

Development of Carbonation-Cured Alkali-Activated Slag Masonry Units

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Abstract – This paper investigates the impact of early accelerated carbonation curing on the performance of alkali-activated slag concrete masonry blocks. The study employed crushed dolomitic limestone aggregates, with a binder-to-aggregate ratio of 1:6. A sodium-based alkaline solution, prepared with a sodium silicate to sodium hydroxide ratio of 1.5, was incorporated into the concrete batch at a solution-to-binder ratio of 0.6. The influence of carbonation curing parameters on the performance of concrete masonry blocks was evaluated, including initial ambient curing duration (0, 2, and 4 hours), carbonation duration (4 and 24 hours), and carbonation pressure (1 and 5 bars). These parameters were analyzed in relation to carbon uptake, compressive strength at 1, 7, and 28 days, and water absorption of the concrete masonry blocks. Additionally, a comparative assessment of the global warming indices associated with the production of cementitious and carbonation-cured alkali-activated slag masonry units was developed. The results reveal that carbon uptake and water absorption capacity increased with longer initial and carbonation curing durations and higher carbonation pressures. The compressive strength was either maintained or decreased due to the decalcification of calcium-rich gel produced during the activation reaction by carbonation curing. However, all mixes demonstrated suitability as non-load-bearing blocks, achieving strengths higher than 4.1 MPa after 1 day of batching. The environmental impact analysis indicator a reduction of 79.2% in the production of 1 m³ of carbonation-cured alkali-activated slag concrete in comparison to cement-based counterparts.

Keywords: Masonry blocks; Slag; Alkali-activated materials; Carbon uptake; Compressive strength; Economic impact.

1. Introduction

Concrete masonry units (CMUs), primarily manufactured with ordinary Portland cement, are extensively utilized in the construction and building industry as load-bearing and non-load-bearing elements [1]. In addition, CMUs can serve as dual purpose for the construction of partition walls while providing thermal insulation for fire and heat protection [2]. The American society for testing and materials (ASTM) classifies CMUs based on their compressive strength values. Blocks with a strength greater than 13.8 MPa are designated as load-bearing blocks [3], whereas those with a strength ranging between 4.1 and 13.8 MPa are referred to as non-load-bearing masonry units [4]. According to the Global Concrete Block and Brick Manufacturing Report for 2021 [5], the production of CMUs is projected to experience an annual growth rate of 2.4%. By the end of 2027, it is predicted to reach a production value of 2.3 trillion units, compared to the annual production of 1.9 trillion units in 2020. This surge is attributed to the exponential increase in the global population and the growing demand for expanding urbanization areas worldwide. The reliability of Portland cement for CMUs production raises significant environmental concerns, as it is the primary contributor to the high global potential indices of cement-based building material production [6]. Therefore, there is a pressing need to find sustainable alternatives and solutions to mitigate and decrease the environmental impact resulting from concrete production.

One of the many proposed alternative binders to cement are geopolymers. Geopolymers or alkali-activated building materials have been suggested as a solution to mitigate such detrimental environmental impact of concrete production, as they are cement-free materials primarily produced with calcium and/or alumina-silicate-rich waste and by-products from various industries [7]. Calcium and alumina silicate binders, such as slag, fly ash, perlite, and metakaolin, among others, are activated with sodium or potassium alkaline solutions to produce mortar or concrete. A scientometric review focusing on the use of geopolymer composites in various construction applications revealed a steady annual research production increase from 23.9% to 45.2% between 2003-2013 and 2014-2022 [8]. This rise is attributed to the promising mechanical, fire-

resistant, corrosion-resistant, and bonding properties of geopolymer composites, making them suitable for diverse construction applications [8]–[10].

Carbonation curing has also been introduced as a solution to reduce greenhouse gas emissions [11]. This curing regime captures and stores carbon dioxide within the paste matrix by reacting with calcium-based materials to produce calcite and calcium silicate hydrate gel [12]. The formation of new reaction products and an increase in the volume of existing compounds resulted in the densification of the microstructure of building materials, leading to a reduction in the porosity and an enhancement in the mechanical properties [13]. El-Hassan and Shao [12] developed carbonation-cured limestone cement-based CMUs with a carbon sequestration potential comparable to that of Portland cement-based units. The results showed that longer initial air curing and carbonation durations led to superior carbon sequestration potential, i.e., carbon uptake, and mechanical strength. On the effect of carbonation pressure, the increase from 0.1 to 10 bars resulted in an increase in carbon uptake and an improvement in the durability properties of cement-based concrete [14]. Moreover, when comparing different curing regimes, carbonation curing emerged as a more energy-efficient option than steam curing for concrete masonry units and required less time than water curing [14]. In the case of slag and fly ash-based alkaline activated concrete, it was found that alkali-activated slag concrete exhibited greater potential for absorbing carbon dioxide compared to fly ash-based counterpart. This was attributed to the higher concentration of calcium-carrying compounds in the binder matrix, which formed during the alkaline activation reaction [15]. On the other hand, the adoption of carbonation curing concrete blocks, bricks, and concrete paver blocks industries could result in the global sequestration of approximately 1 billion tons of CO₂ annually [16].

