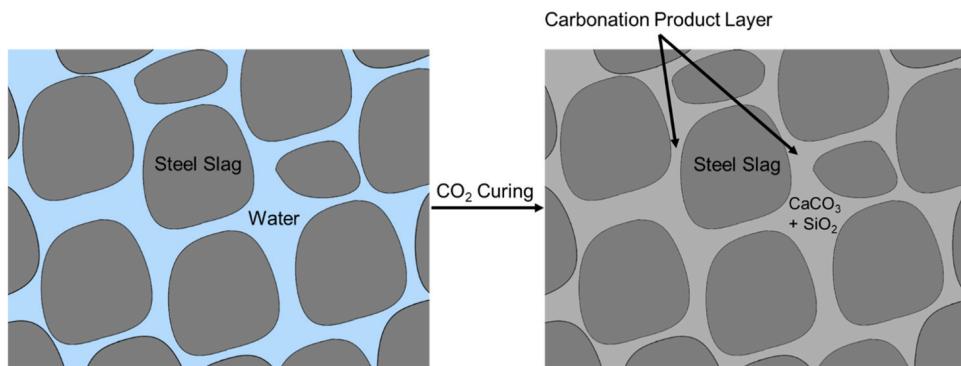


Fig. 10. Schematic diagram of the carbonation cured steel slag binder.

4.4. Collective kinetic effects

To maximize the CO₂ uptake of steel slag carbonation, the rate limiting step must be addressed with the appropriate process parameters. In Fig. 9, each process parameter is listed with their associated effect on the reaction kinetics. The shared impact on reaction kinetics has also been identified as either an opposite or similar effect on the process. From this visualization, the process parameters can be categorized based on their kinetic effect. It should be noted that temperature has several kinetic effects which overlap with the effects from other process parameters. So, when changing the reaction temperature, the impact on other process parameters should be considered. From the review in this section, it is clear no single process parameter can be used to overcome all the potential rate limiting steps, such as CO₂ dissolution, Ca dissolution, and diffusion through the carbonation product layer. Therefore, future research should focus on optimizing and fine-tuning the process parameters that specifically target each rate-limiting step. By addressing these steps individually, researchers can develop strategies to enhance CO₂ uptake. It is equally important to be cognizant of possible negative effects or negative interactions from process parameters which would cause the rate limiting step to shift (e.g. excessive L/S ratio).

5. Value-added use of carbonated steel slag

While the accelerated carbonation of steel slag provides a promising route for permanent CO₂ sequestration, it is evident that identifying the most efficient usage of carbonated slag is critical to close the loop of waste-to-resource economy. Upcycling carbonated slag for usage in the construction industry as a binder or aggregate, is a promising value-added application. During carbonation, the CaCO₃ and SiO₂ gel product layer can act as a binder [113–115]. This approach has successfully been used to produce carbon negative concrete blocks [116–119]. Furthermore, recent studies have found that cementitious properties of steel slag improve after carbonation [120]. The use of carbonated steel slag as either a binder or aggregate can limit the need to procure natural aggregate or CO₂ intensive ordinary Portland cement. To fully leverage the CO₂ benefits of steelmaking slag, post-carbonation applications should be explored further. Then, steel slag will not only act as a CO₂ capture medium but also be part of the waste-to-resource economy. The following sub-sections will summarize the previously mentioned carbonated steel slag applications as an aggregate or binder material.

5.1. Use of carbonated steel slag as aggregates

The carbonation of steel slag enables its use as aggregate in construction materials by addressing volume instability caused by the high f-CaO and f-MgO content. These free oxides are known to cause volume changes in steel slag which is the main issue with the use of steel slag as road or concrete aggregate [121]. As shown in Eqs. 1 and 2 the

carbonation process produces stable carbonates from the f-CaO and f-MgO in steel slag [78,109]. Carbonation effectively eliminates the volume instability of steel slag through this conversion, resulting in improved dimensional stability. The product layer produced during carbonation also limits the leaching of harmful heavy metal ions (Cr, V, Ba, etc.) [122–124]. The use of carbonated steel slag limits the need to procure or produce natural aggregate. Therefore, carbonated steel slag becomes a value-added resource in construction materials while also storing CO₂.

