



# Accelerated Carbonation of Steel Slag and Their Valorisation in Cement Products: A Review

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Mineral carbonation emerges as a promising technology to tackle a contemporary challenge: climate change. This method entails the interaction of carbon dioxide with metal-oxide-bearing materials to produce solid carbonates resembling common substances (chalk, antacids, or baking soda). Given that steelmaking industries contribute to 8% of the global total emissions annually, the repurposing of their by-products holds the potential to mitigate CO<sub>2</sub> production. Steel slag is a by-product of the metallurgical industry which is suitable for capturing CO<sub>2</sub> due to its chemical composition, containing high CaO (24%–65%) and MgO (3%–20%) amounts, which increases the reactivity with the CO<sub>2</sub>. Moreover, the carbonation process can improve the hydraulic and mechanical properties of steel slag, making this by-product interesting to be reused in building materials. Different studies have developed in the last years addressing the possibilities of reducing the environmental impact of steel products, by CO<sub>2</sub> sequestration. This study is dedicated to reviewing the basics of mineral carbonation applied to steel slag, along with recent advancements in research. Special emphasis is placed on identifying parameters that facilitate the reactions and exploring potential applications for the resulting products. The advantages and disadvantages of steel slag carbonation for the industrialization of the process are also discussed.

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## INTRODUCTION

In recent decades, a critical challenge facing modern society is the surge in greenhouse gas (GHG) emissions, leading to climate change with a global temperature increase of 0.8°C over the last century. Anthropogenic activities, including deforestation, fossil fuel consumption, and the meat industry, primarily contribute to the rise in these gases, notably CO<sub>2</sub>. With its concentration reaching 419 ppm in 2021 (compared to 360 ppm in the 19th century), urgent measures are required to address this environmental concern (Azadi et al., 2019).

Given this scenario, it becomes imperative to explore alternative strategies for mitigating CO<sub>2</sub> emissions and reducing dependence on fossil fuels. Carbon capture and storage (CSS) technology emerges as an economically feasible strategy, potentially preventing the emission of eight billion tonnes of CO<sub>2</sub> by 2050 (IEA50, 2021). This technology encompasses geological, oceanic, and mineral storage, with carbon mineralization being a key component.

Carbon mineralization involves a reaction between CO<sub>2</sub> and Ca and Mg-minerals, forming stable carbonated products (CaCO<sub>3</sub> and MgCO<sub>3</sub>) and providing a permanent solution for CO<sub>2</sub> storage. This process accelerates the natural weathering of rocks, transforming it from a millennia-long

process to mere minutes or hours. The resulting by-products are stable solids with long-term storage capacity (Sanna et al., 2014).

Direct carbonation (Lackner et al., 1995) occurs in one step through a direct gas-solid or gas-liquid-solid reaction. While effective, it requires high temperature and pressure for acceleration. In contrast, indirect carbonation involves two reactions: the extraction of metal ions (mainly Mg and Ca) from corresponding containing phases and subsequent carbonation between CO<sub>2</sub> and the resulting solution. This route is more viable owing to its gentle reaction conditions, elevated carbonation effectiveness, and the generation of high-value secondary materials (Lee et al., 2021).

Notably, industrial wastes like steel slag, with high alkalinity and reactivity, are suitable for this process. The carbonation potential of these wastes for CO<sub>2</sub> sequestration was evaluated suggesting that mineral carbonation could reduce global anthropogenic CO<sub>2</sub> emissions by 12.5% (Pan et al., 2020). Waste materials such as cement waste, red mud, municipal incinerator ash, and steel slag are viable for the carbonation reaction, addressing both environmental concerns and waste disposal challenges (Izumi et al., 2021).

Steel slag, a by-product of steel manufacturing, has garnered significant attention for its potential applications in various sectors, particularly in construction. The importance of steel slag lies in its ability to serve as an alternative to natural resources, which are becoming increasingly scarce due to overexploitation. Its utilization in construction offers a multifaceted approach to resource conservation and environmental sustainability (Sun and Wang, 2024). Substituting traditional aggregates can effectively conserve finite natural resources that are both unevenly distributed and rapidly diminishing (Zhang et al., 2024). This effort holds particular significance in the context of sustainable development, where the responsible stewardship of natural resources is paramount and practices to mitigating the environmental impact associated with conventional construction activities are fundamental. In addition, the extraction and processing of natural aggregate entails significant energy consumption and environmental disturbance, including habitat destruction and landscape alteration. By reducing reliance on these materials through the utilization of steel slag, construction projects can minimize their ecological footprint and lessen pressure on sensitive ecosystems. Finally, repurposing, and harnessing steel slag can yield significant economic advantages, curbing waste and diminishing expenses linked with industrial by-products management.

Steel slag is produced in large quantities, with availability depending on the production of steel in each region.

As per the U.S. Geological Survey, specific data regarding actual ferrous slag production in the U.S. is not available. However, it is estimated that domestic slag sales reached approximately 17 million tons, valued at around \$460 million in 2021. This slag was processed by 28 companies serving operational iron and steel factories or reprocessing previously deposited slag wastes at approximately 124 dedicated plants across 33 states (Cris Candice Tuck, 2022). In the context of

India, each tonne of steel production generates about 200 kg of steel slag (Ministry of India, 2023). Owing to the lack of efficient disposal methods for steel slag, substantial slag piles have emerged around steel plants, evolving into a significant source of water, air, and land pollution (Mayes et al., 2008).

In Europe, an annual generation of approximately 45 million tons of slag (originating from iron and steel waste) is reported (EUROSLAG, 2024). This manufacturing originates from the procedures of ore conversion into iron, the conversion of hot iron into steel, and the melting of scrap in an electric arc furnace, or from the subsequent refining of crude steel.

It is crucial to underline that the availability of steel slag can depend on various conditions, such as the rate of steel production, the type of steel being manufactured, and the methods employed for slag disposal. Consequently, the precise availability of steel slag may fluctuate over time and across different locations.

Calculations indicate that utilizing 4.7 tons of steel slag could result in the absorption of one ton of CO<sub>2</sub>, producing 2.3 tons of CaCO<sub>3</sub>. If the entirety of the slag were employed for CaCO<sub>3</sub> production, it could lead to the consumption of 53 million tons of CO<sub>2</sub> and the generation of 120 million tons of CaCO<sub>3</sub> (Eloneva et al., 2012). Other projected statistical estimates suggest that utilizing all steel slag for mineral carbonation could result in the potential sequestration of between 138 and 209 million tons of CO<sub>2</sub> annually, constituting approximately 9.1%–10.4% of the iron and steel industry's total CO<sub>2</sub> emissions (Pan et al., 2017) and evaluate that the total CO<sub>2</sub> potential for mineralization from 2020 to 2,100 will fall within the range of 26–42 gigatons (Myers and Nakagaki, 2020).

Despite the promising potential of carbon mineralization, its industrial applications face obstacles related to energy and cost consumption. Overcoming these challenges requires a focus on recovering value-added by-products derived from carbonation and generating revenues. The carbonated products can find applications in construction materials, paper, and paint filler. This review focuses on offering a comprehensive overview of the scientific literature concerning steel slag carbonation. It delves into *ex-situ* mineral carbonation methods, analyses slag properties and examines their effects on dissolution and carbonation mechanisms. Additionally, it outlines the potential reuse of carbonated slag in the cement industry, emphasizing how the carbonation process can enhance the hydraulic properties of this by-product. Finally, it also highlights the advantages and challenges associated with the diffusion process. The work is not overly specialistic because it is directed towards all scientific communities engaged in exploring available circular economy strategies to mitigate greenhouse gas emissions.