Based on the presented literature, the implementation of carbonation curing could prove highly advantageous in the production of alkali-activated slag CMUs. This study aims to evaluate the carbon sequestration potential and performance of carbonation-cured alkali-activated slag concrete representing CMUs. The effect of various carbonation parameters, including initial ambient curing duration, carbonation curing duration, and carbonation pressure, on said response criteria was assessed.

2. Materials and Methods

2.1. Materials

Ground granulated blast furnace slag, commonly referred to as slag, and crushed limestone aggregates, with a maximum size of 9.5 mm, were employed as binder material and aggregates, respectively. Slag was selected due to its unique composition, which includes a high amount of calcium oxide (CaO) at a percentage of 59.7%. Meanwhile, crushed limestone aggregates were chosen due to their local abundance and superior abrasion and durability performance in comparison to other aggregates like dune and river sand [9], [17].

A sodium-based alkaline solution was produced by blending sodium hydroxide solution (SH) and Grade N sodium silicate solution (SS), maintaining an SS-to-SH ratio of 1.5. The SH solution was initially prepared by dissolving sodium hydroxide pellets in tap water, resulting in a solution with a molarity of 8 M. This specific ratio and molarity were selected to ensure optimal fresh properties behavior, including workability and setting time, which are necessary for the casting of CMUs [18].

2.2. Mix Design and Curing Regime

The alkali-activated slag CMU mixes were produced following the recommendation of a commercial mix having a binder percentage of 13% of the total mass of concrete [18]. As a result, a binder-to-sand ratio of 1:6 for and solution-to-binder ratio of 0.6 were chosen. Such ratios will aid the penetration of carbon dioxide within the concrete matrix. Following materials batching and mixing, the samples underwent curing under various conditions, as outlined in Table 1. Carbonation-cured mixes were labelled as Xa/Yc/Zb, where X, Y, and Z represent the initial ambient curing duration, carbonation curing duration, and carbonation pressure, respectively. The impact of the initial ambient curing duration was investigated by comparing the performance of mixes 0a/4c/1b, 2a/4c/1b, and 4a/4c/1b samples, where it varied among 0, 2, and 4 hours, respectively. The effect of carbonation duration was assessed by analyzing the properties of

the mixes 0a/4c/1b and 0a/24c/1b, where it extended from 4 to 24 hours. Finally, the influence of carbonation pressure was investigated by comparing the behavior of mixes 4a/4c/1b and 4a/4c/5b at respective pressure of 1 and 5 bars.

Table 1: Curing conditions of CMU mixes.

Mix Code	Ambient Curing (hr)	Carbonation Curing (hr)	Carbonation Pressure (bar)
Control	-	-	-
0a/4c/1b	0	4	1
0a/24c/1b	0	24	1
2a/4c/1b	2	4	1
4a/4c/1b	4	4	1
4a/4c/5b	4	4	5

2.3. Sample Production

The alkaline activator solution was prepared a day before concrete production to ensure the complete dissipation of heat when SH and SS were mixed. For the mixing process, dry materials were batched together for a couple of minutes. Subsequently, the alkaline solution was added, and the freshly mixed concrete was blended for an additional minute to ensure thorough homogeneity. The freshly mixed concrete was then placed into 5 cm cubic molds, vibrated for 10 seconds using a vibration table, and subsequently transferred to a carbonation chamber, following the specified regimes detailed in Table 1. Figure 1 illustrates the carbonation setup employed for the curing of concrete mixes representing CMUs.



Fig. 1: Carbonation setup.

