Effects of CO₂ Curing on Alkali-Activated Slag Paste Cured in Different Curing Conditions

Yubin Jun, Seong Ho Han, Tae Yong Shin and Jae Hong Kim *

Department of Civil and Environmental Engineering, Korea Advanced Institute of Science and Technology, Daejeon 34141, Korea; ssjun97@gmail.com (Y.J.); ha8890@kaist.ac.kr (S.H.H.); tyshin@kaist.ac.kr (T.Y.S.)

* Correspondence: jae.kim@kaist.ac.kr; Tel.: +82-42-350-3639

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Abstract: The effect of CO₂ curing on alkali-activated slag paste activated by a mixture of sodium hydroxide and sodium silicate solutions is reported in this paper. The paste samples after demolding were cured in three different curing environments as follows: (1) environmental chamber maintained at 85% relative humidity (RH) and 25 °C; (2) 3-bar CO₂ pressure vessel; and (3) CO₂ chamber maintained at 20% CO₂ concentration, 70% RH and 25 °C. The hardened samples were then subjected to compressive strength measurement, X-ray diffraction analysis, and thermogravimetry. All curing conditions used in this study were beneficial for the strength development of the alkali-activated slag paste samples. Among the curing environments, the 20% CO₂ chamber was the most effective on compressive strength development; this is attributed to the simultaneous supply of moisture and CO₂ within the chamber. The results of X-ray diffraction and thermogravimetry show that the alkali-activated slag cured in the 20% CO₂ chamber received a higher amount of calcium silicate hydrate (C-S-H), while calcite formed at an early age was consumed with time. C-S-H was formed by associating the calcite generated by CO₂ curing with the silica gel dissolved from alkali-activated slag.

Keywords: alkali-activated slag; CO₂ curing; carbonation curing; calcite; vaterite; C-S-H

1. Introduction

Greenhouse gas emission, including CO₂, is one of the causes of global warming, which increases the frequency and extent of natural disaster in the world. Cement clinker production for Portland cement contributes 4% of global CO₂ emission according to the statistics of 2017 [1], which is taken seriously in the construction industry. As environmental problems of CO₂ emission becomes serious worldwide, there have been considerable efforts to find a way to reduce the CO₂ level in the atmosphere. The construction industry has also tried to reduce greenhouse gas emissions.

The use of CO₂ for accelerated curing of cement-based materials is one of the active responses to decrease CO₂ concentration in the atmosphere. Calcium silicates such as alite and belite in Portland cement are spontaneously carbonated, which mainly results in the formation of calcium carbonate (CaCO₃). Cement-based materials subjected to CO₂ curing at an early age show a rapid development of their strength because in the process of CO₂ curing, CaCO₃ precipitates in pores of cast mortar and concrete. Densifying the pore refines their microstructure, including the cement paste matrix and the interfacial transition zone, which results in a higher strength at a rather early age [2,3]. However, previous studies adopted a very low water-to-cement ratio (w/cm) ranging from 0.06 to 0.28 [4], 0.11 to 0.25 [2], and 0.125 [5] to 0.18 [3]. This limits the application of CO₂ curing in practice [3].

On the other hand, the use of alternative cement replacing Portland cement expectedly contributes to the decrease in CO₂ emission. Calcium sulfoaluminate cement has merit due to the low temperature requirement for its calcination process; an approximately 100–200 °C decrease in the calcination temperature compared to the calcination for Portland cement contributes to lower CO₂ emissions [6,7].
In addition, alkali-activated binders, including geopolymers synthesized with industrial byproducts such as blast-furnace slag and fly ash, are one of the promising alternatives [8–11]. It had been reported that they possess excellent durability [11]. The blast-furnace slag utilized as a raw material also contains a large amount of calcium silicates. It is susceptible to carbonation while its hydration is latent, requiring an alkaline activator. The alkaline activator enables calcium to dissolve from slag particles, and the calcium participates in the formation of calcium silicate hydrate (C-S-H) gels contributing to the development of strength [12,13]. In addition, it is expected that the dissolved calcium could participate in the formation of CaCO3 in the process of CO2 curing.

