

Article

Carbonation Behavior and Pore-Microstructure Evolution of Press-Formed BOF Slag Leaching Residue Briquettes

Jianbao Zhang ^{1,2}, Linfei Li ¹, Junguo Li ^{1,*}, Xuan Guo ¹, Yitong Wang ^{1,3} , Yanan Zeng ¹ and Yajun Wang ^{1,3,*}

¹ College of Metallurgy and Energy, North China University of Science and Technology, Tangshan 063210, China; zhangjianbao@ncst.edu.cn (J.Z.); lilinfei@stu.ncst.edu.cn (L.L.); gx1031@ncst.edu.cn (X.G.); wangyt@ncst.edu.cn (Y.W.); zengyanann@126.com (Y.Z.)

² Graduate School, North China University of Science and Technology, Tangshan 063210, China

³ Yanzhao Iron and Steel Laboratory, North China University of Science and Technology, Tangshan 063210, China

* Correspondence: lijg99@163.com (J.L.); wangyj@ncst.edu.cn (Y.W.)

Abstract

To achieve the stabilization and high-value utilization of Basic oxygen furnace (BOF) slag leaching residues, this study proposes a synergistic strategy combining compaction and pressurized carbonation. The effects of moisture content, molding pressure, holding time, and carbonation duration on the carbonation efficiency, mechanical properties, and surface characteristics of the residue briquettes were systematically investigated. Using a combination of orthogonal and single-factor experiments with the carbonation weight gain rate as the primary indicator, this study reveals the intrinsic mechanism by which forming parameters regulate CO₂ mass transfer and reaction efficiency through pore structure modification. The results indicate that moisture content is the dominant factor influencing carbonation efficiency, with an optimal moisture range achieving a balance between reaction kinetics and gas diffusion. Furthermore, molding pressure and holding time dictate the effective carbonation depth by altering the pore size distribution. The carbonation process significantly enhances the compressive strength of the specimens, reduces the surface pH from 12.45 to approximately 10.2, and decreases surface roughness, thereby improving environmental compatibility. These findings provide a theoretical foundation for the high-value application of leached steel slag in marine ecological materials and carbon sequestration.

Keywords: BOF slag; leaching residue; pressure forming; carbonation; high-value utilization



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1. Introduction

The iron and steel industry is a cornerstone of modern industrial civilization; however, it simultaneously ranks among the most carbon-intensive sectors globally. In China alone, crude steel production exceeded 0.961 billion tonnes in 2025, generating an estimated 115–144 million tonnes of basic oxygen furnace (BOF) slag as a byproduct [1,2]. BOF slag is characterized by its high content of free calcium oxide (f-CaO, typically 5–15 wt.%) and free magnesium oxide (f-MgO), which render it volumetrically unstable and restrict its direct application in civil engineering without prior treatment [3,4]. Conventional stabilization approaches (including open-air weathering, steam aging, and autoclave treatment) are time-consuming, energy-intensive, and produce no value-added products, thereby limiting the overall resource utilization rate of BOF slag, which remains below 30% in China [5,6].

A promising emerging strategy involves the aqueous leaching of BOF slag to extract soluble calcium and alkaline components for indirect mineral carbonation or nutritional supplements for microalgae cultivation [7]. During this process, free CaO and portlandite ($\text{Ca}(\text{OH})_2$) dissolve preferentially, releasing Ca^{2+} ions that subsequently react with dissolved CO_2 to precipitate CaCO_3 , thereby achieving simultaneous CO_2 sequestration and slag stabilization [8]. However, this leaching process invariably generates a substantial volume of solid residue, the so-called leaching residue or leached slag. The leached slag retains significant quantities of silicates (C_2S), calcium ferrites (C_2F), and the RO phase (a FeO-MgO-MnO solid solution) [9]. This residue represents an underutilized secondary solid waste, as its residual reactive phases still possess latent carbonation potential that has thus far not been systematically harnessed [10].

Basic oxygen furnace (BOF) slag is a major metallurgical byproduct formed during steelmaking through reactions of slag-forming materials, furnace lining erosion, and oxidation of metallic elements. Its mineral composition primarily consists of the RO phase, calcium silicate, calcium ferrite, free CaO, and free MgO, with CaO and Fe_2O_3 combined accounting for over 75% of its chemical content, alongside trace heavy metals such as Cr, V, and Pb [11]. Current research and engineering practices have identified multiple potential applications for BOF slag: it can serve as a construction aggregate or cement component in building materials; utilize its alkaline mineral properties for CO_2 mineralization sequestration; extract nutrients for aquaculture; or be processed into artificial reefs for marine ecological restoration. However, large-scale application remains constrained by several limitations—including poor volumetric stability due to free oxides, inadequate environmental compatibility caused by high alkalinity, potential heavy metal leaching risks, low added value from direct utilization, and limited mass transfer efficiency during carbonation reactions—all requiring further process optimization to fully realize its resource potential [12].

Accelerated carbonation of steel slag compacts (briquettes) has attracted considerable research attention as a strategy for simultaneously achieving CO_2 utilization, volume stabilization, and mechanical property enhancement [13,14]. The Carbstone technology developed at KU Leuven demonstrated that steel slag briquettes carbonated under elevated CO_2 concentrations (15–20%) can achieve compressive strengths exceeding 30 MPa with a CO_2 fixation efficiency of approximately 18 wt.%, while maintaining heavy metal leaching concentrations below EU regulatory thresholds [15]. Japan's NEDO research consortium further proposed a “steam activation-briquette carbonation” composite process, in which steam pretreatment increases specific surface area and porosity by 30–40%, enhancing CO_2 diffusion kinetics and improving overall carbonation efficiency by approximately 40% relative to conventional powder-based methods [16]. The resultant carbonated briquettes exhibit surface alkalinity (pH ~9–10) favorable for coral larvae settlement, rendering them promising candidates for marine artificial reef construction [17]. Domestically, collaborative research between Baosteel Group and Shanghai Jiao Tong University demonstrated that briquettes incorporating 10 wt.% fly ash achieve a compressive strength of 25 MPa and a CO_2 fixation efficiency of 15% after 48 h of carbonation.