Since the higher FeO_x content of steel slag poses limitations on the reactivity and grindability, there is an attractive usage as aggregate in concrete to replace natural aggregate either partially or fully [125–127]. The use of carbonated steel slag as aggregate in concrete utilizes the higher hardness (due to the high FeO_x content) to match the properties of concrete with natural aggregate [128]. Rojas et. al replaced natural aggregate with treated EAF slag in concrete mixes which resulted in a more rigid concrete, with greater compressive and flexural strength [129]. As previously mentioned, the carbonation process converts unstable volume phases into stable CaCO₃ while the carbonation product layer reduces the risk of heavy metal leaching, both of which open the door to usage as aggregate.

Artificial aggregates obtained from BOF slag powder, subjected to carbonation showed superior mechanical properties compared to natural aggregates, even at full replacement, due to the CaCO₃ product layer [126]. Carbonated steel slag has also been used to replace fine aggregates obtained from crushing and sieving carbonated slag bricks while achieving a 20% increase in compressive strength compared to concrete with natural aggregates [130]. Bodor et. al [131] found carbonated steel slag used as a partial aggregate in cement mortars performed as well as reference cement mortar, while sequestering up to 190 kg CO₂/tonne slag. Furthermore, Mo et. al [125] used carbonated steel slag as aggregate in concrete and studied the mechanical properties and volume stability. The concrete containing steel aggregate was determined to have enhanced volume stability and comparable compressive strength to concrete made with natural aggregate. The effectiveness of steel slag as an aggregate in concrete has been demonstrated at lab scale meaning the technology is viable path to steel slag valorization and potentially applicable at industrial scale.

Another promising avenue for carbonated slag is their porous structure leading to water absorption and permeability. The higher water permeability is attractive for pervious concrete application [127]. Moreover, the higher FeO_x enables the use of carbonated EAF slag in radiation shielding applications where radiation attenuation depends heavily on the atomic mass of aggregates used.

5.2. Use of carbonated steel slag as a binder

As blast furnace usage declines with the increased use of DRI-EAF steelmaking, a suitable replacement to ground granulated blast furnace slag will need to be identified since it is widely used a

supplementary cementitious material [132]. If the carbonation process can be used to create a cementitious material from steel slag, it will significantly reduce the emissions of the concrete industry. The largest source of CO₂ emissions in concrete, which is cement, will be removed or partially removed using a carbon negative material.

Upcycling carbonated slag for usage in the construction industry as a binder is a promising value-added application. During carbonation, the CaCO₃ and SiO₂ gel product layer can act as a binder [113–115] as shown with the chemical reaction in Eq. 9 and the illustration in Fig. 10. From the schematic, it can be seen that space between the steel slag particles is consumed by the carbonation product layer, hence it acts as a binder in place of cement. However, for this application the L/S is much smaller than the previously discussed carbonation set-ups. Typically for use as a binder the L/S ranges from 0.15 – 0.4 [114]. Since the process requires CO₂ to cure the binder, it can only be done as a batch process which restricts the products that can be made. However, this approach has successfully been used to produce carbon negative concrete blocks [116–119]. A life cycle assessment (LCA) completed by Fuchs et. al [133] on their steel slag blocks found a CO₂ saving potential of 337 kg CO₂/m³. Ideally, block production should take place at the steelmaking facility to reduce slag transportation and potentially utilize steelmaking CO₂ emissions. Lei et al. [134] simulated the use of flue gas for carbonation curing and found it to be a feasible methodology for producing steel slag blocks. This approach would further utilize by-products from steelmaking and integrate block production on site to optimize CO₂ savings.