From the viewpoint of recycling industrial byproducts as well as a reduction in CO2 emission, this paper conducted research on CO2 curing for alkali-activated slag. The setting of a slag paste was obtained by the alkali activation, and the cast samples were subjected to CO2 curing. The CO2 curing was carried out in a pressure vessel of approximately 300 kPa of CO2 pressure or in a chamber maintained at a constant 20% CO2 concentration. The compressive strengths of the samples were studied according to the CO2 curing condition. X-ray diffraction (XRD) and thermogravimetric (TG) analyses on the samples quantitatively analyzed the quantity and crystallization of the reaction products.

2. Experiments

2.1. Materials

Ground-granulated blast-furnace slag (GGBFS) used in this study was produced by H Steel Company in Dangjin, Korea. Table 1 reports the chemical composition of GGBFS by X-ray fluorescence analysis. The GGBFS consists of lime and silica to a high extent. Figure 1 presents the XRD pattern of the GGBFS, which corresponds to the general pattern of blast-furnace slag [14–16]. Sodium hydroxide and sodium silicate were used as an alkaline activator. Sodium hydroxide pellets (NaOH; reagent grade with a molar ratio of CaO/SiO2 of 2.8) were acquired from S company in Pyeongtaek, Korea.

Table 1. Chemical composition (oxides in wt.%) of ground-granulated blast-furnace slag (GGBFS).

|     | CaO   | SiO2  | Al2O3 | Na2O  | K2O   | MgO   | MnO   | TiO2  | SO3  | P2O5  | Fe2O3 | Others |
|-----|-------|-------|-------|-------|-------|-------|-------|-------|------|-------|-------|--------|
|     | 47.97 | 30.76 | 13.26 | 0.23  | 0.54  | 3.06  | 0.52  | 0.87  | 1.81 | 0.01  | 0.61  | 0.25   |

Note: Others include BaO, ZrO2, V2O5, SrO, and Y2O3.

![Figure 1. X-ray diffraction (XRD) patterns of raw GGBFS.](image-url)
2.2. Experimental Design and Sample Preparations

The mixture proportions of samples are shown in Table 2. In this study, alkali activators (i.e., activator type and activator concentration) were chosen to harden samples within 1 h for the given GGBFS. The alkaline activator was prepared by blending 5 M NaOH solution with liquid Na$_2$SiO$_3$. The blend ratio of the NaOH solution to the liquid Na$_2$SiO$_3$ was 1.0 by mass. The 5 M NaOH solution was prepared by dissolving NaOH pellets in deionized water. The weight ratio of the alkali activator (5M NaOH + Na$_2$SiO$_3$) to the binder (GGBFS) was 0.4.

Table 2. Mix proportions of samples.

| Binder (g) | Activator (g) | Activator/Binder |
|------------|---------------|-----------------|
| GGBFS 2400 | 5M NaOH 480   | Liquid Na$_2$SiO$_3$ 480 | 0.4 |

After 4 min of mixing with a planetary mixer, the fresh paste was cast in 25 mm cube molds. Each sample was taken from a mold after 1 h of pre-curing in an environmental chamber at 85% relative humidity (RH) and 25 °C. Table 3 summarizes the designed curing conditions including the control of conventional humid curing (85% RH and 25 °C). The first set of samples (denoted with CO2P-##) was subjected to 99.9% purified CO$_2$ in a pressure vessel, where the initial pressure was set between 3 and 4 bar. The demolded samples were placed in the pressure vessel and then the pressure vessel was vacuumed before injecting the CO$_2$. The 3 bar CO$_2$ curing continued for 3 h, 23 h or 167 h, and additional CO$_2$ was injected in the middle of the 167 h curing. Pressure loss in the pressure vessel was monitored using a pressure digital gauge (PDR1000; Pressure Development of Korea Co., Daejeon, Korea). The sampling rate for the pressure measurement was 1 record per second. A total curing time of 4 h, 24 h, and 7 days (168 h) included 1 h of pre-curing. After the 3 bar CO$_2$ curing, the samples were placed in the environmental chamber (85% RH and 25 °C) for further hydration. The samples cured for 3 h, 23 h, and 167 h in the 3 bar CO$_2$ pressure vessel were labelled CO2P-T1, CO2P-T2, and CO2P-T3, respectively. The other sample, labeled CO2-HC, was cured in a CO$_2$ chamber controlled at 20% CO$_2$ concentration, 70% RH and 25 °C until its strength measurement.