Despite these advances, critical knowledge gaps persist. The vast majority of prior studies have focused on fresh or mildly aged BOF slag as the starting material, with the carbonation behavior of leached slag residues remaining largely unexplored. Leaching fundamentally alters the phase assemblage, pore structure, and surface chemistry of the slag, generating a precursor whose carbonation kinetics and structure-property evolution differ substantially from those of untreated slag [18]. Furthermore, the interrelationships among compaction parameters (pressure and holding time), moisture content, pore structure development, CO_2 mass transfer efficiency, and ultimate carbonation degree have not

been systematically quantified for leached slag briquettes. The absence of such mechanistic understanding constitutes a critical barrier to the rational design of high-performance carbonated products from this secondary waste stream.

Current research on carbonation of steel slag pellets exclusively employs original converter slag as raw material. The high content of free CaO in this material often leads to poor volumetric stability, with reaction activity concentrated at the surface, resulting in limited carbonation uniformity and structural stability [19]. However, after water extraction of nutrients from the steel slag, the residue—classified as secondary solid waste—exhibits significantly reduced free CaO content and conversion to $\text{Ca}(\text{OH})_2$. This modification alters the mineral phase composition and reaction active sites, shifting the carbonation rate-limiting mechanism from “surface chemical reactions” to “coordinated pore mass transfer and reaction kinetics,” fundamentally differing from the carbonation behavior of original steel slag. Currently, the carbonation mechanism for pellets derived from leached steel slag residues remains unclear, hindering the development of a cascaded utilization pathway for steel slag as “efficiently leached carbonated building material.”

To bridge this knowledge gap, the present study proposes a synergistic processing approach that couples press-forming with pressurized accelerated carbonation for the stabilization and valorization of BOF slag leaching residues. A three-factor, three-level orthogonal experimental design [20], supplemented by single-factor experiments, was employed to systematically elucidate the effects of moisture content, compaction pressure, pressure-holding time, and carbonation duration on: (i) the carbonation weight gain rate (as the primary kinetic index), (ii) the compressive strength, (iii) the pore structure, and (iv) the surface physicochemical properties (pH and roughness). The overarching objective is to establish a quantitative structure-mass transfer-reaction coupling model, thereby providing a rigorous theoretical and experimental foundation for the high-value application of leached BOF slag briquettes as marine ecological materials and geological carbon sinks.

2. Materials and Methods

2.1. Experimental Materials Preparation

2.1.1. Original BOF Slag

BOF slag was sourced from a local steel plant (Tangshan, China) and subjected to jaw crushing followed by ball milling. The resulting powder was sieved through a 200-mesh standard sieve (aperture ≤ 0.074 mm) to ensure particle size uniformity. The powder was subsequently oven-dried at 105°C for 24 h to eliminate adsorbed moisture before characterization and experimentation. X-ray fluorescence (XRF, Axios MAX, PANalytical, Almelo, The Netherlands) analysis revealed that CaO (36.8 wt.%) and Fe_2O_3 (38.8 wt.%) were the predominant oxide phases, with SiO_2 (9.2 wt.%), MgO (4.5 wt.%), and Al_2O_3 (1.3 wt.%) as secondary constituents (Table 1). The high content of CaO is characteristic of BOF slag and underlies its substantial carbonation potential.

Table 1. Chemical composition of steel slag.

Oxide	CaO	SiO_2	Fe_2O_3	Al_2O_3	MgO	Others
Mass percent/%	36.8	9.2	38.8	1.3	4.5	5.2

2.1.2. Leaching Procedure

Aqueous leaching was conducted by dispersing 200 g of BOF slag powder in 2000 mL of deionized water (liquid-to-solid ratio = 10:1 mL/g) in a sealed polyethylene vessel at ambient temperature ($25 \pm 2^\circ\text{C}$) for 7 days under continuous magnetic stirring at 200 rpm. Upon completion, the suspension was vacuum-filtered through a $0.45\ \mu\text{m}$ membrane to

separate the leachate from the solid residue. The leaching residue was oven-dried at 105 °C for 24 h and stored in a desiccator until use. Phase evolution resulting from leaching was monitored by X-ray diffraction (XRD, D8 Advance, Bruker, Karlsruhe, Germany; Cu K α radiation, $2\theta = 10\text{--}90^\circ$, scan rate = $2^\circ/\text{min}$).

Experimental measurements revealed that the dry mass loss rate of steel slag after leaching treatment reached 3%. Meanwhile, the pH value of the leaching supernatant was measured to be 12, indicating strong alkalinity. Analysis showed that free calcium oxide and other soluble alkaline components in the steel slag were largely released during leaching, which is the primary cause of mass loss in the solid fraction and the high alkalinity of the leaching solution. On the other hand, iron primarily exists in chemically stable RO-phase minerals within the steel slag; consequently, its migration to the liquid phase during leaching was minimal, resulting in extremely low leaching concentrations.

2.2. Orthogonal Experimental Design

A standard $L_9(3^3)$ orthogonal array was employed to efficiently evaluate the combined effects of three process variables—compaction pressure (10, 15, and 20 MPa), molding holding time (1, 2, and 3 min), and moisture content (4.76, 6.25, and 9.09 wt.%)—on the carbonation behavior of the leached slag briquettes (Table 2). Before compaction, deionized water was uniformly incorporated into the leached slag powder, and the mixture was homogenized by hand-mixing for 5 min. The conditioned mixture was then uniaxially pressed into cylindrical specimens (diameter = 20 mm, height $\approx$ 10 mm) using a hydraulic press. Each experimental condition was replicated three times to ensure statistical reliability. The carbonation weight gain rate served as the primary response variable and was calculated according to Equation (1):

$$\gamma = \frac{m_2 - m_1}{m_1} \times 100\% \quad (1)$$

Table 2. Orthogonal experimental design for carbonation of leached slag Blocks.

NO.	Pressure (MPa)	Molding Holding Time (min)	Moisture Content (wt.%)
1	10	1	4.76
2	10	2	6.25
3	10	3	9.09
4	15	1	6.25
5	15	2	9.09
6	15	3	4.76
7	20	1	9.09
8	20	2	4.76
9	20	3	6.25

γ represents the carbonation gain rate (%), m_2 denotes the mass of the sample after carbonation and drying to a constant weight (g), and m_1 indicates the dry mass of the sample before carbonation (g).

