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Research Article

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A Mini Review of Direct Aqueous Carbonation of Calcium Carbide Residue and Ferrous Slag for Carbon Mineralization and Cementitious Applications

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Abstract

Direct aqueous carbonation of alkaline industrial residues has emerged as a promising pathway for CO₂ sequestration and waste valorization; however, its effectiveness is strongly constrained by material-specific reaction mechanisms. Carbonation of calcium carbide residue (CCR) alone is limited by rapid surface passivation caused by localized CaCO₃ precipitation, while iron- and steel-making slags often exhibit sluggish carbonation due to kinetically inert silicate matrices and restricted calcium availability. Recent studies demonstrate that combining CCR with slag fundamentally alters the carbonation pathway by redistributing carbonate precipitation away from CCR particle surfaces, mitigating passivation, and enabling sustained calcium dissolution. This mini review synthesizes recent advances in aqueous carbonation of CCR and CCR–slag systems with a mechanistic focus on rate-controlling regimes, precipitation topology, and interfacial processes. Particular attention is given to reconciling contradictory trends in operating parameters and elucidating how carbonation-derived products perform as supplementary cementitious materials (SCMs). Available evidence indicates that carbonated CCR and CCR–slag products primarily function as fillers and hydration nucleation agents, with strength enhancement confined to low replacement levels and fresh-state behavior governed by carbonate morphology and surface characteristics. Durability performance remains largely unverified, representing a critical barrier to practical deployment. By integrating mechanistic insights with performance considerations, this review identifies key design principles and research priorities necessary to advance carbonation-based valorization of CCR and related industrial residues.

Keywords: Calcium carbide residue, Direct aqueous carbonation, CCR-slag synergy, Supplementary cementitious materials, CO₂ sequestration

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1. Introduction

The escalating emission of anthropogenic CO₂, a predominant greenhouse gas, continues to drive critical ecological challenges, including global warming, extreme weather patterns, and ocean acidification [1]. The cement industry, as a major contributor to these emissions, underscores the urgent need for sustainable alternatives and effective carbon mitigation strategies [2]. In this context, carbon capture, utilization, and storage (CCUS) technologies, particularly CO₂ mineralization using industrial alkaline wastes, present a promising pathway to concurrently address emissions reduction and waste valorization [3, 4].

A significant source of such alkaline waste is calcium carbide residue (CCR), a by-product from acetylene production with calcium hydroxide (Ca(OH)₂) as its primary component [5]. Global CCR generation exceeds 40 million tons annually, yet its utilization remains below 30%, with the majority subjected to landfilling or stockpiling [4, 6]. This practice not only wastes land resources but also poses environmental risks due to CCR's fine particles and high alkalinity (pH > 12), which can pollute water and air [7]. Parallely, the iron and steel industry generates vast quantities of slag by-products. Steel slag output accounts for 10-15% of crude steel production, while granulated blast furnace slag reaches approximately 390 million tons globally [8, 9, 10]. Conventional disposal via dumping occupies extensive land and poses persistent pollution threats. The construction sector's pressing need to reduce its carbon footprint thus intersects with the challenge of managing these voluminous alkaline wastes.

Direct aqueous carbonation has emerged as a viable, moderate-severity technology to stabilize these wastes while sequestering CO₂. In this process, CO₂ dissolves in an aqueous medium to form carbonic acid, which dissociates into carbonate ions (CO₃²⁻) and protons (H⁺). The protons promote the leaching of calcium ions from the solid matrix, which subsequently precipitate as stable carbonates [11, 12]. The aqueous environment lowers the activation energy for mineralization, enhances reactivity, and achieves higher carbonation rates and efficiency at lower temperatures compared to direct dry routes [6, 13]. CCR, with its high Ca(OH)₂ content, small particle size, and reported CO₂ sequestration capacity of 0.18-0.47 g CO₂/g CCR, is particularly suitable for this process [8, 14].

Despite growing interest in direct aqueous carbonation of alkaline industrial residues, the carbonation of individual waste streams such as calcium carbide residue (CCR) and iron- or steel-making slags remains constrained by inherent material-specific limitations. CCR, while highly reactive due to its portlandite-rich composition, undergoes rapid surface passivation during aqueous carbonation, leading to diffusion-controlled kinetics and incomplete conversion [4]. In contrast, steel and blast furnace slags often exhibit sluggish carbonation behavior arising from low calcium leaching rates and kinetically inert silicate matrices [15]. As a result, carbonation performance in these systems is frequently evaluated in terms of aggregate CO₂ uptake or reaction efficiency [16, 17], with limited consideration of how carbonation pathways, precipitation topology, and interfacial mechanisms govern both process effectiveness and the functionality of the resulting products.

Recent studies indicate that combining CCR with iron- and steel-making slags fundamentally alters the carbonation mechanism by redistributing CaCO₃ precipitation away from CCR particle surfaces and onto slag substrates, thereby mitigating passivation and enabling higher overall conversion. However, these mechanistic insights remain fragmented across individual studies and

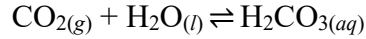
are often obscured by system-specific experimental conditions. This mini review synthesizes recent advances in aqueous carbonation of CCR and CCR-slag systems with a mechanistic focus on rate-controlling regimes, synergistic precipitation behavior, and their implications for the performance of carbonated products as supplementary cementitious materials (SCMs). By reconciling apparent contradictions and identifying unresolved structure–property relationships, this review aims to establish design principles for carbonation-based waste valorization, rather than cataloging isolated experimental outcomes.

2. Fundamentals of Direct Aqueous Carbonation

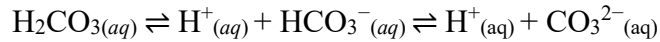
The direct aqueous carbonation process involving calcium-based systems follows well-defined reaction pathways. Initially, gaseous CO_2 dissolves in water, resulting in the formation of carbonic acid (H_2CO_3). This acid then dissociates into bicarbonate (HCO_3^-) and carbonate (CO_3^{2-}) ions [6]. At the same time, calcium ions (Ca^{2+}) are released from the calcium-based solid residue into the solution. Calcium carbonate (CaCO_3) forms when aqueous Ca^{2+} ions react with CO_3^{2-} ions [7].

The direct aqueous carbonation process can be summarized as follows:

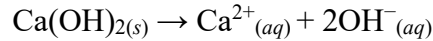
- i. CO_2 Dissolution and Hydration



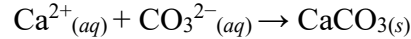
- ii. Acid Dissociation



- iii. Calcium Dissolution



- iv. Carbonate Precipitation



A schematic overview of a typical direct aqueous carbonation configuration is shown in figure 1.