Table 3. Curing conditions of samples.

| Sample Label | Curing time |
|--------------|-------------|
|              | 1 h         | 3 h         | 20 h        | 6 Days | 35 Days |
| Control      | Chamber at 25 °C and 85% RH | Chamber at 25 °C and 85% RH | Chamber at 25 °C and 85% RH | Chamber at 25 °C and 85% RH |
| CO2P-T1      | Chamber at 25 °C and 85% RH | Chamber at 25 °C and 85% RH | 3-bar CO$_2$ pressure vessel | Chamber at 25 °C and 85% RH |
| CO2P-T2      | Chamber at 25 °C and 85% RH | Chamber at 25 °C and 85% RH | 3-bar CO$_2$ pressure vessel | Chamber at 25 °C and 85% RH |
| CO2P-T3      | Chamber at 25 °C and 85% RH | Chamber at 25 °C and 85% RH | 3-bar CO$_2$ pressure vessel | Chamber at 25 °C and 85% RH |
| CO2-HC       | Chamber at 25 °C and 85% RH | Chamber at 25 °C and 85% RH | 3-bar CO$_2$ pressure vessel | Chamber at 25 °C and 85% RH |

Note. Demolding after 1 h pre-curing; Strength, XRD, and TG tests at 4 h: Control, CO2P-T1, and CO2-HC samples; Strength, XRD, and TG tests at 24 h: Control, CO2P-T1, CO2P-T2, and CO2-HC samples; Strength, XRD, and TG tests at 7 days: Control, CO2P-T1, CO2P-T2, CO2P-T3, and CO2-HC samples; Strength test at 42 days: Control, CO2P-T1, CO2P-T2, CO2P-T3, and CO2-HC samples.

The compressive strengths of the samples were measured at the age of 4 h, 24 h, 7 days, and 42 days after the mixing (Table 3), where an average of the strengths of four replicated specimens was reported. The strength development of alkali-activated slag paste was faster than normal cement concrete. Approximately 90% of the strength at 42 days was obtained even with a 7-day old sample. (see Table 4). This study therefore conducted a detailed investigation until 7 days for CO$_2$ curing. After each measurement of compressive strength at the age of 4 h, 24 h, and 7 days, fractured specimens were
finely powdered and then immersed in isopropanol to stop their hydration. The XRD and TG analyses were applied to the powdered samples after vacuum drying. The XRD and TG were performed for the phase analyses of the reaction products formed in samples. The XRD measurement was carried out using a high-power X-ray diffractometer (Rigaku Corp., Tokyo, Japan) with an incident beam of Cu Kα radiation for a 2θ scanning range of 5–60°. The XRD patterns of samples were analyzed with the International Center for Diffraction Data (ICDD) PDF-2 database [17] and the Inorganic Crystal Structure Database (ICSD) [18]. The TG measurement was performed in nitrogen gas at a heating rate of 10 °C/min from room temperature to 950 °C using an SDT Q600 thermal analyzer (TA Instruments, Inc., New Castle, Delaware, USA).

| Sample Label | Compressive Strength (Standard Deviation), MPa |
|--------------|-----------------------------------------------|
|              | 4 h    | 24 h    | 7 Days | 42 Days |
| Control      | 14.78 (1.33) | 65.56 (1.88) | 85.77 (0.88) | 108.83 (3.52) |
| CO2P-T1      | 18.27 (1.1) | 55.72 (1.14) | 80.62 (2.99) | 85.6 (1.62) |
| CO2P-T2      | -      | 53.47 (2.09) | 69.11 (2.6) | 74.69 (1.9) |
| CO2P-T3      | -      | -      | 61.96 (0.48) | 73.87 (2.57) |
| CO2-HC       | 24.81 (1.96) | 81.77 (2.86) | 111.89 (1.88) | 121.98 (1.43) |