2.3. Carbonation Procedure and Performance Testing

Immediately after demolding, the briquettes were transferred to a custom-built stainless-steel pressure reactor (volume = 2 L). The reactor was purged with 99.9% purity CO $_2$ for 5 min to displace residual air, and the CO $_2$ pressure was then regulated to 0.5 MPa using a pressure-reducing valve. Carbonation was conducted at room temperature ($25 \pm 2^\circ\text{C}$) for the designated duration (0.5, 1, 2, 4, 6, or 8 h for single-factor experiments; 6 h for orthogonal experiments). Upon completion, the reactor was slowly depressurized,

and the carbonated briquettes were retrieved and oven-dried at 105 °C for 24 h before weighing and further characterization.

Compressive strength testing: A single-axis compression test was conducted using a microcomputer-controlled electronic universal testing machine at a loading rate of 1 mm/min. The specimen consisted of a cylinder with a diameter of 20 mm and a height of 20 mm. The formula for calculating single-axis compressive strength is as follows:

$$f_c = \frac{F}{S} \quad (2)$$

In the formula: f_c represents the uniaxial compressive strength (in MPa); F is the failure load of the specimen (in N); S is the compressive area of the specimen (in mm²), calculated as $S = \pi d^2 / 4$ (where d is the specimen diameter in mm).

2.4. Surface Property Detection of Briquettes

Three-dimensional surface topography and quantitative roughness parameters were measured using a 3D optical surface profilometer (Contour GT-K, Bruker, Karlsruhe, Germany) [21]. For each specimen, three randomly selected areas (500 µm × 500 µm) were measured, and the arithmetic mean roughness (R_a), root-mean-square roughness (R_q), and maximum height (R_z) were extracted.

Scanning electron microscopy coupled with energy-dispersive X-ray spectroscopy (SEM-EDS, ZEISS Sigma 300, Oberkochen, Germany) was employed to characterize surface microstructure and elemental distribution [22].

Surface pH was measured using a flat-membrane pH electrode (FiveEasy Plus, Mettler-Toledo, Greifensee, Switzerland) in contact mode, with three replicate measurements per specimen.

3. Results and Discussion

3.1. Mineral Phase Evolution During Leaching

XRD analysis (Figure 1) of the as-received BOF slag revealed a phase assemblage comprising the RO phase (a FeO-MgO-MnO solid solution, ~38 wt.%), dicalcium silicate (β -C₂S, 33.9 wt.%), dicalcium ferrite (C₂F, 20.5 wt.%), and free CaO (f -CaO, 16.9 wt.%). The elevated f -CaO content is a critical source of both volume instability and carbonation reactivity. After 7 days of aqueous leaching, significant mineralogical transformations were observed: (i) the diffraction peaks of the RO phase decreased markedly in intensity, indicating partial dissolution of the FeO and MnO end-members; (ii) f -CaO content was drastically reduced from 16.9 to 2.1 wt.%, confirming near-complete hydration and dissolution of the free lime fraction; (iii) a pronounced portlandite (Ca(OH)₂) peak emerged at 18.0° 2 θ (relative content: 15.2 wt.%), reflecting the hydration reaction: $\text{CaO} + \text{H}_2\text{O} \rightarrow \text{Ca(OH)}_2$; and (iv) trace calcite (CaCO₃, 1.8 wt.%) was detected, attributable to natural atmospheric carbonation during processing. Collectively, these transformations effectively suppress volume instability by converting the expansive f -CaO phase into stable Ca(OH)₂, while simultaneously providing a Ca²⁺-rich precursor that is kinetically favorable for subsequent accelerated carbonation.

3.2. Influence of Process Parameters on Carbonation Behavior of Slag Briquettes

3.2.1. Orthogonal Experiment Analysis

The carbonation weight gain rates (γ) obtained across the nine experimental conditions of the L₉(3³) orthogonal array ranged from 5.76% to 8.93% (Table 3), confirming that the leached slag residue retains appreciable carbonation reactivity despite the prior extraction of a substantial fraction of its soluble alkaline components.

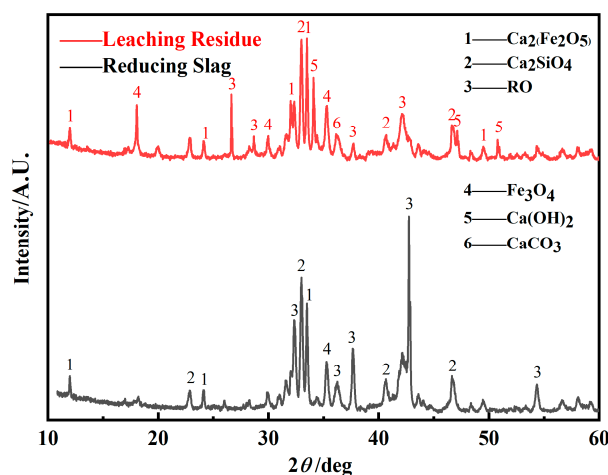


Figure 1. XRD patterns of the original and the 7-day leached BOF slags.

Table 3. Carbonation weight gain rate under different block-making processes for leaching slag Briquettes.

NO.	Pressure (MPa)	Molding Holding Time (min)	Moisture Content (wt.%)	Weight Gain Rate (%)
1	10	1	4.76	5.76 ± 0.21
2	10	2	6.25	7.66 ± 0.18
3	10	3	9.09	7.34 ± 0.25
4	15	1	6.25	7.66 ± 0.15
5	15	2	9.09	8.93 ± 0.22
6	15	3	4.76	6.22 ± 0.19
7	20	1	9.09	8.35 ± 0.17
8	20	2	4.76	7.55 ± 0.20
9	20	3	6.25	8.79 ± 0.16

Range analysis was performed to rank the relative importance of the three process variables (Table 4). The calculated range values were: $R_{\text{moisture}} = 1.697$, $R_{\text{pressure}} = 1.310$, and $R_{\text{holding time}} = 0.790$, yielding the factor priority order: moisture content > compaction pressure > pressure-holding time. This ranking indicates that moisture content exerts a dominant influence on carbonation efficiency, which can be rationalized by its dual role: (i) water serves as the reaction medium for Ca^{2+} dissolution and CO_3^{2-} formation, both of which are prerequisite steps for CaCO_3 precipitation; and (ii) water partially fills the pore network, and its spatial distribution critically modulates the tortuosity and effective diffusivity of CO_2 through the briquette. An optimal moisture content strikes a balance between maximizing dissolved ion availability and preserving sufficient open porosity for gas-phase CO_2 transport.