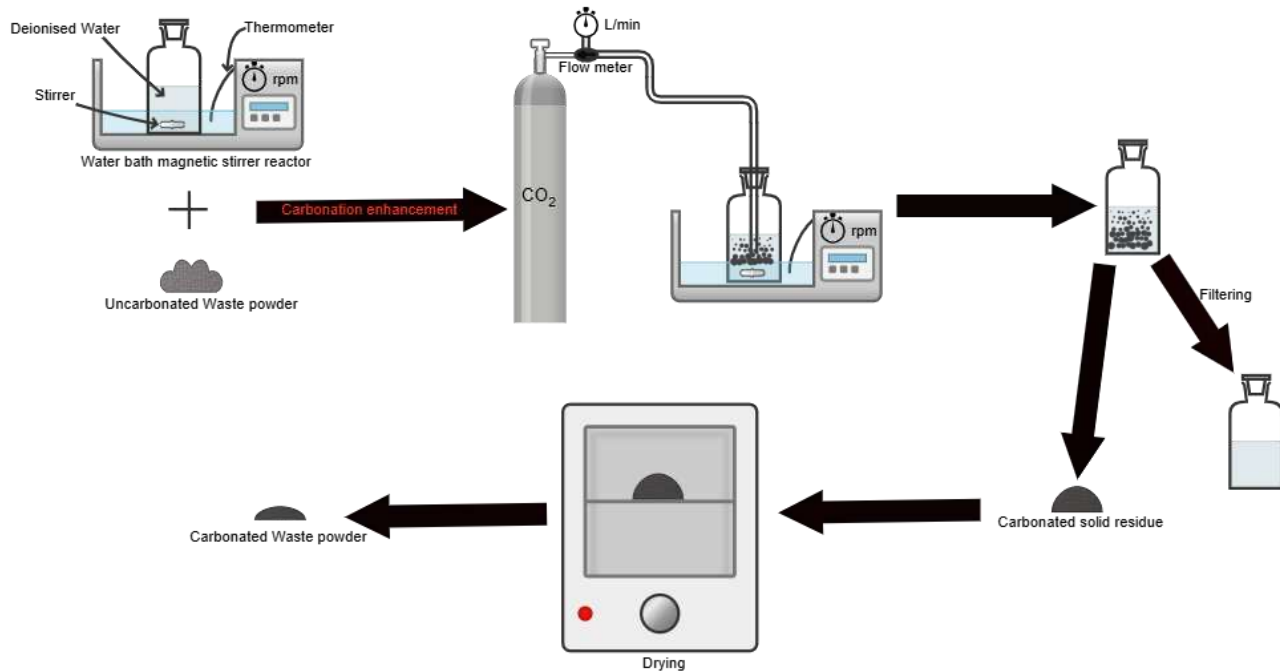


Figure 1. A typical direct aqueous carbonation experimental setup

The setup in figure 1, highlights the gas-liquid-solid interaction central to aqueous mineral carbonation, wherein CO_2 is introduced into an alkaline slurry to promote calcium dissolution, aqueous transport, and subsequent carbonate precipitation.

The process is driven by a decrease in pH as CO_2 dissolves, creating conditions that encourage the dissolution of calcium-based solids and the subsequent release of calcium ions (Ca^{2+}) into the solution. This leads to precipitation of stable carbonate (CaCO_3). The reaction occurs spontaneously until it approaches equilibrium, which is typically signaled by the system's pH stabilizing close to neutral [18, 19, 20] or when the CO_2 concentration in the outlet gas matches that of the inlet [21].

2.1. CCR Aqueous Carbonation Mechanism

The carbonation mechanism for CCR specifically involves a series of interfacial and solution-phase steps. The process begins with the dissolution of portlandite ($\text{Ca}(\text{OH})_2$) from the CCR matrix into the aqueous phase. Parallely, CO_2 dissolution and hydrolysis provide a source of carbonate ions. The nucleation of CaCO_3 occurs once the ion activity product exceeds the solubility product, followed by crystal growth and precipitation [6, 7].

The primary solid product formed is mainly calcite, which is the most thermodynamically stable polymorph of CaCO_3 under standard carbonation conditions [7, 17]. However, the resulting polymorph can be influenced; for example, the incorporation of crystal modifiers like glycine may stabilize vaterite by modifying nucleation energies and adhering to crystal surfaces [19].

Kinetic investigations indicate that the carbonation of CCR starts rapidly but soon encounters limitations. The main kinetic barrier is attributed to “surface passivation,” resulting from the formation of a dense, low-permeability layer of CaCO_3 on the surfaces of CCR particles. This layer significantly restricts the outward diffusion of Ca^{2+} ions from the center and the inward diffusion of CO_2 [17]. Therefore, while the initial phase might be affected by gas-liquid mass transfer [7], overall conversion is often governed by solid-liquid diffusion through this product layer [6]. This represents the dominant mechanistic model for CCR carbonation in isolation. The reaction pathway governing direct aqueous carbonation of CCR, including the development of surface passivation, is schematically illustrated in figure 2.

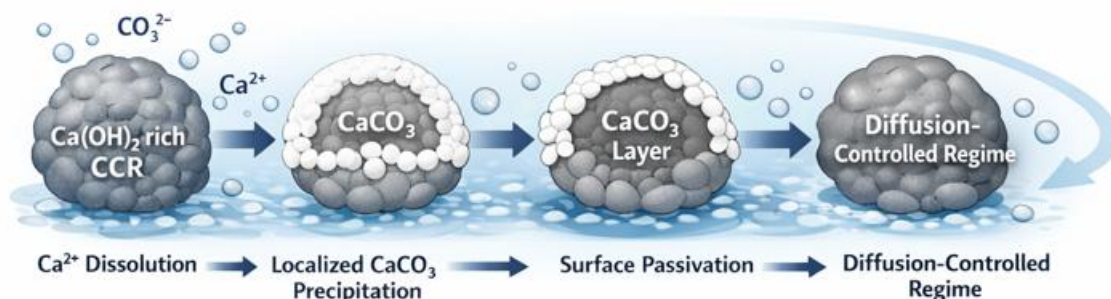


Figure 2. Schematic illustration of the dominant reaction pathway during direct aqueous carbonation of calcium carbide residue (CCR). Rapid calcium dissolution is followed by localized CaCO_3 precipitation on CCR particle surfaces, leading to surface passivation and a transition to diffusion-controlled kinetics that limits overall conversion.

As depicted in Figure 2, localized CaCO_3 precipitation directly on CCR particle surfaces progressively restricts Ca^{2+} outward diffusion and CO_2 ingress, causing a transition from reaction-controlled to diffusion-controlled kinetics.