3. Results and Discussion

CO2 curing reportedly accelerates the strength development of Portland cement-based materials [5,19]. Alkali-activated slag pastes were cured in various CO2 curing environments, and Table 4 reports the effect on their compressive strength. Figure 2 directly compares the effect of CO2 curing at the moment when curing finished. The time shown in Figure 2 includes the 1 h pre-curing required to have acceptable demolding in common (Table 3). The effect of CO2 curing for 3 h was compared in Figure 2a: the sample cured in the 20% CO2 concentration chamber (CO2-HC) showed the highest strength, followed by the sample cured in the 3 bar CO2 pressure vessel (CO2P-T1). Both CO2 curing conditions provide a higher strength than the control curing environment. However, a longer period of CO2 curing had a negative effect on strength. As seen in Figure 2b, the samples cured in the 3 bar CO2 pressure vessel (CO2P-T2) for 23 h gave a lower compressive strength than the control sample. In contrast, the samples cured in the 20% CO2 concentration chamber still showed a higher strength than the control. The trend on the strengths at 168 h (CO2P-T3 for 167 h) was the same as CO2P-T2 for 23 h, as shown in Table 4.

Figure 2. Compressive strengths at 4 h (a) and 24 h (b).
Here it should be noted that CO$_2$ was sufficiently supplied for CO$_2$ curing. Figure 3 shows the measured pressure loss in the pressure vessel during CO$_2$ curing. The CO$_2$ pressure decreased over time while the initial pressure was controlled between 3 and 4 bar as previously described. This implies that the CO$_2$ was consumed with time. About 24 h after injecting CO$_2$, the CO$_2$ pressure dropped to a level of approximately 0.3 bar. In this study, CO$_2$ was reinjected so that the CO$_2$ pressure did not drop to 0 bar during CO$_2$ curing. For the CO2P-T3 case (CO$_2$ curing for 167 h), CO$_2$ was reinjected one time during the 167 h of CO$_2$ curing (Figure 3).

![Figure 3. Pressure loss in the pressure vessel during CO$_2$ curing.](image)

A successive moisture curing after CO$_2$ curing reportedly causes the hydration of calcium silicates, which results in further increases in the strength of Portland cement-based materials [2,20,21]. Figure 4 presents the compressive strength development of the samples taken after further moisture curing following CO$_2$ curing. In the case of the CO2-HC samples, since they were cured in a 20% CO$_2$ concentration chamber maintained at 70% RH, we can presume that the samples were subjected to moisture curing. It achieved the highest strength of 111.89 MPa at seven days while the control alkali-activated slag reached 85.77 MPa at seven days. Further hydration on the samples cured in the 3 bar CO$_2$ pressure vessel (CO2P-T1) obviously provided strength development, but its increase was lower than the control. The strength of the control sample exceeded that of CO2P-T1 at 24 h and seven days while the 4 h strength of the control sample was lower than that right after CO$_2$ curing.

![Figure 4. Compressive strength developments of paste samples cured under different curing conditions with curing time.](image)
It was again confirmed that the long period of CO$_2$ curing (CO2P-T2 for 23 h and CO2P-T3 for 167 h) in the 3 bar CO$_2$ pressure vessel was not effective. Their compressive strengths were lower than that of the control sample. The effect of further moisture curing on the CO2P-T2 sample was also not effective.

The XRD patterns of all samples are shown in Figure 5. Earlier studies have shown that calcite, C-S-H, hydrotalcite, and calcium alumina silicate hydrate (C-A-S-H) were present in NaOH/Na$_2$SiO$_3$-activated slag [12,22,23]. It was also reported that the reaction products of alkali-activated slag depend on the composition of slag and the activator. Nevertheless, C-S-H is the main reaction product in alkali-activated slag [12,13] regardless of the activator used. As shown in Figure 5a, the main reaction products of the control sample were calcite and C-S-H. Figure 5b shows the detailed XRD pattern, where the strongest peaks of calcite (29.395° 2θ) [18] and C-S-H (29.356° 2θ) [17] were overlapped at 29° to 30° 2θ. The formation of C-S-H(I), which is a more crystalline form of C-S-H [24], was additionally found at 24 h and seven days.

