

Research Article

Mechanical Properties of Na_2CO_3 -Activated High-Volume GGBFS Cement Paste

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Received 30 January 2018; Revised 10 April 2018; Accepted 20 May 2018; Published 25 June 2018

Academic Editor: Flora Faleschini

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The use of Na_2CO_3 to improve the mechanical properties of high-volume slag cement (HVSC) is experimentally investigated in this study. Ordinary Portland cement (OPC) was replaced with 50, 60, 70, 80, and 90% ground-granulated blast-furnace slag (GGBFS) by weight. Na_2CO_3 was added at 0, 1, 2, 3, 4, and 5 wt.% of HVSC (OPC + GGBFS). The compressive strength, water absorption, ultrasonic pulse velocity, dry shrinkage, and X-ray diffraction spectra of the Na_2CO_3 -activated HVSC pastes were analyzed. The results indicate that Na_2CO_3 was effective for improving the strength of HVSC samples at both early and later ages. There was a trend of increasing HVSC sample strength with increasing Na_2CO_3 content. The 5% Na_2CO_3 -activated HVSC (50% OPC + 50% GGBFS) paste had the best combination of early to later-age strength development and exhibited the highest UPV and the lowest water absorption among the Na_2CO_3 -activated HVSC samples at later age.

1. Introduction

Ground-granulated blast-furnace slag (GGBFS) is a by-product of steel manufacturing that has been used as a mineral admixture in concrete for many years. Because this material is cheaper than Portland cement, its incorporation as a replacement for Portland cement in concrete mixtures provides both economic and environmental benefits [1, 2].

Cement containing high volumes of GGBFS is referred to as high-volume slag cement (HVSC) and has been widely studied for its role in industrial by-product recycling and substantial reduction in Portland cement usage. Several studies have noted that the use of HVSC material can provide benefits [3–8] including reduced hydration heat and improved sulfate resistance, chloride resistance, durability, and corrosion resistance. However, there are still problems with the mechanical properties of HVSC [7, 9]. For instance, HVSC results in lower early strength, higher drying shrinkage, and lower carbonation resistance. In order to safely use HVSC in concrete production, their drawbacks should be improved. Some studies have indicated that the low early

strength of HVSC can be overcome by adding alkali activators (e.g., sodium hydroxide, sodium silicate, or sodium sulfate) that are typically used for activation of GGBFS. Singh et al. [10] studied the effect of 4 wt.% Na_2SO_4 on HVSC and reported that addition of the alkali activator to GGBFS-blended Portland cement results in an acceleration of the hydration reaction due to the activation of GGBFS. The activation of GGBFS by the alkali leads to formation of an additional amount of calcium silicate hydrate (C–S–H), which increases the compressive strength. Sajedi and Razak [11] studied the effect of chemical activators (NaOH, KOH, and Na_2SiO_3) on the early strength of HVSC mortars. They mentioned that Na_2SiO_3 gives the highest strength value, whereas NaOH results in the lowest strength. Wu et al. [12] reported the effect of additives (Na_2SO_4 , calcium aluminate cement, and potassium aluminum sulfate) on the compressive strength of HVSC mortars. They noted that the early-age strength of mortar with additives is higher than that without additives and proposed that increasing the amount of additive could result in an increase in the early-age strength. Acevedo-Martinez et al. [13] studied the strength development and

TABLE 1: Chemical composition of OPC and GGBFS (wt.%).

	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	TiO ₂	MnO	SO ₃	K ₂ O
OPC	21.32	5.61	3.12	61.72	3.94	—	—	2.51	0.79
GGBFS	35.30	12.58	0.79	41.30	3.19	0.70	0.22	4.75	0.63

microstructures of HVSC activated with different concentrations of Na₂O using water glass (Na₂SiO₃). The results indicated that increasing the percentage of Na₂O produced denser and more compact matrices of hydration products in HVSC, which was reflected in improved strength. Previous studies have explored the use of sodium carbonate (Na₂CO₃) source for activating GGBFS [14–17]. Abdalqader et al. [17] reported the effect on the compressive strength of using Na₂CO₃ for the activation of fly ash and slag blends. They noted that the strength of Na₂CO₃-activated fly ash-slag blends increased with curing time and Na₂CO₃ content, and the strength at early ages depended on the Na₂CO₃ content. Thus, it can be expected that Na₂CO₃ could play a role in the early-age strength development of HVSC. However, research on the early-age strength of HVSC with Na₂CO₃ as the main activator is limited in comparison with research on HVSC focused on Na₂SiO₃, Na₂SO₄, and NaOH activators.

The primary objective of this study was to carry out an experimental investigation to explore the effect of Na₂CO₃ for improving early strength and other mechanical properties of pastes with high volumes of slag. The compressive strength, water absorption, ultrasonic pulse velocity, dry shrinkage, and X-ray diffraction of HVSC pastes activated with varying Na₂CO₃ content were analyzed.