There has been success using steel slag as a binder in a CO₂ curing batch process, but carbonation may potentially create a pathway for usage in ready mix concrete. While steel slag is known for its low reactivity, recent studies have found that cementitious properties of steelmaking slag improve after carbonation [120] which opens the possibility to directly replace cement in concrete. The fine CaCO₃ particles resulting from the carbonation reaction provide nucleation sites to foster higher degree of hydration and improve mechanical properties [24,135]. Moreover, CaCO₃ can react with C₃A contained in Portland cement to form hydrates (namely, C₃A•CaCO₃•xH₂O) with higher stability and hardness compared with sulphaaluminate resulting from the hydration of plain Portland cement [136]. Cement blends that included carbonated BOF slag as a replacement for Portland cement showed almost comparable strength to Portland cement [117,136]. Chen et al. [137] found the relative compressive strength of cement blended with carbonated steel slag could reach 98% of pure Portland cement compressive and flexural strength after 28 days curing. Furthermore, in shorter curing times of 3 and 7 days, the blended cement compressive strength was 105% and 102% respectively with 98% and 103% flexural strength. Other work by Pan et al. [95] showed that cement with high carbonation conversion steel slag exhibited better mechanical strength than Portland cement in the early stages of curing. Both studies show the suitable mechanical properties of carbonated steel slag as a supplementary cementitious material, particularly as an early strength cement. Additionally, enhancing the carbonation efficiency not only improves the mechanical performance of carbonated steel slag cement but also facilitates the increased sequestration of CO₂, presenting a dual benefit for sustainable construction practices.

There are some challenges associated with the usage of steel slag in concrete. Firstly, the difficulty of grinding to a desirable granulometry due to the RO phase and FeO_x content. Secondly, CaCO₃ from carbonated slag can reduce the pH of the pore solution of concrete. The drop in pH of the pore solution increases the likelihood of reinforcement corrosion [138], thereby limiting the applications to non-reinforced concrete uses.

6. Outlook and future research needs

Our current review indicates that considerable work has been undertaken on the carbonation of steel slag over the past decade. Many of

Table 3
Summary of future research needs with research motivations provided.

Research Aspect	Research gap, motivations, opportunities, and embodied challenges
Reaction Time Optimization	While available literature indicates the presence of a threshold reaction time beyond which further reaction is less efficient for CO ₂ sequestration, how other process parameters—e.g., temperature, CO ₂ pressure, slag composition, etc.—influence the optimum reaction time is yet to be unveiled.
Reaction Temperature Optimization	Literature has shown that while raising the reaction temperature heightens CO ₂ uptake, excessive reaction temperature decreases CO ₂ solubility [80]. Therefore, identifying the optimum reaction temperature is phenomenal. Furthermore, the mutual interaction between the reaction temperature and other process parameters and their synergistic effects on CO ₂ uptake need more research. Analytical modelling of such interactions will be helpful to obtain reasonably fair initial values of optimum temperature, to reduce depended on the serendipity of trials and error experiments. Whereas the relationship between CO ₂ pressure and CO ₂ uptake is theoretically linear, this is not supported by available literature. More research is needed to unveil the actual relationship between CO ₂ pressure and the extent of the carbonation reaction and how this relationship is influenced by other process parameters.
CO ₂ Pressure Optimization	
Liquid-to-solid (L/S) Interaction with other Process Parameters	While the interference of the effect of temperature on CO ₂ uptake with that of L/S has been documented [98], how the effect of other process parameters on the carbonation reaction interferes with that of L/S is yet to be understood
Particle Size Optimization	Optimum slag granulometry is critical since finer particle sizes lead to higher degree of carbonation while excessively fine particle sizes can result in practical difficulties. Existing challenges include the extra burden of grinding slag to specific particle size to cope with the requirements of optimum carbonation and subsequent use as a value-added product.
Slag Composition Optimization	While it is understood that slag chemistry (particularly Ca and Mg content) greatly influences CO ₂ uptake [107,108], the optimal Ca and Mg contents remain poorly understood. The major challenge is the inherent variability of slag composition dictated by its raw materials and processes challenges associated with controlling the composition of steel slag. Consequently, research directly relating the concentration of Ca or Mg phases to CO ₂ uptake is limited.
Water Source	The effect of the content of the water source in alkalis such as NaCl needs further research. For carbonated slag aimed for utilization in the construction sector, caution should be exercised, given the NaCl's effect on the corrosion of steel reinforcement.
Steel Slag Valorization	Practical challenges include higher iron oxide content hindering energy efficient grinding to desirable granulometry, high content of free lime causing volumetric instabilities and high-water absorption levels. Future research needs include: (i) coupled use of carbonated steel slag powder and high reactivity binders; and (ii) use of carbonated steel slag as aggregate in non-structural applications or structural applications with no-corrosion likelihood, (iii) radiation-shielding concrete where the higher iron content will be advantageous for radiation attenuation.