CO$_2$ curing for the alkali-activated slag pastes (Figure 5c–f) also provided calcite and C-S-H as their main products. However, C-S-H(I) was not present in the CO$_2$-cured samples until seven days. In addition, vaterite, a metastable allotropic form of CaCO$_3$ [25–27], was found in the samples right after the 3 bar CO$_2$ curing (CO2P-T2 at 24 h and CO2P-T3 at seven days). The vaterite was not found in CO2P-T1 (the 3 bar CO$_2$ curing for 3 h) and CO2-HC (the continuous CO$_2$ curing in the 20% CO$_2$ chamber).

TG and derivative TG (DTG) curves of the alkali-activated slag pastes before and after CO$_2$ curing are compared in Figure 6. The first weight loss below 200 °C, clearly identified in the DTG curves, represented the loss of the combined water due to dehydration of C-S-H (50–200 °C [28]) and C-S-H(I) (90–110 °C [13]). The weight loss in the range of 600–700 °C indicated the presence of calcite [29,30]. The amount of calcite in each sample could be roughly quantified on the basis of the weight loss from 600–700 °C [29]. The calcite concentrations at all ages of the samples are tabulated in Table 5.

| Sample Label | Calcite Concentration |
|--------------|-----------------------|
|              | 4 h       | 24 h       | 7 Days   |
| Control      | 1.33%     | 1.51%     | 2.09%    |
| CO2P-T1      | 1.43%     | 2.90%     | 4.07%    |
| CO2P-T2      | -         | 3.64%     | 3.93%    |
| CO2P-T3      | -         | -         | 2.76%    |
| CO2-HC       | 8.37%     | 6.96%     | 2.27%    |

Table 5. Calcite concentrations of the samples.
Figure 5. Cont.
The extent of C-S-H in the control sample obviously increased with conventional humidity curing (see Figure 6a). Although the weight loss of C-S-H and C-S-H(I) overlaps, it can be shown that C-S-H increased with curing time because the C-S-H(I) peak in the XRD result was almost the same between 24 h and 7 days. This observation can also be identified in Figure 5b. In addition, the DTG peak of calcite slightly increased with curing time. The calcite concentrations were evaluated at 1.33%, 1.51%, and 2.09% at 4 h, 24 h and 7 day, respectively. It was reported that C-S-H in alkali-activated slag paste is also carbonated even in conventional humidity curing [31]. As a result, the formation of C-S-H and C-S-H(I) contributes to the strength of alkali-activated binders [29,32], and the formed calcite is helpful in improving the early age strength even though the concentration is as low as 2.8–4.6% [29]. The strength development of the control sample, shown in Figure 4, also supports the correlation. Furthermore, C-S-H is considered as the major reaction product contributing to strength development because the peak intensity of C-S-H(I) in the XRD pattern was relatively small and the growth rate of calcite was low, as reported in Table 5.
In addition, the DTG carbonation of the inside of a cast sample needs CO$_2$ was cured for a sufficient time (167 h) that CO$_2$ diffusion is expected inside. Nevertheless, it gave a low calcite concentration (2.76%). Therefore, the CO$_2$ diffusion related to air-filled porosity cannot be considered as a critical factor.

Successive hydration of the sample after CO$_2$ curing contributed to a higher amount of calcite as shown in Figure 6b. The calcite concentrations in CO2P-T1 increased over time: 1.43%, 2.90%, and 4.07% as reported in Table 5. The sample had a relatively higher degree of carbonation than

![Figure 6. TG and derivative TG (DTG) curves of (a) control, (b) CO2P-T1, (c) CO2P-T2, and (d) CO2-HC samples with curing time.](image-url)
the control sample even though the calcite concentration was still low (less than 5%). However, as shown in Figure 4, the strength of the CO2P-T1 sample was less than that of the control sample at all ages. The small amount of calcite formation by CO2 curing was negative on the strength of alkali-activated slag.