Analytical findings from SEM and EDS indicate a dense Carbonate layer on particle surfaces, accompanied by a diminished carbonate signal in the core, which validates this passivation phenomenon [17].

Reported driving forces: The efficiency of this carbonation process is influenced by several operational parameters, as synthesized from the reviewed studies.

The reported effects of key operational parameters on CCR aqueous carbonation are summarized in Table 1.

Table 1: Reported Influences of Key Parameters on Direct Aqueous Carbonation

Parameter	Reported Direction of Influence	Key Study/Studies
Temperature	Increased temperature (e.g., 25–60°C) generally enhances the CO_2 sequestration capacity and the dissolution rate of $\text{Ca}(\text{OH})_2$. Excessively high temperatures may reduce efficiency.	[6], [17], [21]
CO_2 Concentration/Pressure	Higher CO_2 concentrations (e.g., up to 15 vol%) or pressures improve CO_2 removal efficiency and sequestration capacity by increasing solubility and driving force.	[6], [21], [22]
Particle Size	Reduced CCR particle size (e.g., <75 μm) increases surface area, leading to higher CO_2 removal efficiency and sequestration capacity.	[6], [21]
Liquid-to-Solid (L/S) Ratio	An optimal ratio exists. Low ratios limit mass transfer; high ratios dilute Ca^{2+} concentration. Typical optima range from 8-10 mL/g to 100 g/L.	[6], [16],[21]
Agitation	Increased mixing improves uniformity and efficiency, though optimal speed may depend on system composition.	[20], [22]
Time	Insufficient time leads to incomplete carbonation, while excessive time is energetically inefficient and may be counterproductive due to passivation. The "optimal" time is system-dependent, varying with CCR characteristics, L/S ratio, CO_2 concentration, and temperature.	[16], [18]

2.2. Contradictions and Inconsistency in CCR Aqueous Carbonation

Despite the consensus on the overall process, discrepancies exist in the literature regarding the carbonation of CCR. These contradictions often stem from differences in experimental conditions and analytical focuses.

Key contradictions reported in the literature and their likely origins are summarized in Table 2

Table 2: Key Contradictions and Their Possible Explanations

Contradiction /Inconsistency	Report from Studies	Observation
Rate-Limiting Step	One study identifies gas-liquid mass transfer as the main limit [7], while others emphasize solid-liquid diffusion inhibited by surface passivation as the key barrier [6, 17].	Due to different experimental stages and conditions. Gas-liquid transfer may dominate initial kinetics, while product-layer diffusion controls the later stage and overall conversion yield.
Effect of Temperature	Most report a positive effect, but one entry notes the effect on CO ₂ removal efficiency for CS was "almost negligible" [21].	Studies lack statistical experimental optimization(ANOVA/Signal-to-noise ratio) to determine the most influential aqueous carbonation factors
Dominant CaCO ₃ Polymorph	Most studies report calcite, but one demonstrates that vaterite can be the dominant product with additives [19].	Not a true contradiction, but highlights a condition-dependent outcome. The "base" system favors calcite, while additives can kinetically stabilize metastable vaterite.

3. Synergistic Carbonation of CCR and Iron/Steel Slags

3.1. Why Synergy Matters

The primary driver for combining calcium carbide residue (CCR) with iron/steel slags in aqueous carbonation is to overcome the intrinsic limitations of each material processed alone. For CCR, the rapid formation of a dense, passivating CaCO₃ layer on particle surfaces severely limits conversion yields. For many slags, the low reactivity and slow leaching of calcium from silicate matrices constrain carbonation kinetics. Synergistic systems aim to create chemical complementarities that bypass these individual barriers [16, 17].

The proposed rationales center on a division of labor and interactive chemistry. CCR, rich in portlandite (Ca(OH)₂), acts as a readily available and highly soluble calcium reservoir. The slags, depending on their type, contribute complementary properties: they provide abundant solid surfaces for heterogeneous nucleation, certain phases can release additional Ca²⁺, Mg²⁺, or Fe²⁺ upon dissolution, and their surface chemistry may interact electrostatically with ions from CCR. For instance, Ma et al. [20] proposed that steel slag particles, with a negative zeta potential, preferentially attract Ca²⁺ ions leached from CCR. Furthermore, the high alkalinity of CCR slurry can act as an activator, promoting the dissolution of less-reactive phases within the slag. This combination transforms the slag from a slow-reacting solid into an active participant and beneficiary in the carbonation process.

The complementary roles of CCR and different slag types reported in synergistic carbonation systems are summarized in Table 3.

Table 3: Reported Chemical Complementarities and Roles in CCR-Slag Synergy

Study	Slag Type	Stated Role of CCR	Stated Role of Slag	Proposed Interactive Effect
[17]	Electric Arc Furnace (EAF) Slag	Primary Ca^{2+} source.	Substrate for heterogeneous nucleation.	Ca^{2+} from CCR migrates to slag surface, inducing carbonate precipitation there and avoiding CCR surface passivation.
[16]	Ground Granulated Blast Furnace Slag (GGBS)	Supplementary calcium source (high soluble Ca).	Primary nucleation sites.	Ectopic CaCO_3 formation on GGBS enhances Ca^{2+} leaching from CCR, boosting sequestration.
[20]	Basic Oxygen Furnace (BOF) Slag	Alkaline activator; primary Ca^{2+} source.	Preferential site for Ca^{2+} adsorption & precipitation.	Electrostatic attraction directs Ca^{2+} to slag surface; CCR alkalinity activates slag dissolution.

3.2. Proposed Mechanisms

Figure 3 provides a conceptual comparison between the dominant carbonation pathway of CCR alone and the modified pathway that emerges in CCR–slag systems. The core synergistic mechanism, consistently highlighted across studies, is the spatial redistribution of carbonate precipitation, often termed "ectopic cover" or "heterogeneous nucleation on slag." This modifies the pathway dominant in CCR-alone carbonation, where passivation occurs [16, 17].