2.4. Testing Procedures

The mass gain approach was employed to calculate the CO₂ uptake of carbonation-cured CMU mixes. Equation (1) was used to determine the increase in mass attributed to the sequestration of CO₂ by the binder. The water loss observed within the chamber was collected using tissue paper and incorporated into the equation.

$$\text{CO}_2 \text{ uptake (\%)} = \frac{(\text{Mass after carbonation curing} - \text{Initial mass}) + \text{Water loss}}{\text{Binder mass}} \times 100 \quad (1)$$

The compressive strength was measured after 1, 7, and 28 days of concrete production, following the ASTM C109 standard [19]. The water absorption of the 28-day samples was calculated according to the ASTM C642 procedure [20]. Furthermore, the environmental impact was analyzed using the global warming potential indices (GWP), representing the mass in kilograms of CO₂ emitted during the production of 1 cubic meter of CMUs (kg CO₂ eq/m³). The GWP of carbonation-

cured mixes was determined by subtracting the CO₂ uptake by the binder from the GWP of the mix. Additionally, the GWP of alkali-activated CMUs was compared with that of cement-based counterparts. The GWP indices for the materials used in this study were as follows: 0.0416 kg CO₂ eq./kg for slag, 0.8980 kg CO₂ eq./kg for cement, 0.0052 kg CO₂ eq./kg for crushed limestone, 0.4240 kg CO₂ eq./kg for sodium silicate (SS), and 0.8293 kg CO₂ eq./kg for sodium hydroxide (SH), [6], [21], [22].

3. Results and Discussion

3.1. Carbon Uptake

Figure 2 illustrates the carbon uptake values for CMU mixes cured with different initial ambient curing durations, carbonation curing durations, and pressures. The percentage of carbon uptake based on binder mass ranged between 9.7% and 11.2%. Examining the impact of carbonation duration, the carbon uptake increased from 9.7% to 11.2% as the duration extended from 4 to 24 hours for mixes 0a/4c/1b and 0a/24c/1b, respectively. This increase is attributed to the prolonged contact between CO₂ and concrete samples placed inside the carbonation chamber, enhancing the storage of CO₂ within the calcium-rich binder. Similar findings were reported elsewhere regarding the effect of carbonation duration on carbonation-cured limestone cement-based CMUs [12]. Similarly, an increase in pressure from 1 to 5 bars resulted in an increase in the carbon uptake, i.e., from 9.9% to 11.2%, in mixes 4a/4c/1b and 4a/4c/5b.

As for the impact of the initial ambient curing duration, the carbon uptake was reported to be 9.6%, 9.7%, and 9.9% for initial ambient curing durations of 0, 2, and 4 hours in mixes 0a/4c/1b, 2a/4c/1b, and 4a/4c/1b, respectively. A longer initial ambient curing duration led to higher evaporation of the solution within the concrete matrix, resulting in the formation of pores. These pores facilitated increased penetration of carbon dioxide into the matrix, thereby increasing the carbon sequestration potential of alkali-activated slag CMUs [14].

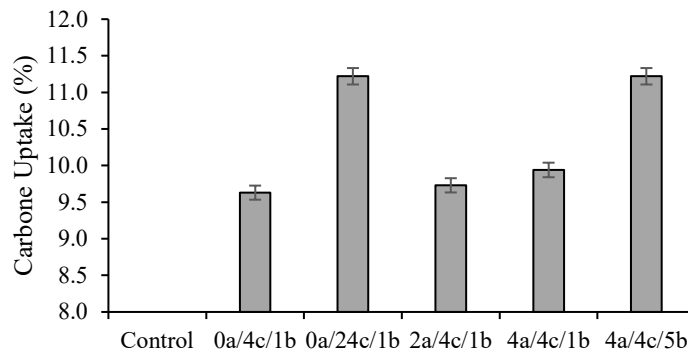


Fig. 2: Carbon uptake of carbonation-cured CMU mixes.

3.2. Compressive Strength

The compressive values of ambient and carbonation-cured alkali-activated slag CMU mixes at 1, 7, and 28 days are illustrated in Fig. 3. The overall trend indicates that the compressive strength of carbonation-cured CMUs either maintained or slightly decreased following accelerated carbonation curing. This reduction is attributed to the reaction between the calcium-rich gel formed during the alkaline activation of slag and CO₂, leading to the formation of calcium carbonate polymorphs [23]. Nevertheless, it is noteworthy that all samples exhibited a strength higher than 4.1 MPa, making them suitable for use as non-load-bearing CMUs [4].

Furthermore, the CMUs mixes demonstrated remarkable strength gain performance by achieving 83 to 95% of their 28-day strength after 1 day of casting. The compressive strength values at 1-, 7-, and 28 days ranged between 8.0-11.9 MPa, 9.2-12.3 MPa, and 9.6-12.5 MPa, respectively. This rapid strength development is owed to the fast and high degree of reaction between calcium-rich binders, i.e., slag, and a sodium-based alkaline solution [24]. Consequently, the carbonation-

cured mixes exhibit great potential for use in the construction industry as non-load-bearing units after only 1-day of production.