2. Experimental Methods

2.1. Materials. The cement used in this study was ordinary Portland cement (OPC) with a specific gravity of 3.15 g/cm³ and a Blaine fineness of 330 m²/kg. The cement was supplied by S company in South Korea. GGBFS was produced by the K iron steel company in South Korea; it has a specific gravity and Blaine fineness of 2.8 g/cm³ and 420 m²/kg, respectively. The chemical composition of the cement and GGBFS was determined by X-ray fluorescence (XRF) spectrometry and is summarized in Table 1. Figure 1 shows the X-ray diffraction (XRD) spectra of the raw materials. Sodium carbonate (Na₂CO₃, anhydrous, ≥99%, C company, South Korea) was used as the alkali activator.

2.2. Mixture Proportions, Sample Preparation, and Curing. Mixture proportions of the samples are summarized in Table 2. The OPC was partially replaced with 50, 60, 70, 80, or 90% GGBFS (by weight). The weight ratio of water to the binder (OPC + GGBFS) was 0.45. Na₂CO₃ was added at 0, 1, 2, 3, 4, and 5 wt.% of the binder for each mixture. The 50, 60, 70, 80, and 90% GGBFS replacement levels are denoted as R5, R6, R7, R8, and R9, respectively. A total of 30 different paste mixtures were prepared in the laboratory according to ASTM C305 [18]. After mixing, cubic samples of 50 × 50 × 50 mm were cast for use in the determination

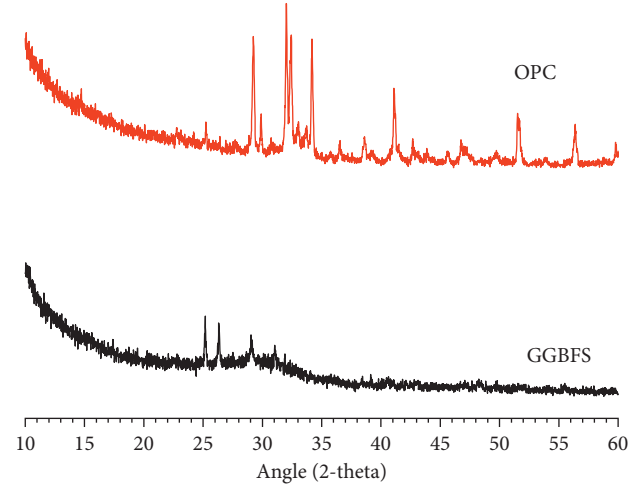


FIGURE 1: XRD spectra of the raw materials.

TABLE 2: Mixture proportions in the test pastes.

Sample ID	OPC (wt.%)	GGBFS (wt.%)	Na ₂ CO ₃ content (wt.% of OPC + GGBFS)
R5	50	50	0, 1, 2, 3, 4, and 5
R6	40	60	0, 1, 2, 3, 4, and 5
R7	30	70	0, 1, 2, 3, 4, and 5
R8	20	80	0, 1, 2, 3, 4, and 5
R9	10	90	0, 1, 2, 3, 4, and 5

of compressive strength and water absorption. Samples of 40 × 40 × 160 mm were cast for determination of dry shrinkage and ultrasonic pulse velocity (UPV). All samples were cured at a temperature of 23 ± 2°C and 80 ± 5% relative humidity until testing.

The compressive strengths of the hardened HVSC pastes were measured at 3, 7, and 28 d. The results are reported as an average of three samples. XRD was performed at 3 and 28 d. Fractured specimens from the strength tests were finely powdered and immersed in isopropanol to stop hydration. After vacuum drying, particles passing through a no. 200 sieve were used for XRD analyses. The water absorption, dry shrinkage, and UPV tests were conducted in accordance with ASTM C642 [19], C157 [20], and C597 [21], respectively.

3. Results and Discussion

3.1. Compressive Strength Development. The compressive strengths of HVSC pastes with and without Na₂CO₃ are shown in Figure 2. Previous studies [7, 11, 22, 23] reported a decrease in compressive strength with increasing slag content in slag-cement systems. For the R5, R6, R7, R8, and