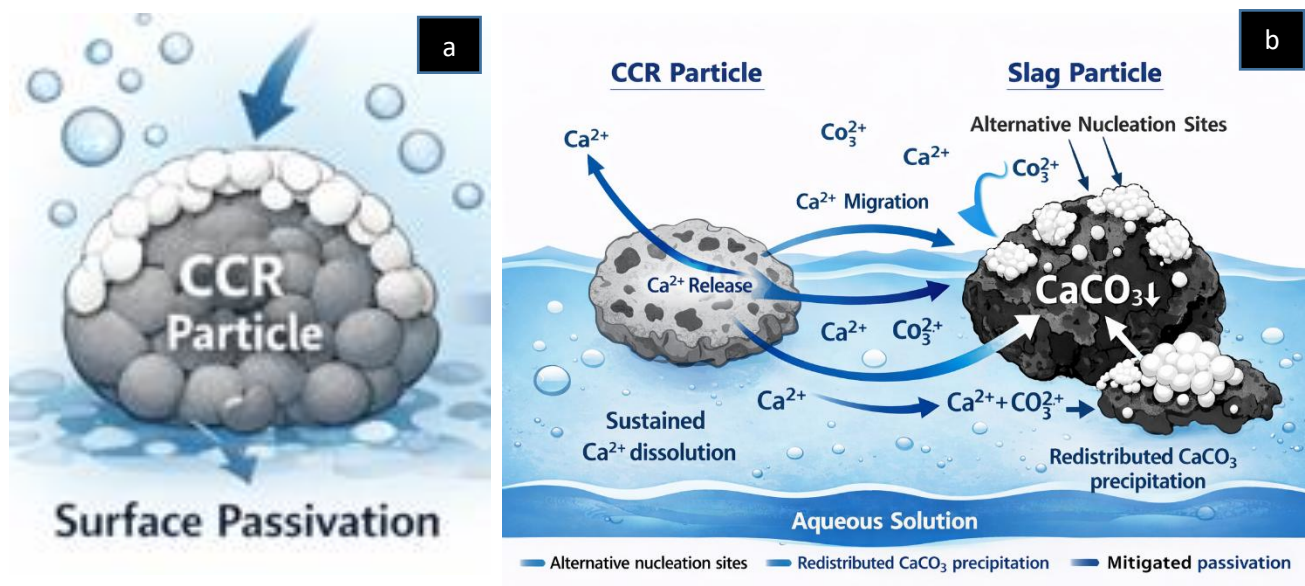


Figure 3. Conceptual comparison of carbonation pathways in (a) CCR-only and (b) CCR-slag systems. In the presence of slag, CaCO_3 precipitation is redistributed away from CCR particle surfaces toward alternative nucleation sites, mitigating passivation and enabling sustained

calcium dissolution and higher overall carbonation conversion.

Reported modifications to the carbonation pathway in CCR-slag systems relative to CCR alone are summarized in Table 4

Table 4: Synergistic Carbonation Mechanisms

Study	Key Modified Pathway vs. CCR Alone	Observed Outcome
[17]	Shift from homogeneous passivation on CCR to heterogeneous nucleation on slag.	No passivation shell on CCR; carbonate forms as fine coating on slag.
[16]	Nucleation and growth of CaCO_3 occurs primarily on GGBS surface.	Higher CO_2 sequestration than theoretical mix; enhanced early-age strength.
[20]	Preferential ("ectopic") growth of CaCO_3 on slag surface.	Avoids coating on CCR particles; increased mineralization rate and efficiency.

In the combined system (Figure 3b), Ca^{2+} ions rapidly released from CCR migrate unidirectionally through the aqueous phase toward slag particle surfaces, where they preferentially react with dissolved CO_3^{2-} to form CaCO_3 . This process is proposed to be facilitated by the slag's surface properties, which may provide favorable nucleation energies [16]. Consequently, a dense carbonate layer forms on the slag particles instead of encapsulating the CCR grains. This keeps the CCR reaction sites open, allowing for more complete consumption of $\text{Ca}(\text{OH})_2$ and significantly improving the overall carbonation rate and CO_2 uptake efficiency. S. Li et al. [17], provided analytical evidence supporting this, noting the absence of a "shell-like structure" on CCR particles in the blend and a more even calcium distribution. Ma et al. [20] further suggested that the electrostatic properties of slag drive this preferential localization.

The spatial redistribution of carbonate precipitation away from CCR particle surfaces mitigates passivation and enables sustained Ca^{2+} dissolution, in sharp contrast to the homogeneous surface passivation observed in CCR-only systems (Figure 3a).

3.3. Contradictions and Comparisons

While all reviewed studies report positive synergy, the described mechanisms and quantified outcomes are not uniform, indicating system-dependent behavior influenced by slag composition and experimental conditions.

A key point of divergence is the primary mechanism for the preferential nucleation on slag. S. Li et al. [17] and Zhang et al. [16] emphasize a physical nucleation site effect. In contrast, Ma et al. [20] introduces a physicochemical mechanism based on electrostatic attraction, suggesting a more active role for the slag's surface charge. Furthermore, the nature of synergy is quantified differently: Zhang et al. [16] uses a synergistic effect ratio (>1), while the others report percentage increases in rate or efficiency. The impact on product morphology is also noted differently; S. Li et al. [17] is the only study to explicitly state that the composite system produces a carbonate coating "more favorable for sustained carbonation" than that formed on slag carbonated alone.

These differences likely stem from the distinct slag types used (EAF slag, GGBS, BOF slag), which possess varying mineralogy, glass content, reactivity, and surface properties. For example, the high glass content and latent hydraulic properties of GGBS [16] may lead to different

interfacial interactions compared to the more crystalline, steel-making derived EAF and BOF slags.

The literature consistently confirms that combining CCR with iron/steel slags creates a synergistic system that enhances aqueous carbonation performance by redirecting carbonate precipitation. However, the specific mechanisms, whether dominated by physical nucleation, electrostatic forces, or alkaline activation, and the magnitude of benefit, are not universal. They appear to be closely tied to the mineralogical and chemical identity of the slag used, underscoring the need to consider slag composition as a critical variable when designing such synergistic processes.

A comparative summary of consistent findings, divergent outcomes, and likely controlling factors across CCR–slag systems is provided in Table 5

Table 5: Comparison of Outcomes and Potential Influencing Factors

Aspect	Consistent Finding	Divergence / Condition-Dependent Outcome	Potential Factor Linked to Divergence
Synergistic Effect	All studies report enhanced carbonation efficiency vs. individual components.	Metric of synergy varies (ratio vs. % increase). Mechanism proposed differs (nucleation site vs. electrostatic).	Study objectives, analytical focus; specific slag surface properties.
Slag's Role	Slag provides a surface for CaCO_3 precipitation, mitigating CCR passivation.	Proposed as passive nucleation site [16] vs. electrostatically active surface [20].	Slag mineralogy and surface charge (zeta potential).
Impact on Products	CaCO_3 forms on slag surfaces (ectopic cover).	Morphology of carbonate on slag may be altered in composite system [17].	Local supersaturation and ion availability influenced by CCR proximity.
System Outcome	Improved CO_2 sequestration.	Degree of improvement and optimal blend ratio are system-specific.	Slag composition (Ca/Mg/Si content, reactivity), particle size, and process conditions (L/S, CO_2 concentration)

4. Reaction Parameters Governing Aqueous Carbonation of CCR and CCR–Slag Systems

The efficiency of direct aqueous carbonation of calcium carbide residue (CCR) and CCR–slag blends is governed by a coupled set of physicochemical parameters that control dissolution kinetics, CO_2 mass transfer, and carbonate precipitation pathways. Rather than acting independently, these parameters define distinct rate-controlling regimes, which explains the non-monotonic trends and apparent contradictions frequently reported in the literature. This section synthesizes the dominant effects of pH, temperature, pressure, CO_2 concentration, liquid-to-solid (L/S) ratio, and particle size, with emphasis on consistent trends and system-specific limitations.