Table 4. Range analysis results.

NO.	Pressure (MPa)	Molding Holding Time (min)	Water Addition Rate (wt.%)
Mean value 1	6.920	7.257	6.510 ± 0.20
Mean value 2	7.603	8.047	8.037 ± 0.16
Mean value 3	8.230	7.450	8.207 ± 0.21
Range	1.310	0.790	1.697

Upon further integration with the mean analysis of each factor, it is evident that at a compaction pressure of 20 MPa, a holding time of 2 min, and a moisture content of 9.09 wt.%,

the carbonation weight gain rate of the leached slag briquette peaks. This finding suggests that enhancing the molding density moderately while maintaining an appropriate moisture content facilitates the efficient diffusion and reaction of CO₂ within the slag briquette under specific CO₂ partial pressure and reaction time conditions. It is noteworthy that the optimal combination identified through orthogonal analysis does not solely rely on “high pressure or high water content” scenarios but rather signifies a harmonious equilibrium among pore structure, liquid phase distribution, and gas mass transfer. This outcome lays the groundwork for mechanistic analysis in subsequent single-factor experiments [23].

To further verify the statistical significance of each factor’s impact, an analysis of variance (ANOVA) was performed on the orthogonal experimental data, with results presented in Table 5.

Table 5. ANOVA results of carbonation weight gain rate.

Factor	Quadratic Sum	Mean Square
Pressure	2.576	1.288
Molding holding time	0.973	0.486
Water addition rate	4.921	2.460
error	0.280	0.140

The analysis of variance results demonstrated that water content had a highly significant effect on the carbonation weight gain rate, while forming pressure exhibited a significant influence; however, the impact of forming pressure holding time was not statistically significant. These findings fully align with the order of effects identified in the range analysis, providing statistical validation that moisture is the key factor governing the carbonation efficiency of BOF steel slag briquettes during carbonation.

3.2.2. Single-Factor Experimental Analysis

Figure 2 demonstrates the effect of process parameters on carbonation weight gain rate: (a) compaction pressure, (b) pressure-holding time, (c) initial moisture content, and (d) carbonation time. Each subplot shows the mean carbonation weight gain rate ($\pm$ standard deviation) plotted against the corresponding process variable, with all other parameters maintained at their baseline values.

The relationship between compaction pressure and carbonation performance is governed by a finely balanced interplay of competing processes, as shown in Figure 2a, which reveals a strong non-monotonic correlation between carbonation weight gain rate (γ) and forming pressure. Quantitatively, γ increases from 6.60% at the lowest pressure (5 MPa) to a maximum of 9.08% at 20 MPa (+37.6% relative), then decays to 7.53% at 25 MPa (−17.1% from peak). This response profile indicates that neither minimal nor excessive consolidation is optimal; rather, an intermediate pressure window (~20 MPa) achieves a maximum through the reconciliation of competing kinetic and transport constraints. At sub-optimal pressures, insufficient interparticle contact and high open porosity reduce the reactive surface area accessible to dissolved CO₂; at supra-optimal pressures, pore closure impairs CO₂ diffusion, thereby shifting the rate-limiting step from surface reaction control to diffusion control.

As the holding time increased from 0.5 min to 2 min (Figure 2b), the carbonation weight gain rate rose from 7.29% to 8.83%. Beyond 2 min, the carbonation weight gain rate slightly decreased, then stabilized. These findings suggest that the molding holding time indirectly influences the carbonation reaction by altering the pore structure, although a distinct “time threshold” exists for this effect [22]. During the short-term pressure-holding phase, the particles gradually rearrange and compact, leading to improved pore connectivity and

structural stability. Once the holding pressure time reaches the critical value, the pore structure stabilizes. Extending the holding pressure time further does not enhance CO₂ mass transfer conditions; instead, it may increase the proportion of local closed pores, which could slightly inhibit the carbonation reaction.