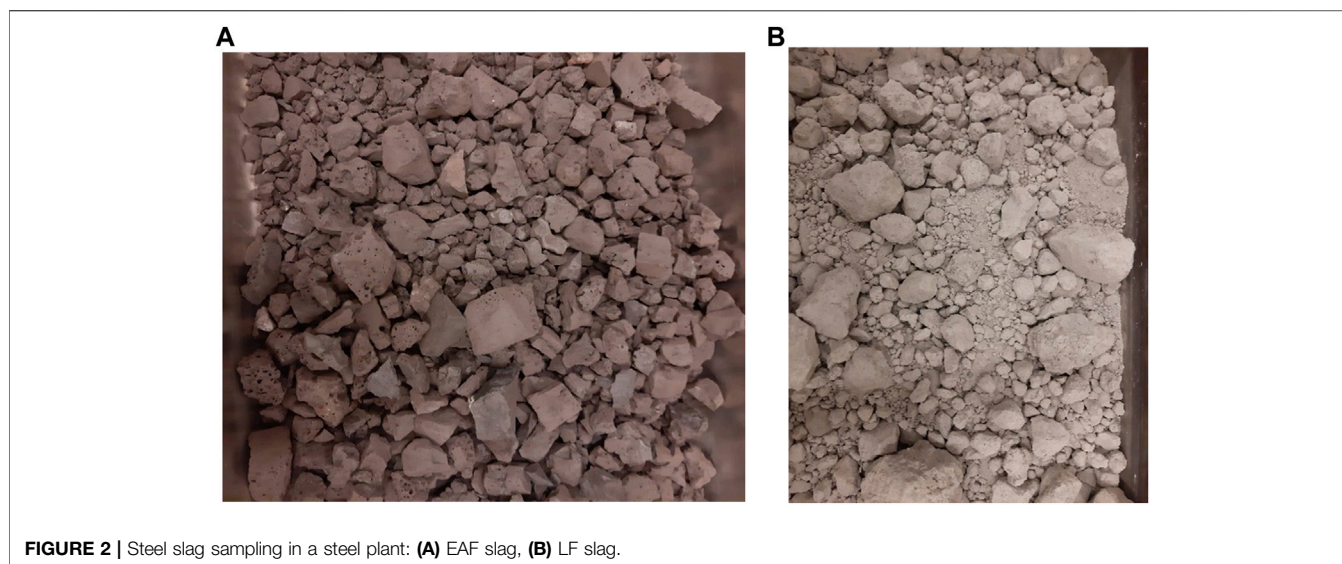
## REVIEW METHODOLOGY

To investigate the literature data about steel slag carbonation, text research was addressed to academic journals and scientific publications by Scopus.

Relevant keywords in the abstract and paper titles “steel slag” and “carbonation” were used. 437 papers were found on January 18, 2024. Among them, 25 works are review papers.







**FIGURE 2** | Steel slag sampling in a steel plant: **(A)** EAF slag, **(B)** LF slag.

*Steel Slag Characterization* section characterizes steel slag by mineralogical and chemical composition, *Carbonation and Reaction Parameters* section describes the carbonation reaction and the influence of reaction parameters, *Kinetics Models* section compares the two main kinetic models, proposed by literature data, *Methodologies to Improve the Mineral Carbonation Efficiency* section describes different methodologies to improve the carbonation degree and *Carbonated Slag Application in the Cement Industry* section takes into account the reuse of carbonated steel slag in the cement industry as aggregates and supplementary cementitious material and as soil stabiliser, *Advantages and Limitations of Steel Slag Carbonation* section resumes the advantages of carbonation process for steel slag and *Conclusion and Perspectives* section concludes the manuscript.

## STEEL SLAG CHARACTERIZATION

Steel slag is a waste of steel plants, comprising various types such as Electric Arc Furnace (EAF) slag, Argon Oxygen Decarburization (AOD) slag, Basic Oxygen Furnace (BOF) slag, Ladle Refining Furnace (LF) slag, and Blast Furnace (BF) slag (Zhang et al., 2023). The categorization is based on the diverse steelmaking procedures employed. **Figure 2** shows what the steel slag looks like after its production. Indeed, the chemical and physical properties of steel slag are influenced by different production conditions and feedstock inputs. For example, in the process of EAF steelmaking and oxygen converter steelmaking, CaO and MgO are added to the smelting furnace to remove dangerous elements such as P and S in molten steel. Oxygen is used to remove impurities and carbon into Basic-Oxygen Furnaces and Electric Arc Furnaces. By this treatment, elements such as Al, Si, Mn, and P are oxidized to respective oxides. The last step of the steelmaking process is the refining by desulfurization process to remove impurities, oxygen, nitrogen, hydrogen, and carbon in the ladle furnace, where LF slag is a

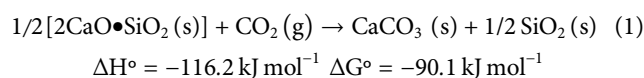
carbon by-product (Humbert and Castro-Gomes, 2019; Luo and He, 2021; Zhang et al., 2023).

Steel slag encompasses various valuable components, including 24%–65% CaO, 10%–32% SiO<sub>2</sub>, 3%–20% Al<sub>2</sub>O<sub>3</sub>, 0%–37% Fe<sub>2</sub>O<sub>3</sub>, and 3%–20% MgO and the chemical composition, based on the type of slag, is reported in **Table 1**. As illustrated, the chemical composition is influenced by the origin of slag. EAF and AOD slag may have elevated levels of Cr, posing a risk of significant pollution if it is directly discharged into soil and water resources (Chen et al., 2021a; Zhang et al., 2023).

CO<sub>2</sub> sequestration is influenced by the mineralogical composition of steel by-products (see **Table 2**).

Except for BF slag, which is completely amorphous, other slags contain mainly Ca and Mg-phases as larnite, periclase and Ca-Mg-Al silicate. The mineral composition of steel slag primarily consists of 15%–25% dicalcium silicate (Ca<sub>2</sub>SiO<sub>4</sub>, C<sub>2</sub>S), 20%–25% tricalcium silicate (Ca<sub>3</sub>SiO<sub>5</sub>, C<sub>3</sub>S), 40%–45% RO (inert phases and combination between Mg, Fe and Ca), along with small quantities of free CaO and MgO (f-CaO and f-MgO). The primary constituents of Electric Arc Furnace slag (EAF) are CaO, SiO<sub>2</sub>, and Fe<sub>2</sub>O<sub>3</sub>. In BOF and EAF slag Fe-phases are identified as FeO or Fe<sub>2</sub>O<sub>3</sub> due to high FeO quantity. Argon Oxygen Decarburization slag (AOD) contains a substantial amount of Cr<sub>2</sub>O<sub>3</sub>, and its direct discharge poses a significant threat to water and soil resources (Chen et al., 2021b).

Mineral compositions of steel slag have a considerable influence on CO<sub>2</sub> sequestration and the main reactive species are periclase (MgO), calcium silicate (C<sub>3</sub>S and C<sub>2</sub>S) and merwinite. Thermodynamics calculations demonstrate that the carbonation of these phases is a spontaneous process ( $\Delta G$  is negative) at standard temperature and pressure as reported in the formula (1) (Ragipani et al., 2021; Zhang et al., 2023):



**TABLE 1** | Chemical composition of steel slag and pH range.

Steel slag	CaO	MgO	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	MnO	Cr <sub>2</sub> O <sub>3</sub>	pH	References
Units	[%wt]	[%wt]	[%wt]	[%wt]	[%wt]	[%wt]	[%wt]	[%wt]		
BF slag	28–51	4%–11%	6–20	26–40	<2	<2	<1	<1	11–10	Chang et al. (2017), Ukwattage et al. (2017), Luo and He (2021), Mei et al. (2021), Mohammed (2021), Mohammed et al. (2021), Tran et al., 2021; Younsi (2022), Li et al. (2023a)
BOF slag	42–55	5–11	2–6	10–23	10–37	<2	0.7–5	<1	13–11	De Windt et al. (2011), Salman et al. (2014), Ko et al. (2015), Quaghebeur et al. (2015), Costa et al. (2016), Yadav and Mehra (2017a), Chang et al. (2017), Chang et al. (2018), Chen et al. (2019), Librandi et al. (2019), Wang et al. (2019), Ragipani et al. (2020), Li et al. (2021), Luo and He (2021), Ragipani et al. (2021), Zhang et al. (2023)
EAF slag	24–40	3–15	6–12	6–32	20–30	<2	0.4–7	1–6	12–10	Suer et al. (2009), Capobianco et al. (2014), Gurtubay et al. (2014), Salman et al. (2014), Librandi et al. (2019), Tong et al. (2019), Chen et al. (2020a), Wang et al. (2021a), Luo and He (2021), Ragipani et al. (2021), Biava et al. (2023), Zhang et al. (2023)
LF slag	38–65	3–20	2–20	12–30	0.7–7	<1	0.4–4	<1	13–11	Araizi et al. (2016), Luo and He (2021), Zhang et al. (2021), Xu and Yi (2022a), Xu and Yi (2022b), Zod et al. (2022), Biava et al. (2023), Wu et al. (2023), Zhang et al. (2023)
AOD slag	34–60	2–10	1–12	24–32	0.5–12	<1	<1	<3	13–12	Santos et al. (2013), Capobianco et al. (2014), Salman et al. (2014), Baciocchi et al. (2015a), Moon and Choi (2018), Moon and Choi (2019), Park et al. (2020), Biava et al. (2023), Zhang et al. (2023)