4.1. pH Evolution and Aqueous Carbonate Chemistry

In most reported studies, pH is not actively controlled but instead monitored as an indicator of reaction progress. The aqueous carbonation of CCR systems typically begins at highly alkaline conditions ($\text{pH} > 12$) due to rapid dissolution of $\text{Ca}(\text{OH})_2$ and progressively decreases toward near-neutral values ($\text{pH} \sim 7$) as CO_2 dissolves and hydroxide ions are consumed [18, 19]. Stabilization of pH is commonly interpreted as a proxy for reaction completion.

Despite this consistent qualitative trend, a major limitation across the literature is the lack of quantitative linkage between pH evolution and carbonation rate or conversion yield. pH is widely described as the thermodynamic driving force for Ca^{2+} leaching and carbonate precipitation, yet its role remains largely descriptive rather than mechanistically resolved.

Chemical modifiers can alter this behavior. For example, Zhou et al. [19] demonstrated that glycine delayed pH stabilization and shifted the dominant CaCO_3 polymorph from calcite to vaterite, indicating that additives influence carbonation kinetics and interfacial energetics without changing the thermodynamic endpoint. Overall, while pH evolution is universally reported, its predictive value for process optimization remains underdeveloped.

4.2. Temperature and Pressure Effects

Temperature and pressure exert competing influences on carbonation kinetics and CO_2 solubility, resulting in characteristic optimum operating windows rather than monotonic improvements.

Across CCR-only and CCR–slag systems, most studies report enhanced carbonation rates and CO_2 uptake as temperature increases from ambient to approximately 55–60 °C [17, 20, 21]. This enhancement is attributed to accelerated $\text{Ca}(\text{OH})_2$ dissolution, increased ion diffusion, and faster nucleation kinetics. Beyond this range, performance typically plateaus or declines due to reduced CO_2 solubility in water, which offsets kinetic gains.

Contradictory observations exist. Li et al. [21] reported that temperature had an “almost negligible” effect on CO_2 removal efficiency for CCR alone, highlighting that temperature sensitivity is system-dependent and influenced by reactor configuration, carbonation stage, and performance metric (e.g., removal efficiency versus sequestration capacity).

Pressure generally increases total CO_2 uptake by enhancing gas solubility and driving force. However, elevated pressures can reduce carbonation efficiency by promoting rapid surface precipitation and accelerated passivation of reactive solids [22]. This behavior underscores a recurring theme: conditions that maximize instantaneous reaction rates may simultaneously limit overall conversion by intensifying product-layer diffusion resistance.

4.3. CO_2 Source and Concentration

The rate of mass transfer for CO_2 between gas and liquid phases, as well as the concentration of carbonate ions in the solution, are influenced by the concentration and flow rate of CO_2 . This, in turn, affects the driving force for precipitation. Conditions can range from pure CO_2 [16, 20] to simulated flue gases that contain 10–25% CO_2 [7, 21]. Research often identifies optimal concentrations such as 15 vol% CO_2 for steel slag [21], and optimal flow rate of 9 L/min for a GGBS–CCR blend [16], and 2 L/min for a CCR–EAF slag blend [17]. Concentrations or Flow rates exceeding these optimal levels can lead to reduced efficiency.

Generally, an increase in CO_2 concentration enhances the carbonation driving force and the capacity for sequestration, at least up to a certain limit. Multiple studies have indicated that rather than a consistent improvement, optimal values exist, reflecting a shift from a regime limited by the supply of CO_2 to one constrained by other factors. Li et al. [21] observed that while the

sequestration capacity for CCR alone slightly increased with higher CO₂ concentrations, its removal efficiency actually declined.

Higher CO₂ levels contribute to improved gas-liquid mass transfer and increased supersaturation of the solution [21]. However, excessive CO₂ influx can lead to rapid surface passivation, a phenomenon also noted with increased pressure. Zhang et al. [16] specifically pointed out that performance declines observed at elevated flow rates (12 L/min) were due to a side reaction: $\text{CaCO}_3 + \text{CO}_2 + \text{H}_2\text{O} \rightleftharpoons \text{Ca}(\text{HCO}_3)_2$, which results in the redissolution of carbonate and wasted CO₂.

4.4. Liquid-to-Solid (L/S) Ratio

The liquid-to-solid (L/S) ratio is crucial for determining the fluidity of the slurry, which subsequently influences reactant concentrations, mixing effectiveness, ion transport, and overall mass transfer. This ratio can be expressed in various forms, such as L/S (mL/g) or solid-to-liquid (S/L, g/L). The reported ranges for optimal L/S ratios vary widely: between 5 and 50 mL/g [7], 50 to 150 g/L [21], and between 6 and 9 for liquid-to-solid ratios [16].

A clear optimum exists, but its value is specific to the system in question. For instance, approximately 100 g/L is optimal for CCR-steel slag [21], L/S = 8 for ground granulated blast furnace slag and calcium carbide residue blend (GGBS/CCR) [16], and L/S = 30 mL/g for CCR in a bubble column setting [7].

An ideal L/S ratio is essential because very low ratios can impede mixing and mass transfer, while excessively high ratios dilute the concentration of calcium ions (Ca²⁺), which diminishes supersaturation levels and slows the rate of precipitation.

The main issue lies in the significant differences in reported optimal values, which can vary by an order of magnitude across different studies. Low L/S ratios produce viscous and poorly mixed slurries, thereby hampering the transport of CO₂ and ions [6, 22]. Conversely, high L/S ratios decrease the concentration of leached Ca²⁺, leading to lower ion activity and a reduction in nucleation and growth rates [6]. W. Li et al. [7] connected their optimum L/S ratio to achieving an ideal "discrete bubble regime," which enhances gas-liquid contact in their specific reactor setup.