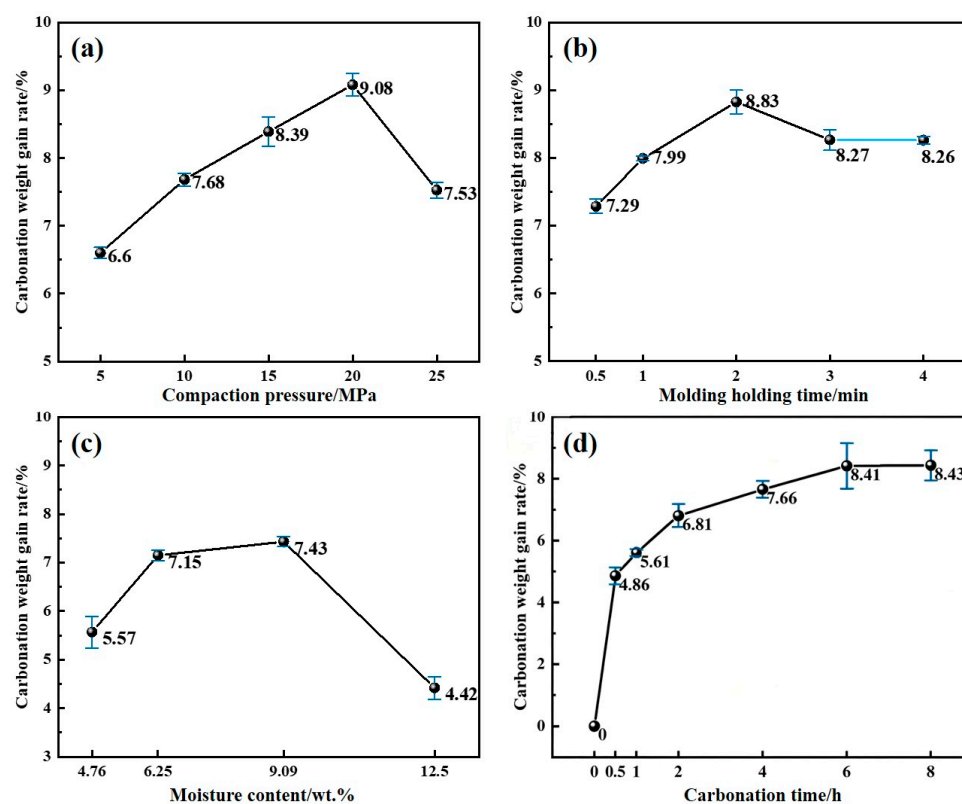


Figure 2. The impact of process parameters on the carbonation weight gain rate. (a) The influence of Compaction pressure: The fixed water content was 9.09 wt.% and the molding pressure holding time was 2 min; (b) The influence of molding holding time: The fixed forming pressure is 20 MPa, and the water content is 9.09 wt.%; (c) The influence of moisture content: Fixed block pressure is 10 MPa, and the holding time is 2 min; (d) The influence of carbonation time: The compression pressure for fixed block molding was 20 MPa, the holding time was 2 min, and the water content was 9.09 wt.%.

At a low moisture content of 4.76 wt.%, the average carbonation weight gain rate is merely 5.57% (Figure 2c). As the moisture content increases to 9.09 wt.%, the rate peaks at 7.43%. However, a further increase to 12.5 wt.% results in a sharp decline to 4.42%. This distinct trend underscores the critical role of water content in dictating the carbonation efficiency of the leached slag briquettes. As indicated by Equations (3)–(8), water functions bidirectionally as both a reactant and a catalytic medium essential for driving the carbonation process. Excessive moisture saturates the internal pores, significantly reducing effective porosity and severely impeding the diffusion and penetration of CO₂ gas [24]. Furthermore, at moisture levels exceeding 12.5 wt.%, a “bleeding” phenomenon occurs under high molding pressures (≥ 15 MPa). This not only deteriorates the structural integrity during molding but also introduces substantial experimental error via mass loss from water seepage; consequently, the molding pressure is strictly maintained at 10 MPa for this single-factor experiment. Conversely, insufficient moisture deprives the system of the adequate liquid phase required for reactive ion dissolution. Therefore, optimizing the water dosage is imperative to satisfy the chemical reaction requirements while maintaining open structural channels for gas diffusion. Ultimately, determining the optimal moisture

content reflects a delicate dynamic balance between the chemical facilitation of water and the physical gas-transport constraints of the porous system.

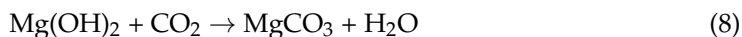
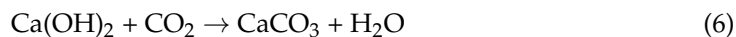
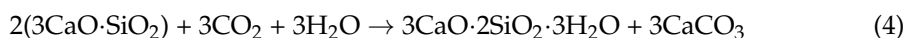
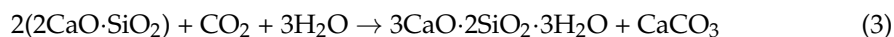


Figure 2d reveals a distinct three-stage kinetic pattern for the carbonation weight gain over time. Initially, during the rapid reaction stage (0–2 h), the average carbonation weight gain rate of the leached slag briquettes surges sharply to 6.81%. Subsequently, the system enters a slow-growth stage (2–6 h), where the weight gain increases from 6.81% to 8.41%, accompanied by a significant deceleration in reaction kinetics. Finally, during the restricted phase (6–8 h), the difference in weight gain between the 6 h and 8 h milestones is negligible, indicating that the surface carbonation reaction of the briquettes has essentially reached completion.

Based on the carbonation weight gain ratio indicator and combined with the conversion results of CO₂ fixation per unit mass of leached steel slag, the optimal process demonstrates a CO₂ absorption capacity of 89.3 mg/g for steel slag. By comparing this with the theoretical carbon sequestration capacity of original BOF steel slag, the carbon sequestration potential of leaching residues is quantified, providing a more intuitive and comparable assessment of carbon absorption performance.

These macroscopic trends are fundamentally governed by the dynamic evolution of the gas–solid reaction interface. During the initial rapid stage (0–2 h), highly reactive components (e.g., free CaO and Ca(OH)₂) on the briquette surface interact directly with dissolved CO₂ without the hindrance of a product layer. At this point, the CO₂ diffusion rate far exceeds the chemical consumption rate, and the exothermic reaction between Ca(OH)₂ and CO₂ during carbonation has an enthalpy change $\Delta H = -113 \text{ kJ}\cdot\text{mol}^{-1}$ (calculated based on each mole of CO₂ involved in the reaction), which raises the temperature and further accelerates the reaction rate [25]. During the subsequent slow-growth phase, the continuous precipitation of CaCO₃ leads to the gradual formation of a dense product barrier. This layer severely restricts inward CO₂ diffusion, shifting the process from a reaction-controlled to a diffusion-controlled regime. As the reaction progresses into the final restricted stage, thermodynamic and kinetic equilibrium is approached. The accumulated CaCO₃ progressively fills the deep pores, drastically reducing pore connectivity and effective porosity. This pore-clogging effect terminates further inward penetration of CO₂, consequently causing the overall reaction rate to plateau [26].

3.2.3. Evolution of the Surface Micro-Morphology of Briquettes

Scanning electron microscopy (SEM) observations reveal that the uncarbonated leached slag briquettes exhibit distinct particle outlines with numerous interparticle pores and micro-cracks, resulting in a relatively rough surface (Figure 3). Following adequate carbonation treatment (6 h), the surface morphology of the briquettes undergoes a significant transformation. An abundance of acicular, clustered, or columnar crystals precipitates on the particle surfaces and along the pore margins, intertwining to form a dense network

structure. These newly formed crystals firmly adhere to the briquette surface and partially fill the interstitial voids, substantially densifying the overall structure.