Table 4

A summary of steel slag carbonation studies with CO₂ uptake highlighted and specific notes given.

Study	Carbonation Type	CO ₂ Uptake (kg/tonne)	Notes
Kurusta et al. [94]	Aqueous	246	<ul style="list-style-type: none"> Used a process that carbonates and grinds slag simultaneously Carbonation done at high pressures
Biava et al. [140]	Aqueous	135	<ul style="list-style-type: none"> Highly pressurized reactor Low L/S Ambient temperature
Kim et al. [62]	Aqueous	213	<ul style="list-style-type: none"> Used super critical CO₂ High pressure Ambient conditions
Nielsen et al. [77]	Aqueous	106	
Omale et al. [63]	Aqueous	58	<ul style="list-style-type: none"> Short reaction time used Ambient temperature High pressure Optimized temperature Fine particles High pressure
Polettini et al. [99]	Aqueous	465	
Polettini et al. [85]	Aqueous	536	<ul style="list-style-type: none"> High temperature and pressure Fine particles Low L/S High temperature and pressure Coarse particles Low L/S
Su et al. [84]	Aqueous	231	
Ghacham et al. [141]	Aqueous	52	<ul style="list-style-type: none"> Short reaction time High pressure Fine particles
Rushendra et al. [53]	Aqueous	82	<ul style="list-style-type: none"> Lower L/S Coarse particles
Baciocchi et al. [92]	Aqueous	403	<ul style="list-style-type: none"> High temperature and pressure Low L/S
Baciocchi et al. [92]	Aqueous	280	<ul style="list-style-type: none"> Similar to previously listed study but different mineralogy
Cheng et al. [91]	Aqueous	283	<ul style="list-style-type: none"> Fine particles Cold rolling wastewater was used Ambient temperature & pressure
Cheng et al. [112]	Aqueous	277	<ul style="list-style-type: none"> Cold rolling wastewater was used Ambient temperature & pressure
Cheng et al. [142]	Aqueous	290	<ul style="list-style-type: none"> 40% CO₂ used Optimized temperature Fine particles
Cheng et al. [82]	Aqueous	270	<ul style="list-style-type: none"> Similar to previously listed study
Bonenfant et al. [143]	Aqueous	17.4	<ul style="list-style-type: none"> Ambient temperature Lower L/S
Kurusta et al. [94]	Gas	83	<ul style="list-style-type: none"> Used a process that carbonates and grinds slag simultaneously Carbonation done at high pressures
Rushendra et al. [53]	Gas	11.1	<ul style="list-style-type: none"> Ambient temperatures and high pressure
Santos et al. [100]	Gas	35	<ul style="list-style-type: none"> High temperature and pressure Fine particles
Santos et al. [100]	Gas	120	<ul style="list-style-type: none"> Similar to previously listed study but higher Ca content in slag
Wang et al. [102]	Gas	73	<ul style="list-style-type: none"> Moderate temperatures
Wang et al. [102]	Gas	83.8	<ul style="list-style-type: none"> 75% CO₂ used High temperature

these studies have focused on exploring the CO₂ sequestration potential of steel slag and the characterization of the carbonated by-products. However, there is a notable gap in the detailed understanding of carbonation reaction kinetics, which warrants further investigation. More specifically, the literature lacks an intricate understanding of the interactions between process parameters. While there is a reasonable comprehension of these parameters individually, their collective interactions have not been scrutinized at a detailed level. Kim et al. [62] conducted an ANOVA study to show the interaction between various process parameters. In a comparable statistical approach, Polettini et al. [85] investigated similar aspects without unveiling the mechanistic effects of the interactions among these process parameters. This review has extensively collected insights on the effect of process parameters for carbonation and identified some possible kinetic interactions between them, which can be used as a new perspective for future research and further process optimization as detailed in Table 3. The collective insights from Table 3 underscore the intricate challenges in optimizing steel slag carbonation. The need for a comprehensive understanding of the interplay among process parameters, such as temperature, CO₂ pressure, and slag composition, is emphasized, with a recognition of the limitations in determining optimal values. Challenges include the complex relationship between temperature and CO₂ uptake, unexplored process parameter interactions with reaction time, and the non-linear correlation between CO₂ pressure and carbonation extent. Variability in steel slag composition, especially Ca and Mg content, poses a significant limitation on understanding carbonation efficiency. This emphasizes a need for lab-scale testing using controlled composition slag samples. Future research should prioritize analytical modeling, explore interdependencies among process parameters, and develop practical applications for steel slag carbonation as aggregate, thereby advancing its sustainability and value-added potential. As previously noted in this review, increased carbonation efficiency typically leads to improved mechanical performance of steel slag in concrete products. Therefore, the utilization of carbonated steel slag products in concrete also stands to benefit from an increased understanding of the carbonation process while potentially being a carbon negative product.