The 20% CO2 concentration curing provided a different trend of calcite formulation as shown in Figure 6d and Table 5. First of all, calcite formation in the short period (3 h) of CO2 curing was substantial: 8.37% at 4 h. Compared with the other samples, the high level of calcite concentration is attributed to the simultaneous presence of moisture and CO2 in the chamber. The RH was also controlled at 70%. The CO2 dissolution is more active with the neutral moisture supplied in the CO2 chamber. The highest concentration of calcite was confirmed at 4 h and then it decreased with curing time. It is worth noting that C-S-H formation kept increasing with curing time. During the hydration of alkali-activated slag paste, Ca as well as Si dissolved first in an alkaline environment and then C-S-H, including the other products that entered a solid phase in the paste. Ca2+ dissolved from the blast-furnace slag is preferably consumed for the calcite formation at an early age. The calcite formation is suppressed when the Ca dissolution in an alkaline environment exceeds the CO2 dissolution in the limited amount of moisture. The calcite is then consumed to form C-S-H with silica gel released from the blast-furnace slag: SiO2·xH2O + yCaCO3 + H2O ↔ yCaO·SiO2·xH2O + H2CO3, as reported in [31]. The major reaction product of C-S-H was dominated in the CO2-HC sample, which results in the highest strength among the samples considered in this study.

On the other hand, in the case of the CO2P-T2 and CO2P-T3 samples, weight loss in the range of 520–580 °C was identified and it was related to the decomposition of vaterite [27]. The difference in the strength development with time in the sample (Figure 4) seems to be due to the formation of vaterite (metastable CaCO3). Among the samples cured under CO2 pressure, the samples that were cured for 23 h or 167 h showed the presence of vaterite, except for the sample that was CO2-cured for 4 h. This probably indicates that the CO2 curing time affects the polymorphs of CaCO3. As shown in Figure 6c, further humidity curing decreased the DTG peak of vaterite at seven days and increased the peak of calcite. Vaterite can be easily recrystallized to calcite when exposed to water [27].

The CO2-HC sample at 24 h also showed the weight loss in the range of 450–550 °C. This weight loss is likely due to the decomposition of amorphous calcium carbonate (CaCO3·xH2O [36]) rather than vaterite, which is because the peaks for vaterite was not detected in the XRD result as shown in Figure 5f. Amorphous CaCO3 was reportedly decomposed at temperatures between 245 °C and 645 °C [37].

4. Conclusions

CO2 curing for alkali-activated slag paste is promising in the view of reducing CO2 emission in the construction industry. In this study, blast-furnace slag is activated with 5 M NaOH solution and liquid Na2SiO3. The alkali-activated slag paste cured in a 20% CO2 concentration chamber (70% RH, and 25 °C) shows a higher compressive strength than the control samples cured at 85% RH and 25 °C. A higher amount of calcite was confirmed in the CO2 cured samples via XRD and TG analyses. The simultaneous supply of water vapor and CO2 in the chamber contributes to the CO2 dissolution, which results in the initial formation of substantial calcite (at 4 h). Continuous CO2 curing allows us to generate more C-S-H by the hydration of the calcite and silica gel dissolved from blast-furnace slag. As a result, the CO2-cured samples show a decrease in calcite concentration while the amount of C-S-H increases with curing time. However, the strengths of alkali-activated slag cured in a 3 bar CO2 pressure vessel were lower than the control samples. CO2 is hardly dissolved at the high pH of the pore solution of the alkali-activated slag, and a lower amount of calcite is formed even after CO2 curing in the pressure vessel. Limiting the supply of moisture in the pressure vessel prohibits the hydration of alkali-activated slag as well as CO2 dissolution. The strength development of the alkali-activated slag cured in the CO2 pressure vessel is therefore lower than the control samples cured.
in a conventional humid environment. This study finally concludes that CO$_2$ curing at a constant CO$_2$ concentration was more effective on alkali-activated slag paste than in a 3 bar-CO$_2$ pressure vessel.

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