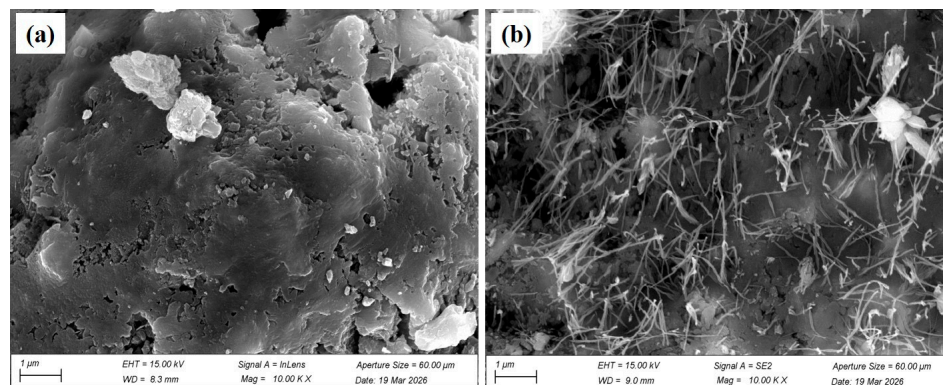


Figure 3. Microscopic morphology of (a) uncarbonated and (b) 6 h-carbonated briquettes.

The morphology of the CaCO_3 crystals formed during steel slag carbonation is not invariable; rather, it is synergistically regulated by multiple factors, including carbonation conditions, initial mineralogical composition, and elemental leaching behavior. Specifically, as reactive mineral phases (such as dicalcium silicate [C_2S] and f-CaO) react with CO_2 , the morphology of the resulting CaCO_3 is dictated by the supersaturation degree at the reaction interface, ion migration pathways, and the competitive precipitation behavior of co-existing elements [27]. For instance, when the RO phase in the steel slag possesses a high $\text{Mg}/(\text{Fe} + \text{Mn})$ ratio, Mg^{2+} is preferentially leached during carbonation. The released Mg^{2+} interferes with the homogeneous nucleation of CaCO_3 , thereby suppressing the growth of thermodynamically stable calcite and instead inducing the crystallization of metastable polymorphs, such as acicular aragonite or vaterite. Furthermore, the crystal structure and surface activity of the parent minerals dictate the nucleation sites and growth orientations of CaCO_3 . Variations in Ca^{2+} dissolution rates and localized ion concentrations among different mineral phases lead to the formation of morphologically distinct product layers. Consequently, the packing density and spatial distribution of aragonite crystals vary significantly with the parent mineral type, ultimately determining the surface micromorphology and structural compactness of the carbonated briquettes.

3.2.4. Effect of Carbonation on the Leaching of Heavy Metal Elements

The BOF slag naturally contains heavy metal elements such as Cr, V, Ni, and Pb. Untreated slag exhibits strong alkalinity, with toxic components like hexavalent chromium readily leaching out through dissolution—a key environmental risk hindering its large-scale application. The pressurized carbonation process stabilizes these metals through a dual mechanism involving chemical transformation and physical encapsulation barriers, significantly reducing leaching risks in applications such as marine artificial reefs and engineering slopes. The mechanisms are as follows: First, chemical stabilization: pH control and valence conversion suppress metal mobility. Carbonation consumes strongly alkaline components like free CaO and $\text{Ca}(\text{OH})_2$, lowering the system pH from 12.5 to around 10; under this weakly alkaline environment, ions such as Cr^{3+} , Ni^{2+} , and Pb^{2+} form highly insoluble hydroxide precipitates, markedly reducing leaching rates. Additionally, reducible components like Fe^{2+} in the slag convert highly toxic Cr^{6+} to less hazardous Cr^{3+} via water-gas reactions during carbonation, forming $\text{Cr}(\text{OH})_3$ for stable storage and eliminating chromium-related environmental hazards at the valence level [28]. Experimental data show that Cr leaching concentrations in carbonated slag decrease by over 88%, far below the limits specified in the “Toxicity Standards for Solid Waste Leaching”.

The second mechanism is the barrier effect of physical encapsulation: it blocks the pathways for heavy metals to interact with the external environment. As the carbonation reaction progresses, the formed CaCO_3 and C-S-H gel continuously fill the pores within steel slag pellets and form a dense, continuous mineral coating on particle surfaces, physically isolating the native mineral phases containing heavy metals from external leaching media such as rainwater and seawater, thereby inhibiting long-term leaching of heavy metals through mass transfer pathways.

Considering anticipated application scenarios such as marine artificial reefs and river-bank stabilization, the surface pH of carbonated steel slag blocks approaches that of natural water environments, with heavy metal leaching concentrations far below environmental standard limits; simultaneously, the dense carbonated layer provides resistance to seawater erosion and long-term effects of wet-dry cycles, ensuring durable heavy metal stabilization that meets environmental safety requirements for ecological applications.

3.3. Evolution of Mechanical and Surface Properties of Briquettes with Carbonation

3.3.1. Mechanical Properties

Based on the optimal molding process (forming pressure of 20 MPa and a holding time of 2 min), the strengthening effect of carbonation treatment on the compressive strength of the briquettes was evaluated, as illustrated in Figure 4. Compared to the physical pressing process alone, the carbonation reaction exerts a fundamentally more decisive regulatory impact on the mechanical properties of the leached slag briquettes. The average compressive strength of the uncarbonated briquettes is merely 93 N. However, after 0.5 h of carbonation, this value surged dramatically to 573 N, eventually peaking at 1262 N when the carbonation duration was extended to 6 h. By 8 h, the strength plateaued at approximately 1260 N, indicating that the surface reaction had essentially reached saturation. Consequently, the carbonation treatment facilitates an order-of-magnitude leap in the compressive strength of the briquettes.

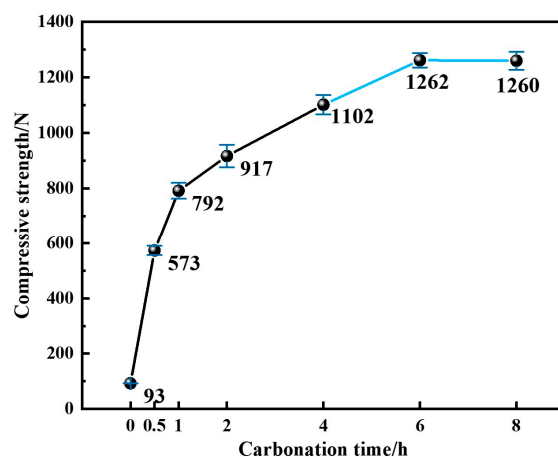


Figure 4. Effect of carbonation on the compressive strength of leached slag briquettes.