**TABLE 2** | Mineralogy of slag.

Steel slag	Mineralogical phases	References
BF slag	Amorphous, SiO <sub>2</sub> , CaSO <sub>4</sub>	Ukwattage et al. (2017), Moon and Choi (2018), Moon and Choi (2019), Tran et al. (2021), Li et al. (2023a)
BOF slag	C <sub>2</sub> S, C <sub>3</sub> S, C <sub>2</sub> AF, RO phase (FeO-MgO-CaO-FeO), MgO, f-CaO, C <sub>12</sub> A <sub>7</sub> , β-C <sub>2</sub> S, FeO, γ-C <sub>2</sub> S	Baciocchi et al. (2015b), Quaghebeur et al. (2015), Yadav and Mehra (2017b), Chang et al. (2017), Wang et al. (2019), Ragipani et al. (2021), Mei et al. (2022), Zhang et al. (2023)
EAF slag	γ-C <sub>2</sub> S, β-C <sub>2</sub> S, Ca <sub>7</sub> Mg(SiO <sub>4</sub> ) <sub>4</sub> , Mg <sub>2</sub> Si <sub>2</sub> O <sub>6</sub> , Ca <sub>3</sub> Mg(SiO <sub>4</sub> ) <sub>2</sub> , CaSiO <sub>3</sub> , MgO, Ca <sub>2</sub> Al <sub>2</sub> SiO <sub>7</sub> , Fe <sub>2</sub> O <sub>3</sub> , MgFe <sub>2</sub> O <sub>4</sub> , FeO, Fe <sub>3</sub> O <sub>4</sub> , SiO <sub>2</sub> , MnO-FeO	Baciocchi et al. (2015a), Baciocchi et al. (2015b), Wang et al. (2021a), Ragipani et al. (2021), Mei et al. (2022), Zhang et al. (2023)
LF slag	C <sub>2</sub> S, CaO-Al <sub>2</sub> O <sub>3</sub> ·2SiO <sub>2</sub> , CaS, Al <sub>2</sub> O <sub>3</sub> , Ca <sub>12</sub> Al <sub>14</sub> O <sub>33</sub> (C <sub>12</sub> A <sub>7</sub> ), MgO, β-Ca <sub>2</sub> SiO <sub>4</sub> , γ-Ca <sub>2</sub> SiO <sub>4</sub> , Ca <sub>2</sub> Al <sub>2</sub> SiO <sub>7</sub>	Araizi et al. (2016), Zhang et al. (2021), Xu and Yi (2022a), Zod et al. (2022), Zhang et al. (2023)
AOD slag	γ-C <sub>2</sub> S, β-C <sub>2</sub> S, FeMg <sub>2</sub> O <sub>4</sub> , FeCr <sub>2</sub> O <sub>4</sub> , C <sub>3</sub> S, Ca <sub>14</sub> Mg <sub>2</sub> (SiO <sub>4</sub> ) <sub>8</sub> , CaF <sub>2</sub> , MgO, Ca <sub>3</sub> MgSi <sub>2</sub> O <sub>8</sub> , Ca <sub>3</sub> Al <sub>2</sub> O <sub>6</sub> , MgSiO <sub>3</sub> , MgCr <sub>2</sub> O <sub>4</sub> , Ca <sub>3</sub> Fe <sub>2</sub> (SiO <sub>4</sub> ) <sub>3</sub>	Baciocchi et al. (2015a), Moon and Choi (2018), Moon and Choi (2019), Park et al. (2020), Mei et al. (2022), Zhang et al. (2023)

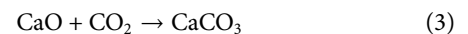
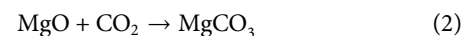
## CARBONATION AND REACTION PARAMETERS

Carbonation reaction occurs in nature but its kinetic is slow, also requiring millions of years. It is divided into two pathways, as shown in **Figure 3**. Direct carbonation happens when a step reaction can be highlighted, whereas, in the indirect process, two subsequent steps can be enhanced: extraction of alkaline metals by acidic solvents from steel slags and carbonation.

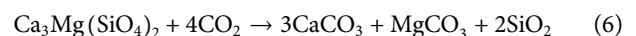
### Direct Carbonation

#### Direct Gas-Solid Carbonation

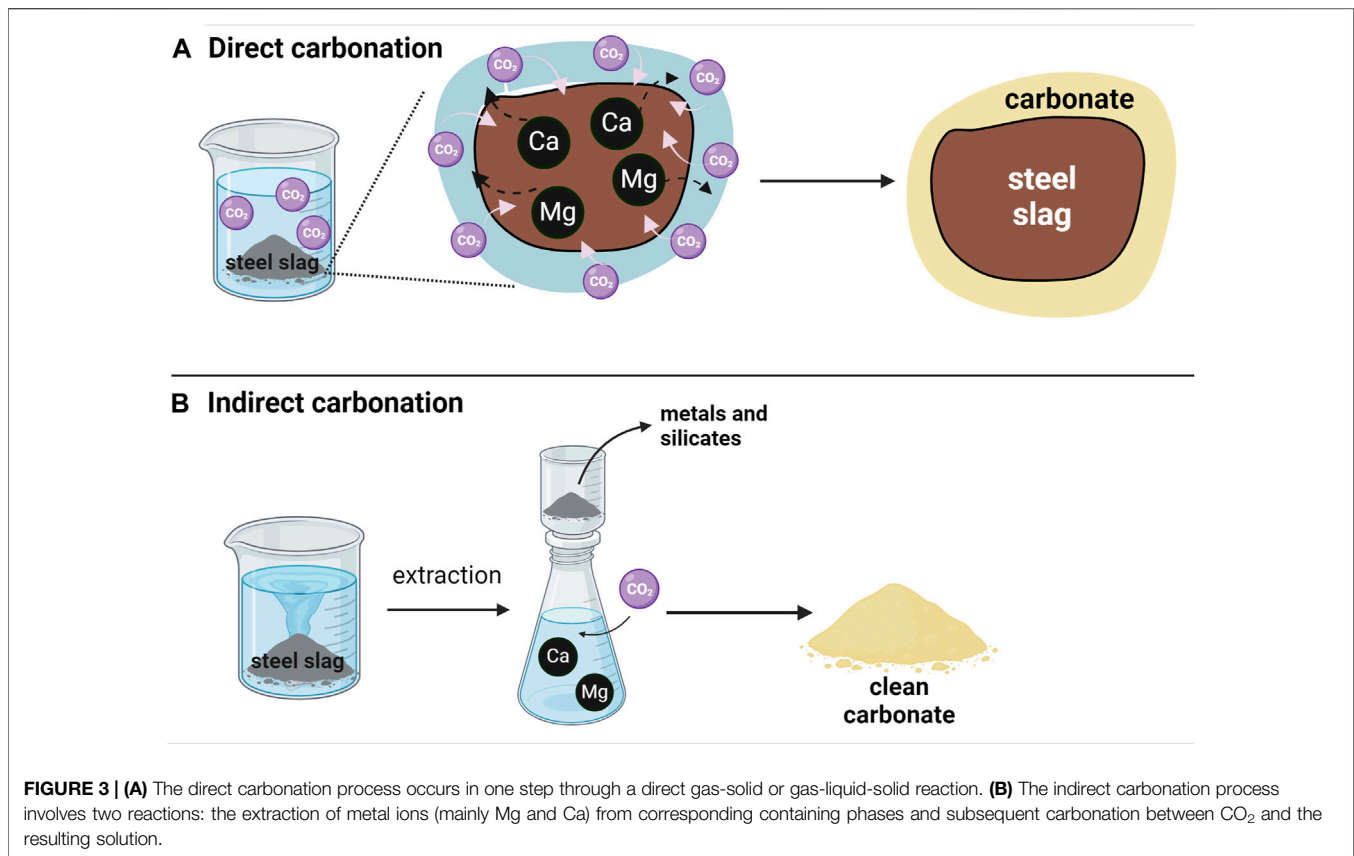
First proposed by Lackner (Lackner et al., 1995), this method closely resembles the natural weathering process. The reactions entail oxides and hydroxides containing calcium and magnesium reacting with CO<sub>2</sub> to form calcite (CaCO<sub>3</sub>), magnesite (MgCO<sub>3</sub>), and/or dolomite (CaMg(CO<sub>3</sub>)<sub>2</sub>) as shown in the formulas 2–4:



Moreover, some studies have demonstrated that also larnite (Ca<sub>2</sub>SiO<sub>4</sub>) and merwinite (Ca<sub>3</sub>Mg(SiO<sub>4</sub>)<sub>2</sub>), contained in steel slag, can react with CO<sub>2</sub> as follows (Chen et al., 2021a; Yadav and Mehra, 2021) in the Eqs 5, 6:



The capacity CaO and MgO to generate carbonates depends on the degree of chemisorption of carbonate ions, the solubility product constant in aqueous carbonation, and the presence of alkaline active sites on the particle surface (Rahmanianzaki and Hemmati, 2022).