Given the comprehensive examination of studies quantifying CO₂ uptake, as presented in Table 4, it can be deduced that the maximum observed CO₂ uptake is 536 kg CO₂ per tonne of steel slag. However, it appears that ~275 kg CO₂ per tonne of steel slag is the mostly commonly reported metric for aqueous carbonation. These values should be considered in the grand scheme of CO₂ emissions during iron and steelmaking. From the global average, 1 tonne of steel produced emits ~2 tonnes of CO₂ [3] and approximately 0.170 tonnes of steelmaking slag [139]. This means for each tonne of steel produced 0.035 – 0.085 tonnes of CO₂ can be captured (using current carbonation process technology). This is clearly a small percentage of the CO₂ emitted during the iron and steelmaking process. It should be considered that CO₂ emissions are expected to decrease by up to 70% as the industry transitions to DRI-EAF steelmaking, which will increase the percentage stored by steel slag. However, the CO₂ uptake potential of steel slag alone will not completely offset the CO₂ emissions during iron and steelmaking. Further benefits can be realized by the usage of carbonated steel slag in the concrete industry.

Considering that 1 tonne of concrete emits 180 kg of CO₂, the usage of carbonated aggregate in concrete could have a considerable impact on the carbon footprint. Natural aggregate does not significantly contribute to the CO₂ emissions from concrete since emissions mostly come from the cement portion. However, the use of ‘carbon negative’ aggregate can offset the CO₂ emissions. Concrete is typically 75% aggregate (fine and coarse) which means 750 kg would be used in 1 tonne of concrete. Pan et al. [109] showed that it is viable to carbonate the steel slag at the site of steelmaking using CRW and flue gas, which will be assumed in this case. If a 75% replacement ratio is used and assuming 100 kg of CO₂ per tonne of slag is captured, the net CO₂ captured would be ~54 kg CO₂ after accounting for the emissions of the carbonation process [144]. This

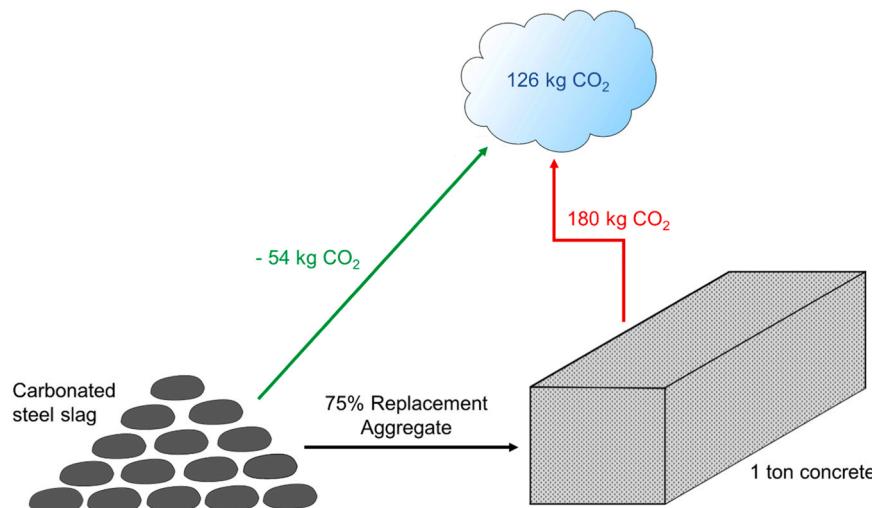


Fig. 11. Diagram showing the usage of carbonated steel slag at a replacement ratio of 75% can lead to 30% reduction in net concrete CO₂ emissions.

replacement ratio will lead to a 30% reduction in CO₂ for a tonne of concrete. A diagram of this scenario is shown in Fig. 11. Of course, if a higher replacement ratio is used, the reduction will increase. It is also important to note that Pan et al. [109] showed that the CO₂ sequestration potential of steel slag in these carbonation conditions is nearly 275 kg of CO₂ per tonne of slag. If the process is further studied to reach this potential, concrete could be made carbon neutral.