This remarkable leap in strength originates from a dual strengthening mechanism induced by the carbonation process [29]. On the one hand, the carbonation reaction substantially consumes the expansive free oxides (i.e., f-CaO and f-MgO) inherently present in the steel slag, converting them into volumetrically stable carbonates [30]. This transformation fundamentally eliminates the internal expansion stress triggered by aqueous hydration, thereby resolving the critical volumetric instability issue that typically hinders the application of steel slag as a structural material. On the other hand, the in situ generated high-strength CaCO_3 crystals, alongside the calcium silicate hydrate (C-S-H) gel formed via the partial conversion of silicate minerals, effectively fill the internal micropores. The

synergistic interaction between the gel phase and the crystalline phase provides a continuous interfacial bonding force, significantly enhancing the overall integrity and compactness of the matrix structure.

Given the experimental conditions of this study—including small sample size ($\Phi 20 \text{ mm} \times 20 \text{ mm}$), long reaction time (6 h), and excellent heat conduction and dissipation in the metal reactor—this demonstrates that the exothermic reaction proceeds slowly and continuously, making it difficult for a significant temperature gradient to form within the sample. Under actual operating conditions, sustained heat dissipation results in a far lower actual temperature rise than predicted, confirming the validity of the isothermal hypothesis for this experimental system.

For the application of marine artificial algal reefs, a compressive strength exceeding 30 MPa is an indispensable prerequisite for withstanding continuous wave impacts and the mechanical loads associated with biological attachment. This requirement aligns with the Technical Specifications for the Construction of Artificial Reefs (T/SCSF 0005-2020), which explicitly mandates that concrete utilized in marine artificial reefs (including algal reefs) must meet or exceed the C30 strength grade [31]. Therefore, carbonation not only transforms the briquettes from a “low-strength and porous” state into a “high-strength and dense” structural entity, but simultaneously optimizes the biocompatibility for microalgal attachment by reducing surface alkalinity. Ultimately, pressurized carbonation serves as a pivotal technological pathway for realizing the high-value ecological application of steel slag.

3.3.2. Surface Chemical Environment

To elucidate the impact of carbonation on the surface chemical environment, the surface pH of the briquettes was monitored across various carbonation durations. As depicted in Figure 5, the evolution of surface pH is intrinsically tied to the degree of carbonation, unfolding in four distinct stages. In its initial uncarbonated state, the leached slag briquette exhibits a highly alkaline surface with a pH of 12.49, reflecting the inherent alkalinity of the steel slag. During the subsequent rapid alkali-reduction stage (0–2 h), the pH undergoes a sharp decrease from 12.49 to 10.44 as the carbonation reaction vigorously progresses. This is followed by a slow alkali-reduction phase (2–6 h), where the pH gradually declines from 10.44 to 10.21 in tandem with the decelerating reaction kinetics. Finally, during the reaction-restricted stage (6–8 h), the surface pH essentially stabilizes.

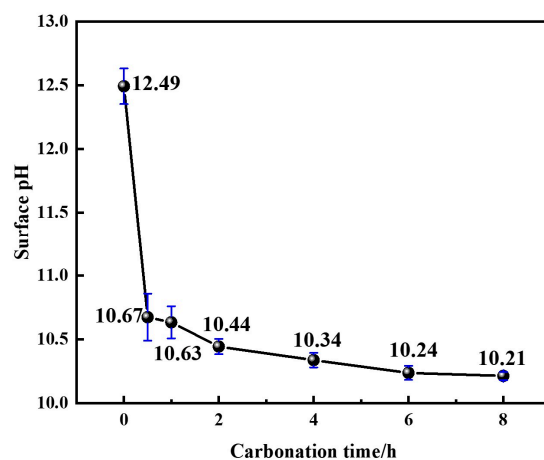


Figure 5. The effect of carbonation on the surface pH of leached slag briquettes.

Mechanistically, this pH moderation is driven by the progressive consumption of strongly alkaline phases (e.g., free CaO and $\text{Ca}(\text{OH})_2$) and their in situ conversion into CaCO_3 precipitates. As the reaction advances, the accumulation of this dense CaCO_3

product layer restricts the inward diffusion of CO_2 , thereby isolating the reaction interface and decelerating the rate of pH decline. The systematic transition of the briquette surface from a highly alkaline state to a moderate, environmentally benign pH fundamentally represents the transformation of steel slag from a “strongly alkaline hazardous waste” into an “eco-friendly material.” This surface neutralization provides a critical theoretical foundation for evaluating its biocompatibility in marine ecological applications, particularly for artificial coral reef substrates.

3.3.3. Surface Roughness

To evaluate the influence of carbonation duration on surface topography, the surface roughness of the leached slag briquettes was systematically assessed over time. As illustrated in Figure 6 and detailed in Table 6, the surface roughness is intrinsically correlated with the degree of carbonation. Mechanistically, carbonation dictates the evolution of surface morphology by modulating the spatial distribution and crystallization behavior of the CaCO_3 precipitates. As the reaction proceeds, highly reactive components (e.g., f-CaO and $\text{Ca}(\text{OH})_2$) are progressively transformed into CaCO_3 [31]. This in situ precipitation continuously infills surface pores, micro-cracks, and topographical asperities, thereby driving a distinct three-stage reduction in surface roughness.