Under specific conditions, the direct gas-solid pathway can achieve a maximum sequestration capacity of more than 10 g of CO<sub>2</sub> per kilogram of steel slag at room temperature (Rushendra Revathy et al., 2016).

This value can increase if the temperature increases (Tian et al., 2013). Nevertheless, the reaction rate might be sluggish owing to the restricted interaction between the gas and solid phases, necessitating elevated temperatures for the process to proceed at a practical rate (Sanna et al., 2014).

Despite these obstacles, the direct gas-solid carbonation method holds the advantage of simplicity and the prospect of achieving high carbonation efficiency. This area remains an active focus of research in the field of CO<sub>2</sub> sequestration.

### Direct Aqueous Carbonation

Direct aqueous carbonation is an extensively studied method. It is a more complex process in comparison to direct gas-solid carbonation because it involves some steps: dissolution of carbon dioxide (CO<sub>2</sub>) in water, the formation of carbonic acid and the reaction with metal oxides to produce carbonates (Veetil and Hitch, 2020).

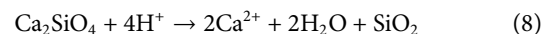
The reaction can be represented as follows in the formula 7:



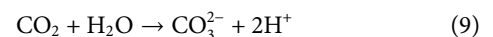
Where M represents a divalent metal cation, such as calcium (Ca) or magnesium (Mg), commonly present in steel slag.

This reaction can also involve calcium species other than CaO, such as C<sub>3</sub>S and C<sub>2</sub>S (β-C<sub>2</sub>S and γ-C<sub>2</sub>S) which hydrate forming Ca(OH)<sub>2</sub> and calcium-silicate-hydrates (C-S-H) (Chen et al., 2021a). At the beginning, the pH value of this system is alkaline but, when the CO<sub>2</sub> is dissolved in water, carbonic acid is formed at equilibrium and the fully dissociation of CO<sub>3</sub><sup>2-</sup> decreases the pH (Yadav and Mehra, 2021). The anion reacts with Ca<sup>2+</sup> or Mg<sup>2+</sup> ions forming Ca(or Mg)CO<sub>3</sub> (Schnabel et al., 2021) as shown in following reactions 8–10:

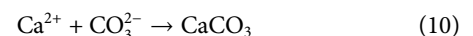
a) The leaching of minerals:



b) The carbon dioxide dissolution leads to the liberation of carbonate ions:



c) The initiation and expansion of mineral carbonate formations:



These three steps occur concurrently within a single reactor. The procedure is conducted at relatively low temperatures and pressures, making it less energy-intensive compared to alternative carbonation methods. However, the reaction rate may be sluggish due to the limited solubility of CO<sub>2</sub> in water and the passivation of the metal oxide surface by the formed carbonates.

To address these challenges, various process modifications have been suggested as the use of chelating agents, some pre-treatments, autoclave curing and continuous wet carbonation (Wu et al., 2017; Yadav and Mehra, 2021).

Despite the obstacles, direct aqueous carbonation has demonstrated a carbonation efficiency higher than 30% under specific conditions. During accelerated aqueous carbonation, steel slag has exhibited an impressive capacity for carbon fixation, reaching effective CO<sub>2</sub> storage ranging from 130 to 330 g CO<sub>2</sub> (kg slag)<sup>-1</sup> (Wang F. et al., 2021).

### Influence of Reaction Parameters

Direct-solid carbonation is primarily influenced by temperature and pressure, where higher pressure and temperature enhance process efficiency by favouring equilibrium shifts to free CO<sub>2</sub> (Chen et al., 2021a). However, several parameters such as temperature, particle size, reaction time, carbon dioxide pressure, pH, and liquid-to-solid ratio (L/S) influence aqueous carbonation (Xiao et al., 2014):

#### Reaction Time

Initially, carbonation is a rapid process, but the rate diminishes over time until reaching equilibrium. This trend is attributed to the high concentration of reactive species (such as CaO and calcium silicates) and the alkaline pH (approximately 11) at the start, facilitating the reaction with CO<sub>2</sub>. Unfortunately, when the time passes, the concentration of free Ca ions decreases, and the pH decreases (around 6.5) making the carbonation process. Additionally, the formation of calcite creates a layer around the dissolved steel slag particles, influencing sequestration (Chen et al., 2021a).

#### Reaction Temperature

In the aqueous carbonation process, both the thermodynamic equilibrium and rate constants, as well as CO<sub>2</sub> dissolution, are influenced by temperature. If it increases, this parameter has a dual effect: it enhances the solubility of cations (such as Mg and Ca) but impacts CO<sub>2</sub> dissolution. It is crucial to emphasize the importance of finding the “optimum temperature,” which is contingent on material characteristics and experimental conditions. Temperatures ranging from 10°C to 40°C promote cation release but decrease the solubility of CO<sub>2</sub>, which is not a limiting step, in this condition. Additionally, low temperatures facilitate CaCO<sub>3</sub> formation on both the solid and liquid slag surfaces due to the high dissolution of CO<sub>2</sub>, whereas at high temperatures, CaCO<sub>3</sub> forms on gas-water interfaces owing to the increased dissolution of Ca ions in the liquid phase (Chiang and Pan, 2017; Chen et al., 2021a; Wang J. et al., 2021; Khudhur et al., 2022). This is in accord with some authors (Ukwattage et al., 2017; Li Z. et al., 2023; Huang et al., 2024) reporting that the “optimum temperature” for steel slag is below 50°C–60°C.

#### Particle Size

The particle size importantly influences the carbonation process: smaller particles provide a greater surface area favouring the reaction. Following carbonation, the specific surface area of steel slag is enhanced, caused by two main factors: enhancement of

slag's surface porosity due to the removal of calcium ions from the solid layer, and irregular layers of calcite formed (Ibrahim et al., 2019).

#### CO<sub>2</sub> Pressure

Generally, when the temperature is constant, the amount of dissolved CO<sub>2</sub> in the liquid is dependent on the carbon dioxide partial pressure, following Henry's law. This occurs when CO<sub>2</sub> solubilization is the rate-limiting step. In such cases, enhancing pressure increases the solubilisation of the mineral and the precipitation of carbonate minerals. However, when the rate-limiting step is calcium extraction, the effect of pressure can be different and less apparent. High pressure may have negative effects, such as unfavourable pH conditions or the rapid formation of calcite, leading to the accelerated presence of a protective layer around the slag, reducing the contact area between the gas and the matrix (Chen et al., 2021a; Khudhur et al., 2022).

#### pH

pH has a variable role in the carbonation process: at lower pH, the carbonation process enhances the leaching of metals (cations), which are the reactants of the reaction. Conversely, in a more alkaline environment (higher pH), the carbonation process favours the precipitation of calcium carbonate (Khudhur et al., 2022).

#### Liquid to Solid (L/S) Ratio

When the L/S ratio is lower than the optimum value, the steel slag is not properly dissolved in water and the interaction of CO<sub>2</sub> and calcium ions decreases. When it is higher, CaO reacts directly with CO<sub>2</sub> forming CaCO<sub>3</sub> and CaO dissolves in water producing Ca(OH)<sub>2</sub> which forms calcite. However, when L/S overcomes a critical value, the excessive water can become a mass transfer barrier, or the concentration of calcium ions and ionic strength are reduced (Chen et al., 2021a; Wang J. et al., 2021).

**Table 3** reports literature results in terms of the CO<sub>2</sub> percentages, as a function of representing the amount of gas sequestration per unit of steel slag, with the corresponding reaction parameters (temperature, carbon dioxide pressure, reaction time, liquid-to-solid ratio (L/S), particle size, carbon dioxide pressure, and pH), found in the literature for direct aqueous carbonation.