4.5. Particle Size and Pre-treatments

The size of particles has a direct impact on the available surface area for reactions and the distance ions must travel within those particles, influencing both the initial reaction rates and the overall conversion efficiency. Current data on this topic is limited. For instance, Li et al. [21] found that reducing the particle size of steel slag to less than 75 µm significantly enhanced the efficiency of CO₂ removal and its sequestration capacity. Similarly, Wang et al. [6] noted that smaller CCR particles tend to increase the carbonation rate. Studies have examined the effects of pre-treatments, such as grinding, on CCR and slag in improving carbonation efficiency [16, 20].

Where research has been conducted, a smaller particle size has been shown to improve carbonation performance. Although no contradictions have emerged, the potential interactions in mixed particle systems remain unexplored. The observed improvements are consistently linked to kinetic factors, including a greater specific surface area for dissolution and shorter diffusion distances for ions within the particles [6, 21].

4.6. Integrated Perspective on Parameter Interactions

Across the reviewed literature, no single parameter independently governs carbonation efficiency. Instead, performance is dictated by the interplay between dissolution kinetics, CO₂ mass transfer,

and product-layer formation, with different parameters dominating at different stages of the reaction. Apparent contradictions, such as positive versus negligible temperature effects or declining efficiency at high CO₂ concentrations, are best understood as transitions between rate-limiting regimes rather than inconsistencies in experimental results.

This regime-based interpretation reinforces the need for system-specific optimization rather than universal operating conditions and highlights why CCR–slag synergistic systems often outperform CCR alone by redistributing carbonate precipitation and mitigating passivation under otherwise unfavorable conditions.

The consolidated effects of key operational parameters, including consistent trends and non-monotonic behavior, are summarized in Table 6.

Table 6: Summary of Parameter Influences

Parameter	Reported Conditions & Outcome	Consistent Trends	Contradictions/ Non-monotonic Outcomes	Author-Stated Causes
pH & Chemistry	Sparse control; used as progress indicator (pH→7). Glycine increases reaction time and stabilizes vaterite.	pH decreases monotonically during reaction.	Lack of quantitative rate-pH relationships.	Thermodynamic driver (pH drop). Kinetic control by modifiers (polymorph selection).
Temperature	Range: ambient to 80°C. Positive effect up to an optimum (55-60°C common).	Increased Temperature enhances kinetics (dissolution, diffusion).	One study found effect on CCR "negligible". Optima exist; very high Temperature can reduce yield.	Kinetic: Faster dissolution/diffusion. Thermodynamic: Reduced CO ₂ solubility at high T.
CO ₂ Concentration/ flow rate	Pure CO ₂ to 25% flue gas. Optimal conc./flow exists (e.g., 15 vol%, 9 L/min & 2L/min).	Higher CO ₂ increases driving force.	Performance declines beyond optimum. Different metrics (capacity vs. efficiency) show opposite trends.	Mass Transfer: Improved driving force. Kinetic/Side Reaction: Rapid passivation; CaCO ₃ to Ca(HCO ₃) ₂ reversal.
L/S Ratio	Wide range (e.g., 5-50 mL/g, 50-150 g/L). Strong, system-	Optimum exists. Low ratio: poor mixing. High ratio: dilution.	Optimal value varies dramatically across studies.	Mass Transfer: Mixing & viscosity. Thermodynamic/Kinetic: Ca ²⁺

	specific optimum.			concentration & supersaturation.
Particle Size	Limited data. Finer particles (<75 μm) improve performance.	Smaller size improves kinetics.	Effect is qualitatively acknowledged but rarely quantified for blends.	Kinetic: Increased surface area; reduced diffusion path length.

5. Carbonated Products as SCMs: Replacement and Performance

The direct aqueous carbonation of calcium carbide residue (CCR) and its blends with various slags not only sequesters CO_2 but also produces materials with potential value as supplementary cementitious materials (SCMs). This section reviews the reported performance of these carbonated products when used in cement paste and mortar systems, focusing on replacement levels, effects on properties, and the proposed underlying mechanisms.

Figure 4 schematically summarizes the dominant roles of carbonation-derived CCR and CCR–slag products when used as supplementary cementitious materials, emphasizing physical filler and hydration nucleation effects rather than latent hydraulic reactivity.

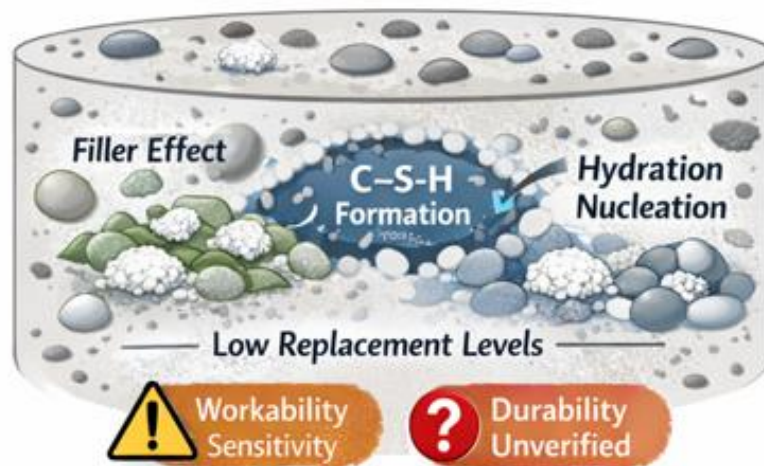


Figure 4. Conceptual representation of the role of carbonation-derived CCR and CCR–slag products when used as supplementary cementitious materials (SCMs). The primary contributions arise from physical filler and hydration nucleation effects rather than latent hydraulic reactivity, with performance strongly dependent on particle morphology, replacement level, and surface characteristics.

5.1. Typical Replacement Percentages

Reported replacement levels of cement with carbonated products vary significantly, reflecting differences in material composition and research focus. The studies can be broadly categorized by the type of carbonated material used, as summarized in Table 7.

Table 7: Reported Replacement Levels of Carbonated Products as SCMs

Study	Carbonated Material	Replacement Level (%) by mass of cement)	Replacement Type	Stated Optimal Level
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[18]	Calcium Carbide Residue (CCR)	10%	Single-level	Not Applicable
[17]	CCR-Steel Slag (SS) Composite	20%	Single-level	Not Applicable
[19]	CCR (Calcite or Vaterite)	10%, 20%	Multi-level	10% for both polymorphs
[16]	GGBS/CCR Blend	50%	Single-level	Not Applicable
[23]	Electric Arc Furnace (EAF) Slag	10%, 20%, 30%	Multi-level	30%
[24]	Steel Slag(specific not specified)	40%	Single-level	Not Applicable

Across the reviewed studies, replacement levels range widely from 10% to 50% by mass of cement, reflecting the absence of a standardized or consensus-driven approach to dosage selection. A large proportion of the studies relied on single-level replacement designs, including Chen et al. [18] at 10% calcium carbide slag (CCS), S. Li et al. [17] at 20% CCS–steel slag composite, Zhang et al. [16] at 50% GGBS/CCS blend, and Biava et al. [24] at 40% steel slag. In these cases, no optimal replacement level was identified, as comparative multi-level evaluation was not conducted. Consequently, performance outcomes reported in these studies remain constrained to the selected dosage and cannot inform broader optimization.