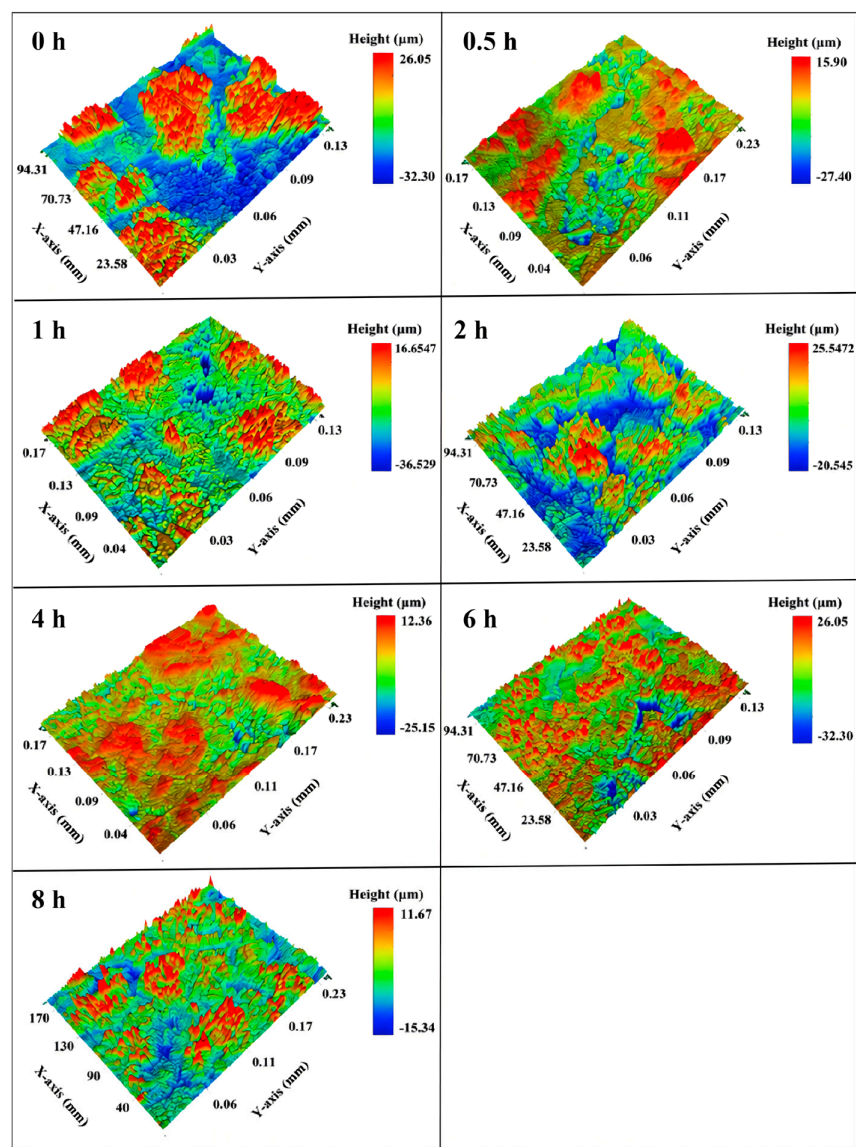


Figure 6. The effect of carbonation on the surface roughness of leached slag briquettes.

Table 6. The effect of carbonation time on the surface roughness of leaching slag blocks.

NO.	Carbonation Time (h)	Ra (μm)	Rq (μm)	Rp (μm)	Rv (μm)	Rt (μm)
1	0	8.58	9.99	26.05	−32.30	58.35
2	0.5	5.98	7.14	25.55	−20.55	46.10
3	1	4.47	6.24	16.65	−36.53	53.18
4	2	3.51	4.84	15.90	−27.40	43.30
5	4	2.90	3.85	12.36	−25.15	37.51
6	6	2.02	3.01	13.69	−25.29	38.98
7	8	1.84	2.42	11.67	−15.34	27.01

During the rapid reaction stage (0–2 h), surface-active substances are swiftly consumed. The data indicate that the most dramatic leveling effect occurs within the first 0.5 h of carbonation. By the end of this 2 h stage, the rapid generation of CaCO_3 effectively masks the original rough structures, causing the average surface roughness to decrease sharply from an initial 8.58 μm to 3.51 μm . Subsequently, the process enters a slow-descent phase (2–6 h), wherein the product layer gradually densifies. As the pore-filling rate decelerates, the average roughness slowly declines to 2.02 μm . Ultimately, during the reaction-restricted stage (6–8 h), the surface pores are almost entirely occluded, and the carbonation reaction approaches completion. A dense, continuous CaCO_3 coating forms on the briquette surface, stabilizing the roughness at approximately 2 μm .

This morphological evolution is intuitively corroborated by the 3D surface profiles (Figure 6). The original uncarbonated briquette surface exhibits numerous micropores and sharp protrusions, whereas extended carbonation (8 h) yields a smooth, integrated surface layer, significantly refining the micro-morphology. The continuous downward trend and subsequent stabilization of the roughness parameters after 6 h align perfectly with the evolutionary patterns observed for both compressive strength and surface pH. This strong correlation suggests that surface morphology serves as a reliable macroscopic indicator of the carbonation progression, providing a theoretical foundation for regulating solid surface properties via reaction time. Furthermore, from an ecological application perspective, this carbonation-induced reduction in roughness not only enhances the structural uniformity of the briquettes but also yields an optimized topographical substrate, which is highly conducive to the adhesion, spreading, and colonization of microalgal spores in subsequent marine environments.

4. Conclusions

This study systematically investigated the carbonation mechanism of leached BOF slag briquettes and evaluated their viability as environmental materials. By integrating orthogonal and single-factor experiments with multi-scale characterizations, this research elucidated the influence of process parameters on carbonation kinetics and the resulting physicochemical properties of the products. The principal conclusions are as follows:

- (1) Both molding pressure and holding time influence carbonation efficiency by regulating pore structure, with the effect of holding time on internal structure being weaker than that of molding pressure, and a critical time threshold exists; beyond this threshold, the pore structure reaches a stable equilibrium, and extending the holding time no longer significantly improves carbonation performance.
- (2) Water exhibits triple functions as a reactant, catalyst, and pore regulator during the carbonation process, demonstrating bidirectional regulatory characteristics; only within the optimal water content range can a balance between chemical reaction rate and gas diffusion capacity be achieved, thereby maximizing carbonation efficiency.

- (3) Carbonation can simultaneously regulate both the mechanical properties and surface physicochemical characteristics of the test specimen: as carbonation time increases, the specimen's compressive strength rises, while its surface pH and roughness decrease; these three trends correspond precisely to the three-stage characteristics of carbonation-induced weight gain, essentially reflecting the macroscopic processes of calcium carbonate formation and pore filling.

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