Typically, better results are achieved with a particle size distribution between approximately 20–200 μm, and with carbonation durations of around 1 h, especially, when steel slag is ground below 100 μm to enhance their reactivity. Regarding temperature, around 100 °C generally supports carbonation, aligning with industrial waste heat ranges. Moreover, a high liquid-to-solid ratio (L/S > 10) is recommended for the reaction; however, significant improvements diminish beyond L/S > 15. Additionally, increasing CO<sub>2</sub> partial pressure enhances CO<sub>2</sub> sequestration, and this is particularly evident at lower gas concentrations due to enhanced solubility through pressurization (He et al., 2023).



**TABLE 3** | CO<sub>2</sub> percentages (amount of gas sequestration per unit of steel slag), with the corresponding reaction parameters (temperature, carbon dioxide pressure, reaction time, liquid-to-solid ratio (L/S), particle size, carbon dioxide pressure, and pH), found in the literature for direct aqueous carbonation.

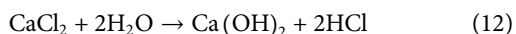
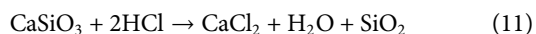
Slag	T	CO <sub>2</sub> partial pressure	Reaction time	[CO <sub>2</sub> ]	Particle size	pH	L/S	CO <sub>2</sub> sequestration	References
Units	[°C]	[Bar]	[hours]	[%]	[µm]		[L/g]	[%]	
SS	20	2.5	0.5	99.9	199	-	0.08	6.14	Liu et al. (2021)
SS	60	1	5	99.9	22.4	-	10	25.7	Tu et al. (2015)
BOF	20	1	96	20	6	-	0.32	15.7	Jiang and Ling (2020)
BOF	40	1	2	99.9	75	-	3.3	16.4	Ma et al. (2021)
BOF	50	2	4	40	43.11	-	5	46	Polettini et al. (2016a)
BOF	90	1	240	99.9	31	-	30	24.5	Yadav and Mehra (2017a)
BOF	100	2	4	40	43.1	12	5	53	Polettini et al. (2016b)
BOF	65	1	0.5	99	62	-	20	30.3	Chang et al. (2012)
BOF	70	1	2	99.9	53	12.5–11.9	10	22.1	Pan et al. (2016a)
BOF	100	10	24	99.9	50.2	-	5	40.3	Baciocchi et al. (2015b)
EAF	100	10	24	99.9	108.1	-	5	28.0	Baciocchi et al. (2015b)
EAF	20	2.5	0.5	18.2	24	11	10	6.4	Ben Ghacham et al. (2016)
EAF	25	5	3	99.9	63	-	5	59.5	Omale et al. (2019)
EAF	20	0.15	40	15	72	11.7	10	1.7	Bonenfant et al. (2008)
LF	20	0.15	40	15	72	12.5	10	24.7	Bonenfant et al. (2008)
LF	20	1	2	99	88	-	5	38	Yi et al. (2021)
LF	175	50	4	-	<63	-	-	30	Matus et al. (2021)
BF	50	30	48	99.9	10	-	1	1.4	Ukwattage et al. (2017)
BF	150	30	24	-	<75	-	10	28	Ren et al. (2020)
AOD	100	2	1.5	-	<74	-	8	66	Tao et al. (2021)
AOD	60	2	1.5	-	<74	-	8	50	Tao et al. (2021)
AOD	90	6	1	99.5	39.3	-	16	44.6	Santos et al. (2013)

## Indirect Carbonation

Unlike direct carbonation, where CO<sub>2</sub> directly interacts with the solid material, indirect carbonation involves a two-step process. Initially, reactive species, such as Ca and Mg ions are pulled out from minerals using solvents, and then they are transformed into respective oxides or hydroxides. Subsequently, these compounds react with carbon dioxide to produce carbonated species. While indirect carbonation offers advantages such as the purity and economic value of the carbonated product and operates under milder reaction conditions (atmospheric pressure and lower temperature), its drawback lies in the use of additional chemical solvents, leading to increased economic costs and necessitating efficient and low-cost recovery methods, which can impact industrial scale-up (Chen et al., 2021a; Yadav and Mehra, 2021; Rahmanihezaki and Hemmati, 2022).

### Indirect Gas-Solid Carbonation

In general, the reaction between CO<sub>2</sub> and Ca and Mg oxides and hydroxides is faster than the direct pathway of respective silicates. Unfortunately, oxides and hydroxides are not present in steel slag. For this reason, their production from silicates is necessary and this is the basis of indirect carbonation. By adding solvent (as HCl) the reactive species are formed (Huijgen et al., 2005; Yadav and Mehra, 2021) as shown in the Eqs 11–13:

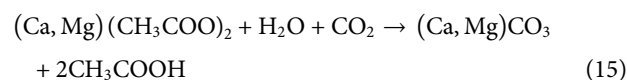
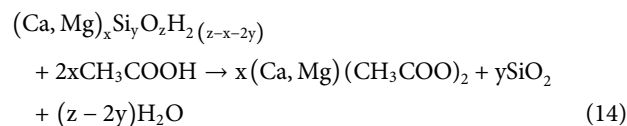


### Indirect Aqueous Carbonation

This procedure involves two successive phases: the extraction of metal ions followed by carbonation in aqueous environments.

The advantage is the optimization of each step. The extraction of ions is optimized using different solvents (acids or bases):

- Strong acids: they are better, in comparison to weak acids, at extracting metal elements from steel slags. Under suitable conditions, the yield of magnesium and calcium solubilisation can result in 46% and 9% respectively (Chen et al., 2021a; Yadav and Mehra, 2021).
- Acetic acid: the limited corrosive properties of this solvent render carbonation a more viable option compared to using strong acids. However, pH of the leaching solution can be adjusted before initiating the carbonation reaction. The acetic acid can be recovered after the extraction process and SiO<sub>2</sub> can be separated by a thickener as shown in the following reactions 14, 15 (Chen et al., 2021a; Yadav and Mehra, 2021):

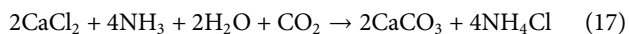
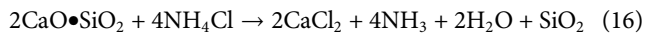


At 1 bar pressure and 25°C of temperature, the conversion is 40% and it can be improved to 75% increasing pressure until 30 bar.

- Ammonium salts: the use of acidic solvents requires a pH adjustment of the solution before the carbonation reaction by the addition of alkaline substances. The crucial aspect of indirect carbonation lies in recuperating



chemical solvents with minimal energy consumption, thereby enhancing the process's economic viability. For this reason, Satoshi et al. (Wang and Maroto-Valer, 2011) evaluated the "pH-swing process" which is based on  $\text{NH}_4\text{Cl}$  use, and extraction and carbonation steps to obtain alkaline and acid conditions without any other chemicals.  $\text{NH}_4\text{Cl}$  exhibits greater selectivity but reduced leachability for the extraction of Ca and Mg ions. In BOF slag the calcium silicate is the more reactive species, and it reacts with  $\text{NH}_4\text{Cl}$  as shown in the following reactions 16, 17 (Chen et al., 2021a; Yadav and Mehra, 2021):



## Influence of Reaction Parameters

### *Solvents Typology and Concentration*

In the indirect carbonation process, the solvent plays a key role in the process. The nature (base or acidic) of the solvent influences the solubility of the metals as follows: strong acids (such as  $\text{H}_2\text{SO}_4$ ;  $\text{HCl}$ , and  $\text{HNO}_3$ ) > acetate (such as  $\text{CH}_3\text{COOH}$  and  $\text{HCOOH}$ ) > ammonium salts ( $\text{NH}_4\text{Cl}$  and  $\text{NH}_4\text{NO}_3$ ) > alkali ( $\text{NaOH}$  and  $\text{KOH}$ ). In fact, a lower pH increases the solubility of metal ions whereas a higher pH favours the precipitation of calcite (Chang et al., 2011).

### *Reaction Temperature*

The effect of temperature is opposing: some studies show temperature has a kinetic effect, while others report that increasing temperature enhances the yield of magnesium and calcium extraction. As for direct carbonation, there exists an optimum value of temperature, under it the speed of the reaction is limited but when it is higher the  $\text{CO}_2$  solubility decreases (Chen et al., 2021a).

### *Particle Size*

Particle size has an important role in the metal solubility in steel slag. In fact, the small particle size favours the solubilization of Mg and Ca ions due to their higher surface area (Chen et al., 2021a).

### *Liquid to Solid (L/S) Ratio*

The L/S ratio mostly influences the extraction step. At higher values, the extraction rate of Mg and Ca ions increases, and it becomes constant when this parameter increases. On the contrary, reducing the liquid-to-solid (L/S) ratio may decrease the reaction rate, but the increase of solvent concentration can solve this effect (Chen et al., 2021a).