The carbonation curing duration and pressure exhibited a similar trend. With prolonged exposure of CO₂ to alkali-activated slag CMU mixes and an increase in carbonation pressure, a slight reduction in compressive strength was noted. For instance, extending the carbonation duration from 4 to 24 hours and elevating the pressure from 1 bar to 5 bars resulted in a reduction of the 28-day strength values from 10.5 to 9.6 MPa and from 11.5 to 10.5 MPa, respectively. This loss in performance is owed to the decalcification of calcium-rich compounds and the formation of calcium carbonate [25].

Concerning the impact of the initial ambient curing duration, the 28-day strength exhibited a notable variation. Indeed, when CMU mixes were placed either directly or 2 hours after casting, the strength increased from 10.5 MPa to 12.4 MPa. However, this strength subsequently decreased to a value of 11.5 MPa when samples were air cured for 4 hours. The observed variation in strength can be associated to several factors. Firstly, the fragile bonding of calcium compounds at the very early stage (0 hours of initial ambient curing) may contribute to this phenomenon. Additionally, the higher amount of evaporated solution with an increase in the initial ambient curing duration from 2 to 4 hours facilitates the penetration of CO₂ into the concrete matrix, consequently enhancing the degree of reaction between slag paste and CO₂. Similar results were found elsewhere in the production of Portland cement-based lightweight CMUs, where a 4-hour initial carbonation curing duration was determined to be optimal, resulting in the highest strength [13].

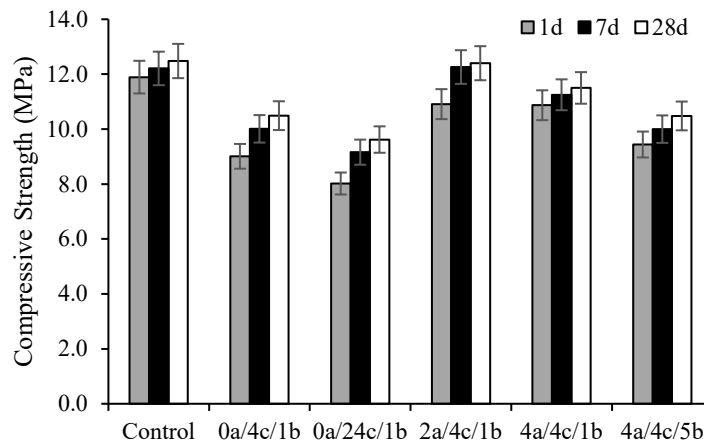


Fig. 3: Compressive strengths of ambient and carbon cured CMUs mixes.

3.3. Water absorption

Carbonation curing demonstrated a positive impact on the water absorption of alkali-activated slag CMU mixes, as presented in Fig. 4. The control samples exhibited the highest value of 9.8%, whereas carbonation-cured counterparts showed water absorption ranging between 7.5% and 9.3%. This decrease in water absorption is attributed to the formation of calcium carbonate within the concrete matrix, leading to a reduction in internal voids. Consistent with previously discussed results, the influence of carbonation curing duration and pressure followed a similar trend. An increase in both parameters resulted in a reduction in water absorption values, owing to longer contact and higher penetration of CO₂ within the matrix, enhancing the reaction with slag paste and expanding the formation of calcium carbonate. X-ray diffractograms on the impact of carbonation curing on alkali-activated material showed an increase in calcium carbonate peak with an increase in the carbonation duration and pressure [18].

Meanwhile, as the initial ambient curing duration increased, a decrease in water absorption was observed, mirroring the trend noted in the carbon uptake analysis. The maximum carbon sequestration by the slag paste led to the densification of the internal structure of concrete through the formation of new calcium carbonate products [13]. Consequently, water absorption values decreased from 9.3% (at 0 hr) to 9.0% (at 2 hr), and further to 8.3% (at 4 hr) of initial ambient curing duration, illustrating a progressive reduction in water absorption with an extended curing period.