7. Conclusions

The carbonation of steel slag is a complex process with several possible rate limiting steps. This scientific review has provided a comprehensive analysis of the key factors influencing the carbonation process, their effect on the chemical reaction kinetics, and the interaction between process parameters' kinetic effects. Furthermore, by outlining the reaction steps, the study provided insights into underlying mechanisms, allowing for a deeper understanding of the sequence of reactions and what role each process parameter plays. This knowledge can aid in developing predictive models, identifying rate-limiting steps, and targeting strategies to enhance carbonation efficiency. Ultimately, the presented perspective can guide future process optimization and the successful implementation of carbonation-based technologies.

The review highlighted the significance of process parameters—such as reaction time, reaction temperature, L/S, CO₂ partial pressure, and particle size—in optimizing the carbonation efficiency and CO₂ uptake. Increasing temperature was identified to have several effects on the reaction kinetics, such as increasing Ca dissolution, decreasing CO₂ dissolution, and increasing atomic movement. From this review, it was determined that each of these effects interacts with other process parameters (particle size, CO₂ pressure, L/S, respectively). Future studies should be carried out to study the process parameter interactions mechanistically, and possibly elucidate other interactions not yet known for complete process optimization. This review also highlights the usage of CRW as a liquid in aqueous carbonation by providing the needed alkalinity to accelerate the carbonation reaction. When combined with flue gas from steelmaking, this presents a logically easy opportunity for CO₂ capture onsite at steel mills.

It is evident that the carbonation of steel slag has the potential to address both environmental and economic challenges of decarbonization. To fully leverage the CO₂ capture benefits, carbonated steel slag should be converted into a durable binder material or aggregate. Rough estimations from this review have shown the potential usage of carbonated steel slag as a carbon negative aggregate product. Carbonated steel slag offers a sustainable solution for reducing CO₂ emissions,

conserving natural resources, and enhancing the performance of construction materials. However, research will need to be carried out with the goal of developing a mechanically sound and carbon negative aggregate, which can be facilitated by carbonation process parameter optimization. Additionally, there is a need to develop standardized testing protocols and regulations for carbonated steel slag as a construction material, which will be instrumental in promoting its widespread acceptance and implementation.

The carbonation of steel slag holds great promise for achieving carbon neutrality ambitions, not only in the steel industry, but also in the cement and concrete industry which is another hard-to-abate sector. As such, the value-added use of carbonated steel slag can contribute to fostering waste-to-resource economy and enabling faster attainment of sustainable development goals. Continued research and collaboration among scientists, engineers, and policymakers are crucial for advancing this field and realizing its full potential. By harnessing the massive potential for carbonation, we can transform steel slag from a waste product into a valuable resource, contributing to a greener future and a more sustainable steel sector.

CRediT authorship contribution statement

Ousmane A. Hisseine: Writing – review & editing, Conceptualization. **Adedapo Noah Awolayo:** Writing – review & editing, Conceptualization. **Christopher DiGiovanni:** Writing – review & editing, Writing – original draft, Funding acquisition, Conceptualization.

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.

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Appendix A. review methodology

The scientific documents contained in this review originate from three primary sources: (i) Scopus database, (ii) research materials obtained from our collaborators in the steel industry, and (iii) secondary references accessed from within the documents in the first two sources. Specifically, the Scopus search was undertaken using a *title-abstract-keyword* search query as follows: TITLE-ABS-KEY ((carbonation OR "CO₂ Sequestration" OR "CO₂ capture"). The search was first refined by limiting it to articles treating direct carbonation, given its economic feasibility compared to indirect carbonation. The study was further refined — in terms of the valorization of slag as a construction material — by excluding documents on geopolymers concrete or alkali activated concrete, thereby maintaining a focus on the application of carbonated slag in well-established mainstream conventional concrete.

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