## KINETICS MODELS

Generally, there are models used to depict the carbonation reaction, namely, the surface coverage model and the shrinking core model as shown in **Figure 4** (Pan et al., 2014; Li et al., 2018; Chen et al., 2020b).

The coverage model proposes that the reaction rate is directly linked to the portion of the surface covered by the reactant. As the reaction progresses over time, the active surface area remains uncovered by reaction products until reaching maximum conversion.

The phase containing calcium reacts with  $\text{CO}_2$ , forming calcium carbonate that covers the active surface of basic oxygen furnaces, leading to surface coverage.

The model shows how ions compete when they dissolve or form solids, how the area that can react changes over time, and how the reactions on the surface affect the movement of matter between the liquid and the solid (Ragipani et al., 2021; Wang et al., 2023a). Additionally, it accounts for the influence of variability in size distribution and diverse morphology in shaping the changes in the active surface area over time.

On the other hand, the shrinking core model has been employed to determine the step that predominantly controls the rate of particle reactions. (Castellote et al., 2008). The primary premise of this model is that the reaction initiates on the outer surface of the solid material, advances through a narrow front, and eventually permeates the entire solid, forming a fully reacted product known as the "ash" layer.

Subsequently, the rate of the reaction is controlled by either the chemical reaction transpiring at the interface between the reacted and unreacted sections or the diffusion of reactants through the product layer.

Within the shrinking core model, an initially unreacted core progressively diminishes in size as the reaction proceeds (Tu et al., 2015). Furthermore, the creation of small calcite crystals on the surface of the solid forms a protective layer that envelops the reactive particles. This layer serves to shield the particles from further reactions (Wei et al., 2021). Initially, the chemical reaction governs the process, particularly in the initial phase. However, as the reaction progresses, it transitions into diffusion through the porous layer, becoming the rate-limiting step. Carbonate conversion occurs at a notably slower rate compared to leaching conversion and is found to be constrained by diffusion through the calcium carbonate layer that forms. The apparent activation energy for the aqueous carbonation of steel slag is determined to be  $4.8 \text{ kJ mol}^{-1}$ . This indicates the amount of energy required to initiate the reaction (Tu et al., 2015).

Moreover, both models are general models, then the actual kinetics may vary depending on the specific conditions and composition of the steel slag (Tu et al., 2015; Ragipani et al., 2021; Wang et al., 2023a).

## METHODOLOGIES TO IMPROVE THE MINERAL CARBONATION EFFICIENCY

The main methodologies to improve the efficiency of mineral carbonation are the use of additives (chelating agents,  $\text{HCl}$ ,  $\text{CH}_3\text{COOH}$ , and  $\text{HCOOH}$ ) and altering conditions such as change of solid-to-liquid ratio, carbon dioxide concentration, pressure, temperature, and particle size (Wu et al., 2017; Yadav and Mehra, 2021). These factors can accelerate the kinetic of mineral dissolution, which is the lowest step, favouring the silicate dissolution.

## Chelating Agents

These agents are molecules capable of establishing multiple bonds with a single metal ion, effectively capturing the ion, and maintaining it in solution (Chen et al., 2023).

They can augment the solubilization of metal ions from the raw materials, thereby improving the efficiency of the carbonation process. Once the metal ions are leached into the solution, they can engage in a reaction with the dissolved CO<sub>2</sub>, resulting in the formation of solid carbonates.

Frequently used chelating agents encompass ethylenediaminetetraacetic acid (EDTA) and its derivatives, etidronic acid, galactaric acid, sodium metasilicate, as well as phosphate derivatives (Chen et al., 2023). As an example, in a study focusing on indirect carbonation, three chelating agents (adipate salts, malonate, and citrate) were used as solvents under ambient temperature and atmospheric pressure conditions. The study found that the concentration of the solvent was the crucial factor affecting calcium extraction from cement kiln dust (CKD), an alkaline industrial by-product (Castellote et al., 2008).

A research study titled “Investigation into the Accelerated Carbonation Behavior of Steel Slag Used as Cementitious Materials with EDTA Effects” delves into the accelerated carbonation process of steel slag in the presence of EDTA (Chen et al., 2023).

The findings indicate that the carbonation process efficiently decreases the free calcium oxide content in steel slag, leading to a significant enhancement in the volume stability of carbonated steel slag (CSS)-based cementitious materials. The presence of EDTA accelerates calcium leaching and acts as a catalyst for the carbonation reaction, leveraging its chelating effect. Steel slag treated with EDTA achieves a CO<sub>2</sub> sequestration rate exceeding 14% within a 60-min carbonation period.

However, it's fundamental to underline that the inclusion of chelating agents may introduce complexity to the process, as there might be a need for the recovery and reuse of these agents to ensure economic viability (Castellote et al., 2008).

## Pre-treatment Process

Enhancing the carbonation process is achievable also through the pretreatment of slags (Quaghebeur et al., 2015). Improving CO<sub>2</sub> absorption can be achieved by milling or grinding steel slags into smaller particle sizes, as this increases their specific surface area (Wang et al., 2023a). Literature shows that the carbonation of the finest particles results in a nearly complete conversion of free lime to carbonate (or portlandite) under the given reaction conditions. In contrast, larger particles consistently retain free lime, possibly due to carbonate mineral precipitation on the free lime particles, creating a layer of a carbonate phase that hinders fluid from reaching the free lime unreacted core of the particle (Quaghebeur et al., 2015).

Other studies revealed that decreasing the slag particle size and enhancing the porosity of the original slag can notably elevate the carbonation efficiency (Polettini et al., 2016a).

Carbonation in an autoclave, also increasing the temperature, can represent an alternative method to enhance the steel slag carbonation (Quaghebeur et al., 2015). Furthermore, elevating the partial pressure of CO<sub>2</sub> not only facilitates CO<sub>2</sub> dissolution in

water but also lowers the pH value, representing a not favourable condition for carbonate precipitation.

Finally, the repetitive effectiveness of the continuous wet carbonation process is also possible, to improve the efficiency of the process (Chen et al., 2023). The research findings reveal that despite a decline in CO<sub>2</sub> sequestration efficiency with each cycle, it consistently outperformed the control group.

## CARBONATED SLAG APPLICATION IN THE CEMENT INDUSTRY

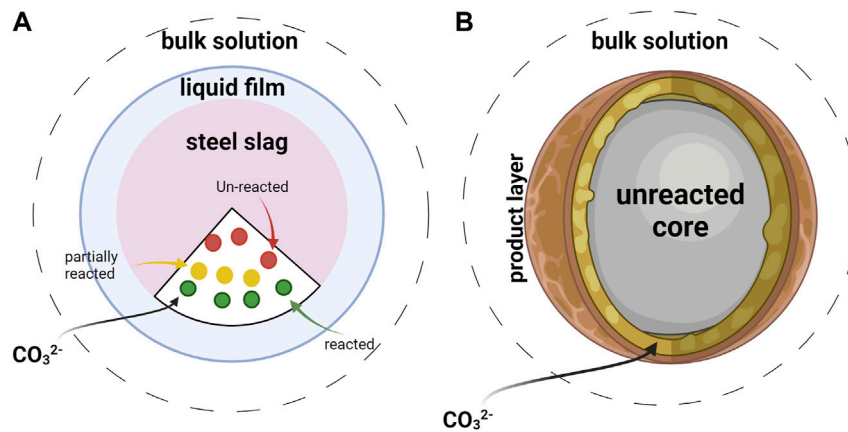
The cement industry is one of the major contributors to the environmental impact of energy consumption and emission of GHGs, including carbon dioxide (Mei et al., 2022). This industrial sector contributes 7.4% of global CO<sub>2</sub> emissions since production of 1 kg of cement releases 0.5–0.7 kg of CO<sub>2</sub> into the atmosphere (Kim et al., 2022; Younsi, 2022). Since cement is an essential component of concrete, its production is responsible for at least 70% of GHGs from the manufacture of concrete. To reduce the CO<sub>2</sub> emissions from concrete products, cement industries have implemented many solutions such as using supplementary cementitious materials (SCMs) as partial replacement for clinker, the main component of Portland cement, or cement during concrete production. Supplementary cementitious materials can be different by-products such as fly ash, steel slag and cement wastes (Younsi, 2022).