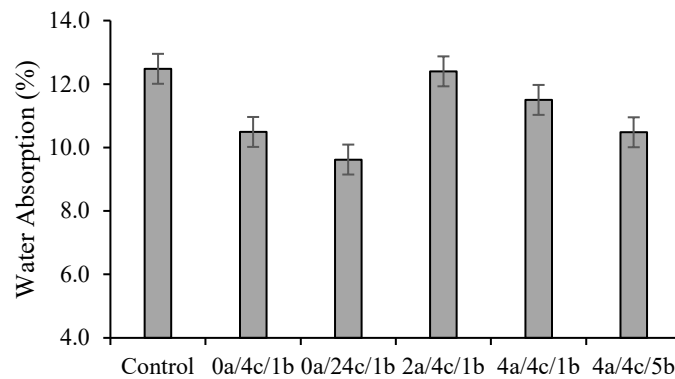


Fig. 4: Water absorption of ambient and carbon cured CMUs mixes.

3.4. Environmental Impact

The environmental impact of producing 1 cubic of various types of CMUs was assessed using GWP indices, as shown in Fig. 5. The GWP indices of two specific types of alkali-activated slag concrete were compared: ambient-cured and carbonation-cured (carbon uptake of 10%, by binder mass). These values were compared with the GWP of CMUs produced using cement as the sole binder. The emission of CO₂ from the production of cement-based concrete mixes was notably high, with a value of 262.3 kg CO₂ eq/m³. However, replacing hydrated cement with alkali-activated slag in the production of CMUs led to a 68% decrease in the GWP, i.e., from 262.3 to 85.2 kg CO₂ eq/m³. Furthermore, the adoption of carbonation curing resulted in an additional GWP reduction of about 36%. Specifically, the GWP of the carbonation-cured alkali-activated slag CMU mixes (with a carbon uptake of 10%) was calculated to be 54.7 kg CO₂ eq/m³. This highlights the environmental benefits of employing carbonation curing in the production of CMUs while also replacing cement with alkali-activated slag as binding material.

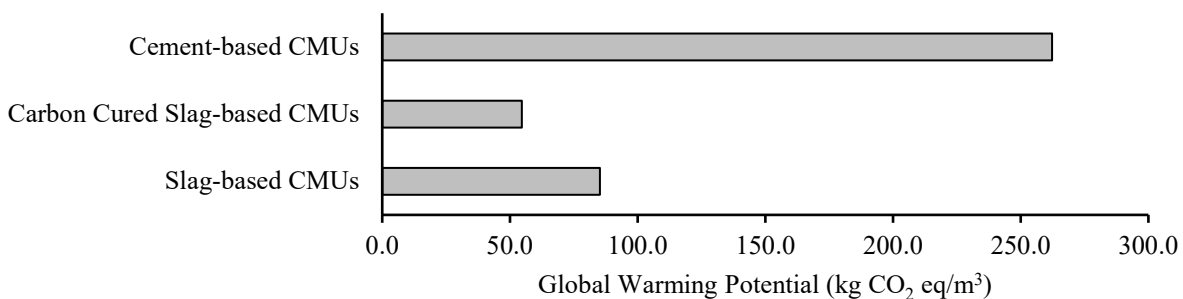


Fig. 5: GWP indices of different types of CMUs.

4. Conclusions

This study evaluated the effect of carbonation curing parameters, such as initial ambient curing duration, carbonation curing duration, and carbonation pressure, on the carbon sequestration capacity, compressive strength, and water absorption of alkali-activated slag concrete masonry units (CMUs). In addition, an environmental impact analysis was performed to compare the amount of carbon dioxide emitted from the production of 1 m³ of cement-based CMUs to that of ambient- and carbonation-cured alkali-activated slag counterparts. The following conclusions summarize the findings of this work:

- Superior carbon uptake for carbonation-cured alkali-activated slag CMU mixes was attained through extended initial ambient curing. This is attributed to the gradual evaporation of the solution, facilitating the penetration of CO₂ and

increasing the carbon uptake. Furthermore, augmenting the carbonation curing duration and pressure resulted in enhanced carbon sequestration, owing to the longer contact between the slag paste and CO₂ under these conditions.

- With prolonged carbonation time and increased pressure, the compressive strength of CMU mixes decreased due to the decalcification of calcium-based rich gels. An optimal initial carbonation duration of 2 hours was identified, resulting in the least impact on the compressive strength. Nevertheless, all mixtures surpassed the strength threshold of 4.1 MPa, stipulated by ASTM C129 for the production of non-load-bearing CMUs.
- Carbonation curing had a positive impact on water absorption, aligning with the trends of carbon uptake and compressive strength.
- Carbonation-cured alkali-activated slag CMUs had a 79% lower environmental footprint than cement-based counterparts. This is owed to the replacement of cement by alkali-activated slag and utilization of carbonation curing.

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