In contrast, only two studies systematically investigated multiple replacement levels. Zhou et al. [19] examined carbonated CCS containing either calcite or vaterite polymorphs at 10% or 20% replacement. Their results clearly indicated 10% as the optimal level for both polymorphs, with higher replacement leading to reduced or non-proportional performance gains. This suggests that, for CCR-derived carbonates, excessive substitution may dilute clinker hydration.

Similarly, Bonfante et al. [23] evaluated carbonated EAF slag at 10%, 20%, and 30% replacement and identified 30% as the optimal dosage in terms of later-age strength development. Unlike CCR systems, the higher optimal level observed in this study was attributed to the formation of amorphous silica during carbonation, which enhanced pozzolanic reactivity and compensated for clinker dilution at higher replacement.

Overall, the data reveal no unified optimal replacement level across carbonated materials, with reported values strongly dependent on material origin, carbonation products, and reactivity mechanisms. The dominance of single-level experimental designs represents a major limitation in the current literature and hampers cross-study comparison.

5.2. Effect on Mortar Properties

5.2.1. Strength Development

The compressive strength development is the most frequently reported hardened property. The effects are age- and material-dependent, as synthesized in Table 8. It should be noted that the reported compressive strength values were obtained using different specimen types and test standards, and therefore should be interpreted in terms of relative trends within each study rather than direct numerical comparison across studies

Table 8. Reported compressive strength development of mortars or pastes incorporating carbonated SCMs, as reported in the original studies. Test methods and specimen geometries vary between studies; values are therefore compared qualitatively rather than quantitatively.

Study	Material	Replacement	Control/Uncarbonated Strength (MPa)		Carbonated Strength (MPa)	Standards
[18]	Carbonated CCR	10%	3 days	~ 16 (control)	~ 19	Not specified
			7 days	~ 19 (control)	~ 28	
			28 days	~ 37 (control)	~ 36	
[17]	Carbonated CCR-SS Synergy	20%	3 days	~ 37 (control)	~ 34	Not specified
			7 days	~ 43 (control)	~ 42	
			28 days	~ 47 (control)	~ 54	
[19]	Carbonated CCR (Vaterite)	10% (optimal)	3 days	~ 35 (control) ~ 33 (Calcite)	~ 40	Not specified
			7 days	~ 42 (control) ~ 41 (Calcite)	~ 47	
			28 days	~ 58 (control) ~ 49 (Calcite)	~ 61	
[16]	Carbonated CCR-GGBS Synergy	50% of binder	3 days	~ 15 (uncarbonated)	~ 19	Not specified
			7 days	~ 25 (Uncarbonated)	~ 30	
			28 days	~ 43 (uncarbonated)	~ 45	
[23]	Carbonated EAF Slag	30% (optimal)	3 days	46 (control); 33(uncarbonated)	28	BS EN 196-1
			7 days	54 (control); 38 (uncarbonated)	38	
			28 days	63 (control); 47 (uncarbonated)	50	

The compressive strength data presented in Table 8 provide a consolidated overview of how carbonated industrial by-products perform as supplementary cementitious materials in mortar systems when benchmarked against control or uncarbonated references. Reported values are taken directly from the cited studies; approximate values (~) reflect data digitized from published figures. Across the studies reviewed, a clear age-dependent behavior emerges: carbonated materials rarely deliver uniform strength enhancement at all curing ages, but instead exhibit a balance between early-age physical effects and later-age chemical contributions that is strongly dependent on material chemistry and replacement level.

At early ages (3–7 days), low to moderate incorporations of carbonated products (typically 10–20% replacement) frequently result in strength levels comparable to, or slightly exceeding, those of control mortars. This trend is consistently observed for carbonated CCR at 10% replacement [18] and for vaterite-rich carbonated CCR systems [19], as well as for carbonated CCR-GGBS blends at higher total binder replacement [16]. These early-age responses are best explained by the nucleation and filler effects induced by finely precipitated CaCO_3 formed during carbonation. This mechanism is consistent with the conceptual framework illustrated in Figure 4, where finely dispersed carbonate phases promote early hydration without contributing significantly to later-age chemical binding. The presence of finely dispersed carbonate particles increases the number of nucleation sites for C–S–H formation, accelerates hydration kinetics, and promotes early microstructural densification, partially compensating for clinker dilution.

However, this early-age benefit is not universal. In systems with higher replacement levels or reduced clinker availability, such as the 20% carbonated CCR–steel slag composite studied by S. Li et al. [17] and the 30% carbonated EAF slag system reported by Bonfante et al. [23], early-age compressive strength is consistently lower than that of the Portland cement control. In these cases, the dilution of clinker and the consumption of reactive Ca-bearing phases during carbonation outweigh the nucleation advantage, resulting in delayed strength development despite improved particle packing.

At 28 days, the trends diverge more clearly depending on whether carbonation produces solely inert carbonate phases or preserves secondary reactivity. Pure carbonated CCR systems tend to show strength parity or marginal reductions relative to control mortars at later ages [18], indicating that once the nucleation effect is exhausted, crystalline CaCO_3 contributes little to continued strength gain. In contrast, systems incorporating carbonated slag or slag-CCR synergies consistently demonstrate strength recovery or enhancement at 28 days. Notably, S. Li et al. [17] reported a substantial increase in 28-day strength for carbonated CCR–steel slag mortars, while Bonfante et al. [23] showed that carbonated EAF slag outperformed its uncarbonated counterpart at the same replacement level, even though both remained below the Portland cement control.

The superior later-age performance of these systems can be attributed to the presence of reactive amorphous phases generated or preserved during carbonation, particularly silica-rich domains capable of participating in secondary pozzolanic reactions. This behavior contrasts sharply with CCR-only systems and highlights that carbonation does not inherently render a material inert; rather, its effect depends on the initial mineralogy and the resulting phase assemblage after treatment.

An additional layer of complexity is introduced by the polymorphism of the carbonate phases formed. Zhou et al. [19] demonstrated that vaterite-rich carbonated CCR consistently outperformed calcite-based equivalents, especially at 28 days. Given vaterite's higher solubility and metastability, its enhanced nucleation efficiency and potential for dissolution-reprecipitation processes provide a plausible explanation for the observed strength differences. This finding underscores that carbonated products cannot be treated as chemically or functionally equivalent solely based on CaCO_3 content.