Steel slag has been proposed to be employed for road filler production. Nevertheless, the inclusion of f-CaO and f-MgO in steel slag and the low content of hydraulic components, such as C<sub>2</sub>S and C<sub>3</sub>S, can induce delayed expansion and cracking in cement-based materials, leading to prolonged volume instability (Li et al., 2022). Moreover, recent studies have indicated significant alterations in the mechanical properties of steel slag and volume instability and a decrease of heavy metals leaching, following carbonation treatment, making it a possible supplementary cementing material or aggregate in concrete and cement applications (Mo et al., 2017; Dong et al., 2021). The accelerated carbonation eliminates the expansive compounds like f-CaO/MgO in fresh slag enabling the potential of these by-products as building materials (Li and Wu, 2022; Wang et al., 2023).

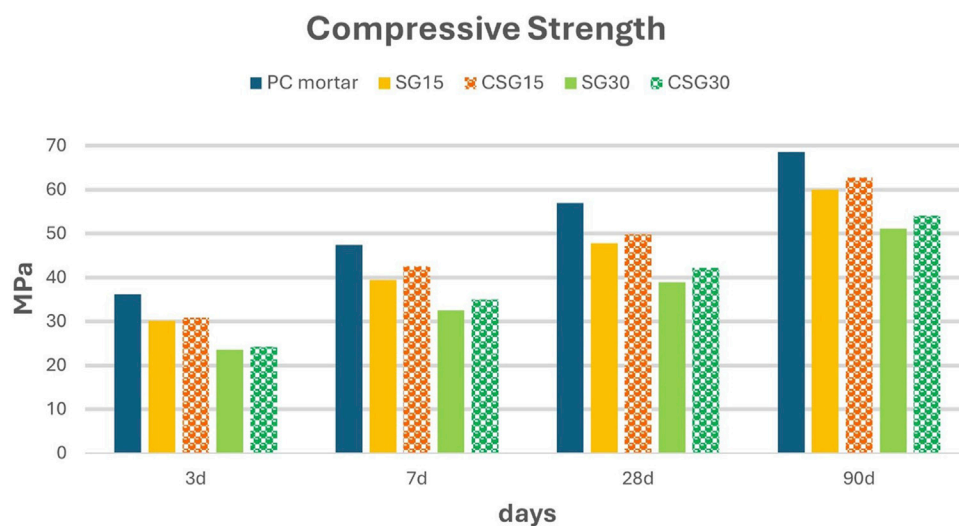
Some opportunities for reuse in building applications are reported in the following.

## Supplementary Cementitious Materials

One possible use of carbonated slag is as a partial replacement of Portland cement. The first results in existing studies are encouraging but the research activities are still ongoing. Chen et al. (2016) demonstrated that the mechanical compressive strength of cement with 10% carbonated BOF slag has higher values than uncarbonated slag in a short time. Moreover, the carbonated BOFs have a better hydration activity than the starting material at 3 days and 28 days increased by 97% and 16% respectively (Wang et al., 2023b). Mechanical properties increase due to the activity of limestone and hydrated products. CaCO<sub>3</sub> densify the microstructure of steel slag, reduces the pores



**FIGURE 4** | Kinetics models of carbonation reaction: **(A)** The surface coverage model shows that the reaction takes place exclusively at unreacted surface sites. In this model, the phase containing calcium reacts with  $\text{CO}_2$ , forming calcium carbonate that covers the active surface of basic oxygen furnaces, leading to surface coverage. As the reaction progresses the active surface area remains uncovered by reaction products until reaching maximum conversion. **(B)** The shrinking core model shows that an initially unreacted core progressively diminishes in size as the reaction proceeds. Furthermore, the creation of small calcite crystals on the surface of the solid forms a protective layer that envelops the reactive particles. This layer serves to shield the particles from further reactions.



**FIGURE 5** | Compressive strength values of mixes blended uncarbonated and carbonated steel slag (Liu et al., 2021). Mortar mixes with respectively 15% and 30% of carbonated slag (CSG15 and CSG30) have higher compressive strength compared to mixes with uncarbonated slag. Otherwise, it is important to underline that the introduction of this by-product decreases the mechanical properties compared to PC mortar due to the presence of non-hydraulic phases as  $\gamma\text{-C}_2\text{S}$ , Fe-containing solid solutions,  $\text{C}_2\text{F}$ ,  $\text{C}_{12}\text{A}_7$ .

size and increases hydration activity providing a better surface for the growth and nucleation of hydrated products (Zajac et al., 2020; Song et al., 2021). Moreover, in the cement matrix, calcite can react with  $\text{C}_3\text{A}$  to form  $\text{C}_3\text{A}\bullet\text{CaCO}_3\bullet x\text{H}_2\text{O}$  which shows better stability and hardness compared with sulphoaluminate, present in the cement (Pan et al., 2016b; Song et al., 2021; Wang et al., 2023b). Another improvement of carbonated steel slag is the reduction free species (CaO and MgO) and consequently the increase in their stability (Song et al., 2021). Peng et al. (Liu et al., 2021) show the positive effects of carbonation on increasing compressive strength. As **Figure 5** shows, mortar mixes with

respectively 15% and 30% of carbonated slag (CSG15 and CSG30) have higher compressive strength compared to mixes with uncarbonated slag. Otherwise, it is important to underline that the introduction of this by-product decreases the mechanical properties compared to PC mortar due to the presence of non-hydraulic phases such as  $\gamma\text{-C}_2\text{S}$ , Fe-containing solid solutions,  $\text{C}_2\text{F}$ ,  $\text{C}_{12}\text{A}_7$  (Liu et al., 2021). The compressive strength decreases when the steel slag percentage is enhanced.

Srivastava et al. show the role of carbonation conditions on compressive strength, using 30% of carbonated BOF slag. Varying

the CO<sub>2</sub> pressure, the calcite content and consequently the mechanical performance change. The sample carbonated at 1 bar (less calcite formed) reaches a double value compared to the mix with slag carbonated at 5 bar (high calcite content) (Srivastava et al., 2023).

Also, particle size can influence the compressive strength of the mix. Bodor et al. show that mix with the finest (<0.08 mm) and the coarsest (<1.6 mm) untreated and carbonated BOFs have lower compressive strength compared to Portland Clinker (Bodor et al., 2013). Otherwise, using a ternary system, as Zhang et al. (2011) proposed, can produce high-performance cement by using a huge quantity of SCMs. This system is a combination of fine and coarse slag to have a discontinuous particle size distribution (Pan et al., 2016b).

## Aggregates

Another use of carbonated steel slag is as a sand substitute for cementitious building materials (Zhang et al., 2011; Wang et al., 2023b). Mo et al. (2020) use carbonated slag as artificial aggregates in a mix with Portland cement and fly ash. The concrete prepared with carbonated slag exhibits high compressive strengths in the range of 36.2–45.5 MPa at 28 days. If this mix is compared to natural limestone aggregates, the internal humidity is enhanced and autogenous shrinkage of concrete decreases. Mo et al. (2017) show the positive influence of the CO<sub>2</sub> curing process in concrete samples with steel slag as coarse aggregates. Concrete containing steel slag shows better mechanical properties (like higher compressive strength) than natural aggregate concrete due to the higher content of mineralogical phases that could react with CO<sub>2</sub>.

In conclusion, using carbonated steel slag in the cement takes some advantages such as 1) increase in volume stability, 2) reduction of heavy metal leaching, 3) improvement of mechanical properties and 4) decrease of corrosion due to sulfates, salts, and acid attacks. Otherwise, the use of uncarbonated slag reduces the compressive and tensile strengths and durability and favors a volume expansion (Li and Wu, 2022).

Furthermore, the carbonation process applied to steel slag seems to involve additional energy consumption, contributing to CO<sub>2</sub> emissions either directly or indirectly (Wang et al., 2023a). As a result, the widespread industrial adoption of carbonation technology for steel slag remains considerably constrained. Future research efforts should concentrate on developing more efficient.

## Soil Stabilization

Historically, several amounts of steel slag have been stockpiled in open-air environments, posing significant risks to safety and environmental integrity, including concerns such as land occupation and the leaching of heavy metal constituents into the surrounding soil (Gao et al., 2023).

Moreover, the possibility of incorporating steel slag into the soil is considered in the literature and presents a multifaceted approach to enhancing soil properties and environmental sustainability. Even with the limited amount of available work, some considerations can be proposed.

Basically, the addition of steel slag to soil is based on the idea of providing the joint application of industrial solid waste and mine voids for the enduring subterranean sequestration of CO<sub>2</sub> as minerals. This method not only ensures the resourceful recycling of CO<sub>2</sub> and the optimal use of subterranean voids but also aids in averting geologic calamities, enhancing the economic viability of the system, and holds great potential for broad-scale implementation (Kang et al., 2024). Enhanced soil aggregate stability is achieved through the binding action of CaCO<sub>3</sub>, particularly evident with steel slag incorporation, thereby improving soil resistance to erosion and enhancing overall structural integrity.

Steel slag modifies the soil's geotechnical characteristics, leading to a reduction in liquid and plastic limits, thereby improving workability and structural integrity. Moreover, the addition of steel slag increases the soil's specific gravity, indicating densification and enhanced particle packing, consequently bolstering soil strength and stability. Additionally, steel slag contributes to carbon sequestration by reacting with CO<sub>2</sub> to form stable carbonates, with CaCO<sub>3</sub> acting as a catalyst, expediting the process, and mitigating greenhouse gas emissions (Rowley et al., 2018). The alkaline nature of steel slag also aids in neutralizing acidic soils (Capobianco et al., 2014), promoting optimal pH levels essential for nutrient availability and plant growth, while CaCO<sub>3</sub> helps maintain this balance. Furthermore, the immobilization of heavy metals present in steel slag is facilitated by CaCO<sub>3</sub>, reducing their mobility and potential environmental impact by forming stable complexes within the soil matrix (Cristina Fernandes Deus et al., 2019).

In summary, the synergistic effects of steel slag carbonation with soil must be studied in more detail, to better understand their pivotal role in soil stabilization, carbon sequestration, pH regulation, nutrient availability, heavy metal containment, and aggregate stability, underscoring the importance of their comprehensive utilization in environmental applications.

**Table 4** summarizes the mechanical properties of cement that are impacted by carbonated steel slag addition.

## ADVANTAGES AND LIMITATIONS OF STEEL SLAG CARBONATION

Leveraging steel slag CO<sub>2</sub> sequestration can provide economic, environmental, and social advantages:

1. Economical carbon capture: the steel slag reuse obviates the necessity of establishing a new carbon capture facility, thereby reducing the costs of CO<sub>2</sub> sequestration (Sarperi et al., 2014).
2. Waste valorization: carbonation provides a means of valorizing an industrial waste product, contributing to a circular economy, and promoting sustainable practices.
3. Carbon-negative potential: through the carbonation process, waste can be used as a carbon sink, effectively sequestering and immobilizing CO<sub>2</sub>. This carbon-negative aspect can contribute to broader climate change mitigation efforts.



**TABLE 4** | The impact of carbonated steel slag on the mechanical properties of cement.

Property	Improvement with carbonated steel slag	References
CO <sub>2</sub> Uptake	Increased CO <sub>2</sub> sequestration capacity	Zeng et al. (2023)
Compressive Strength	Enhanced early and long-term strength	Zhong et al. (2023)
Durability	Improved resistance to environmental factors	Zhang et al. (2022)
Setting Time	Adjusted setting times for optimal performance	Chen et al. (2021c)
Flexural Strength	Improved flexural strength for structural applications	Hou et al. (2018)

- Technologically feasible and energy-efficient: this procedure is recognized to be relatively simple, cost-effective, and technologically feasible.
- Adaptability to existing infrastructure: integrating the process with steel slag is adaptable to existing steel mill infrastructure, minimizing the need for significant modifications or new construction.
- Improved leaching properties: the process of carbonation improves the stability of heavy metals contained in steel slag, reducing concerns related to environmental impact (Boone et al., 2014).
- Reduced raw material needs: by steel slag carbonation, there is a consequential reduction in the demand for fresh raw materials, contributing to overall sustainability and resource efficiency.
- Recovering value-added by-products: products derived from carbonation can be reused and generate revenues.
- Versatile applications of the obtained secondary materials: the carbonation process improves the properties of steel slag, making it more appropriate for use as aggregates in various construction applications, thus contributing to sustainable building practices.
- Reduced environmental footprint: the on-site utilization of secondary materials derived from steel slag after carbon dioxide sequestration reduces the need for transporting materials over long distances, minimizing the environmental footprint associated with transportation.
- Community and stakeholder acceptance: this technology may encounter greater community and stakeholder acceptance compared to establishing new carbon capture facilities.

Although promising advantages, some challenges necessitate careful consideration and innovative solutions:

- Chemical heterogeneity: the chemical composition of steel slags is inherently diverse, introducing significant complexities in accurately modelling the dissolution kinetics. This heterogeneity poses a challenge to researchers seeking to understand and predict the behaviour of steel slag during carbonation, demanding sophisticated modelling approaches (Ragipani et al., 2021).

- Slow carbonation kinetics: the carbonation kinetics, especially under atmospheric conditions, is a notable hurdle that ongoing research endeavours are aiming to overcome. Improving the speed of the carbonation reaction is crucial for enhancing the overall efficiency of the process and its feasibility on an industrial scale.
- Incomplete understanding of rate-limiting steps: the identification and comprehension of rate-limiting steps, coupled with a nuanced understanding of the role played by product layers in reaction kinetics, are aspects still in the process of development (Ragipani et al., 2021).
- Increased process costs: while indirect carbonation shows a promising leaching ratio of alkaline earth ions and a high carbonation conversion rate, the requirement for adding chemical reagents to accelerate the reaction can lead to increased process expenses (Luo and He, 2021).
- Additional processes: the carbonation of natural minerals, including steel slag, involves supplementary processes such as mining, transportation, and grinding of raw metals. These additional steps can contribute to heightened overall costs and energy consumption (Wang et al., 2023a).

Addressing these challenges is critical for establishing steel slag carbonation as a viable and widely adopted industrial process.

To apply the carbonation process at an industrial scale, the sustainability and possible impacts of this technology must be studied focusing on the process's energies. To quantify the impact a life cycle assessment is the instrument. However, only limited studies can be found (Suer et al., 2022; Watjanatepin et al., 2023) and generally, they are referred to as preliminary results.

## CONCLUSION AND PERSPECTIVES

The mineral carbonation process mimics and accelerates natural weathering, where rocks break down over an extended period due to weather conditions, including the reaction of CO<sub>2</sub> with specific rocks to form carbonates. However, the natural process is too slow to significantly impact atmospheric CO<sub>2</sub> levels in a human timescale. Additionally, the abundance of magnesium and calcium silicate deposits in the Earth's crust makes mineral carbonation a potentially scalable solution for CO<sub>2</sub> sequestration.

This study provides insights into the steel slag carbonation process, highlighting its advantages and challenges. While it emphasizes the potential for both CO<sub>2</sub> sequestration and waste valorisation, it also underscores the need for additional research to address challenges and make the technology economically feasible.

Direct gas-solid carbonation is a simple process where CO<sub>2</sub> gas reacts directly with solid metal oxides to produce carbonates. The specific reactions vary depending on the feedstock.

In contrast, indirect carbonation offers benefits in terms of the superior purity of the resulting product. However, it is a more intricate process that may demand more energy due to the supplementary leaching step. Additionally, the challenges of recycling extractants contribute to increase costs in the carbon mineralization process.

Various approaches to accelerate carbonation, such as employing high pressure and/or high temperature, increased stirring speeds, and additives, have been explored. However, these techniques may lead to higher energy consumption and could affect the carbonation reaction. Hence, optimizing the steel slag carbonation process with minimal heat and power usage is essential but presents a significant challenge for direct carbonization.

While research on the reuse and recycle of steel slag after carbonation is not diffuse, the fundamental chemistry of mineral carbonation is well understood. Challenges, including the requirement for substantial starting material, slow reaction rates of natural minerals, and energy demands, need to be addressed.

Prospective research efforts could centre on enhancing the carbonation process for steel slag and optimizing parameters for scaling up applications.

Another big challenge of this topic is the reuse of carbonated steel slag in the cement industry. As different studies show, the introduction of steel slag as SCMs reduces the compressive strength of the cement and this reduction is proportional to the percentage of replacement (Zhang et al., 2011; Srivastava et al., 2023). Otherwise, the carbonation treatment improves the hydraulic properties of steel slag making them suitable for this use. It is mandatory to study deeply the mechanism of hydration of the slag in the matrix cement focusing on the possible leaching of heavy metals and the compressive strength.

In conclusion, mineral carbonation holds promise for mitigating climate change through CO<sub>2</sub> sequestration and adding an extra value to steel slag. Despite existing challenges,

the potential benefits make it a compelling area for further research, and it is a way to favour a circular process.

## AUTHOR CONTRIBUTIONS

EB and GB realized bibliographic analysis and performed draft manuscript preparation. LD contributed to the manuscript review and improvement. All authors contributed to the article and approved the submitted version.

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## CONFLICT OF INTEREST

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