Overall, the apparent contradictions observed across the studies, early-age strength gains versus losses, low optimal replacement for CCR versus higher optimal levels for carbonated slag, are not conflicting outcomes but reflections of competing mechanisms whose dominance shifts with

curing age, replacement level, and material chemistry. The table collectively demonstrates that carbonation-derived SCM performance in mortar is governed by the interplay between nucleation effects, clinker dilution, and secondary reactivity. Failure to account for these interactions, particularly in single-level replacement studies, risks misinterpreting carbonation as either universally beneficial or detrimental. Instead, the evidence supports a more nuanced view in which carbonation can be strategically leveraged to enhance mortar performance, provided that material selection, replacement level, and post-carbonation phase composition are carefully aligned.

5.2.2. Workability

The incorporation of carbonated SCMs consistently influences fresh-state properties, with workability emerging as a critical practical constraint. Across most studies, carbonated materials increase water demand and reduce flowability relative to both Portland cement controls and uncarbonated counterparts [16, 23]. This behavior is primarily attributed to the increased specific surface area generated during carbonation and the presence of fine CaCO_3 precipitates on particle surfaces, which enhance water adsorption and restrict particle mobility as shown schematically in figure 4.

Carbonation also accelerates setting, as reported by Chen et al. [18] and Zhang et al. [16], due to the well-established nucleation effect of CaCO_3 on cement hydration. While early stiffening may be advantageous for rapid strength development, it poses challenges for placement and finishing if not carefully managed.

Importantly, workability penalties are not intrinsic to carbonation but are strongly dependent on carbonate polymorphism and particle morphology. Zhou et al. [19] demonstrated that vaterite-rich carbonated CCR can improve flowability at an optimal replacement level (10%) due to its spherical morphology, which induces a “ball-bearing” effect. In contrast, calcite-dominated systems consistently reduce flow due to the angular and prismatic shapes of their particles. This distinction highlights that carbonated products should not be treated as rheologically equivalent solely based on chemical composition.

From a practical perspective, the literature indicates that carbonated SCMs generally require either reduced replacement levels, optimized polymorph control, or chemical admixture adjustment to maintain acceptable workability. Failure to address these factors risks undermining otherwise favorable mechanical performance, particularly in high-replacement systems.

5.2.3. Durability Parameters: A Critical Unverified Frontier

Durability performance represents the most significant and unresolved barrier to the practical deployment of carbonated SCMs derived from CCR and CCR-slag systems. While multiple studies report microstructural refinement resulting from carbonate precipitation and filler effects [17, 18, 19], these observations remain indirect indicators rather than validated durability outcomes.

Critically, none of the reviewed studies report standardized durability testing, including chloride ingress resistance, water permeability, sulfate resistance, carbonation resistance, or dimensional stability. As a result, the long-term behavior of mortars incorporating carbonated SCMs under aggressive exposure conditions remains unknown.

This gap is particularly consequential for several reasons. First, the stability of carbonate-rich binders in sulfate-bearing environments is uncertain, with potential risks of secondary phase formation such as thaumasite. Second, the interaction between pre-carbonated SCMs and subsequent atmospheric carbonation of cementitious matrices introduces a paradox: while

carbonate precipitation may densify pore structure, it may also reduce alkalinity and compromise steel reinforcement protection. Third, the influence of fine carbonate particles on drying and autogenous shrinkage has not been examined, despite its relevance to cracking susceptibility and transport-controlled degradation.

Based on current evidence, durability claims for carbonated SCMs must be regarded as unsubstantiated rather than implicitly favorable. Future research should prioritize (i) transport properties (chloride diffusion and sorptivity), (ii) chemical resistance under sulfate and acidic exposure, and (iii) dimensional stability, using standardized test methods. Without such data, the transition of carbonation-derived SCMs from laboratory-scale concepts to engineering materials remains speculative

6. Conclusions and Perspectives

Direct aqueous carbonation of calcium carbide residue (CCR) and CCR-slag systems represents a technically promising yet mechanistically constrained route for CO₂ sequestration and waste valorization. Carbonation of CCR alone is intrinsically limited by rapid surface passivation arising from localized CaCO₃ precipitation, which shifts the reaction into a diffusion-controlled regime and restricts overall conversion. Conversely, iron- and steel-making slags exhibit inherently slower carbonation kinetics due to limited calcium availability and kinetically inert silicate matrices. These complementary limitations explain the growing interest in combined CCR-slag carbonation systems.

A central conclusion of this review is that the superior performance reported for CCR-slag systems arises not from increased alkalinity or calcium content alone, but from a redistribution of carbonate precipitation pathways. The presence of slag redirects CaCO₃ formation away from CCR particle surfaces toward alternative nucleation sites, mitigating passivation and enabling sustained calcium dissolution. This mechanistic shift reconciles previously contradictory observations regarding temperature, CO₂ concentration, and pressure effects, which are best interpreted as transitions between competing rate-controlling regimes rather than inconsistencies in experimental outcomes. The properties of carbonation-derived products further depend on the morphology and polymorphism of the precipitated carbonates. When used as supplementary cementitious materials (SCMs), carbonated CCR and CCR-slag products generally act as fillers and hydration nucleation agents rather than latent hydraulic binders. Strength enhancement is therefore most pronounced at low replacement levels and is sensitive to carbonate phase assemblage, while workability and setting behavior are governed by particle morphology and surface characteristics. Importantly, durability performance remains largely unverified, and current evidence is insufficient to support claims of long-term resistance under aggressive exposure conditions.

From a design perspective, effective carbonation-based waste valorization requires system-specific optimization rather than universal operating conditions. Reaction parameters must be selected to balance calcium dissolution, CO₂ mass transfer, and precipitation kinetics while avoiding premature passivation. For SCM applications, carbonation conditions should be tailored to produce morphologically favorable carbonate phases and controlled particle size distributions, rather than maximizing CO₂ uptake alone.

Looking forward, several priorities emerge. Future studies should (i) explicitly link carbonation conditions to carbonate polymorphism and spatial precipitation behavior, (ii) evaluate durability

using standardized transport and chemical resistance tests, and (iii) assess scalability and robustness under realistic gas compositions and reactor configurations. Addressing these gaps will be essential to transition CCR–slag carbonation from laboratory demonstrations to reliable, application-oriented technologies.

7. DECLARATION:

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7.3. Clinical trial number: not applicable

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7.6. Author Contributions

Zoum Fon Alain: Conception, Methodology, Formal Analysis, Visusalisation, Validation
Manuscript drafting and editing

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Graphical Abstract