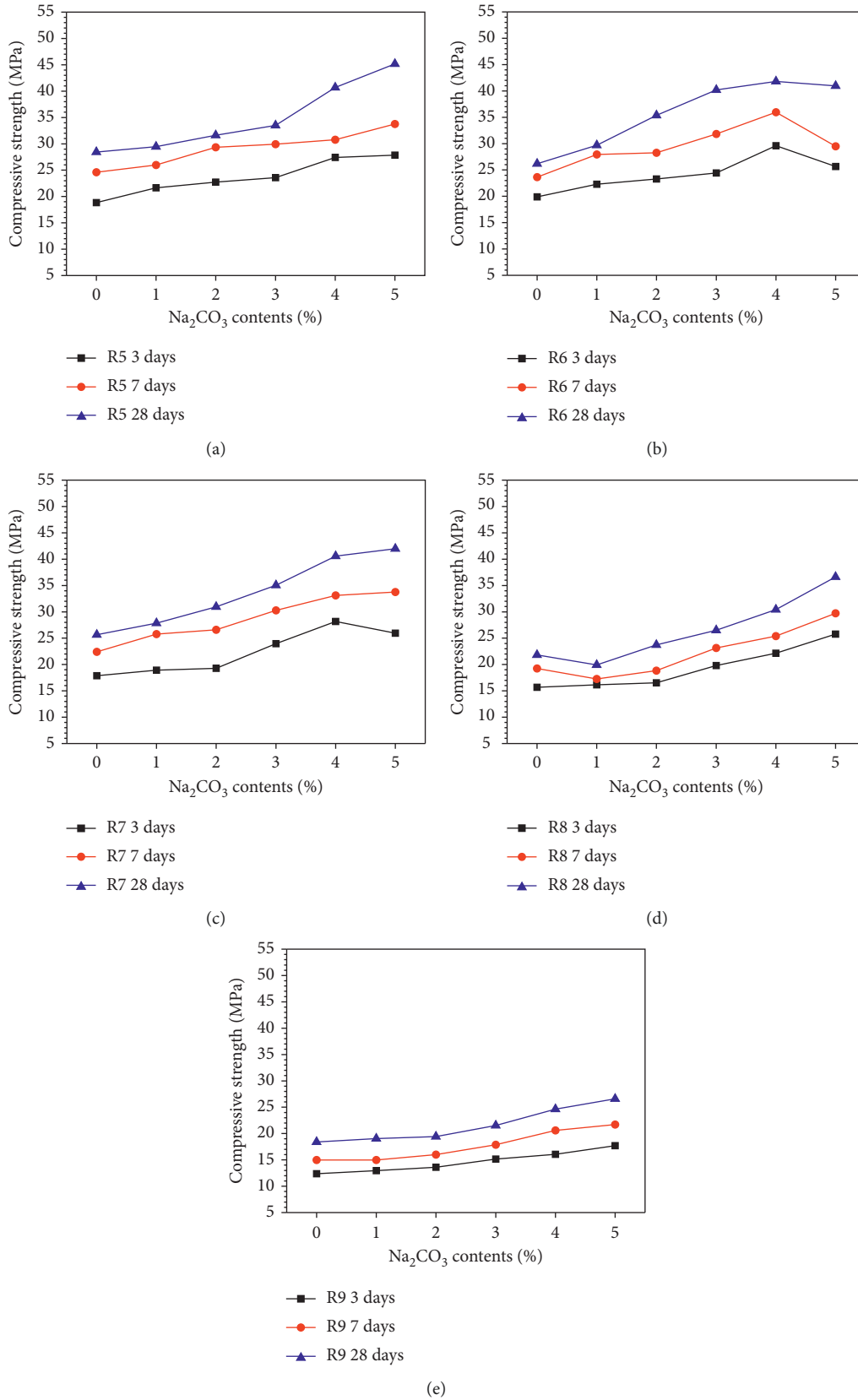


FIGURE 2: Compressive strengths of the Na_2CO_3 -activated HVSC pastes with different replacement levels of GGBFS: (a) R5, (b) R6, (c) R7, (d) R8, and (e) R9 samples.

R9 samples without the Na_2CO_3 activator, the strengths decreased slightly with increasing replacement level of GGBFS. This result is in agreement with previous studies on HVSC [7, 11, 22, 23].

For the R5, R6, R7, R8, and R9 samples with the Na_2CO_3 activator, the strengths of samples were higher than those of the samples without Na_2CO_3 at all ages, regardless of the amount of Na_2CO_3 added. The exception to this was the R8 sample, in which Na_2CO_3 contents of 3% or more showed higher strength than 0% Na_2CO_3 . With respect to the amount of added Na_2CO_3 , the results indicated that there is a trend of increasing strength with increasing Na_2CO_3 content, except for the R6 and R7 samples. The R6 and R7 samples showed a decrease in strength at the addition of 5% Na_2CO_3 .

Table 3 presents a comparison of the strengths of samples with Na_2CO_3 relative to the results without the activator. The results listed are given relative to the strength of the sample without Na_2CO_3 , which was considered to have a value of 100%. The strength of the HVSC with Na_2CO_3 was 3.06~64.29% greater after 3 days of curing, compared to that without Na_2CO_3 . The strength results showed that the addition of Na_2CO_3 contributed to the improvement in both the early- and later-age strengths in the HVSC system.

3.2. X-Ray Diffraction (XRD) Analysis. XRD spectra of the HVSC samples with Na_2CO_3 are shown in Figure 3. Generally, the XRD spectra show the typical contributions of hydrated cement: portlandite [$\text{Ca}(\text{OH})_2$] and C-S-H. As the amount of OPC decreases (R5 \rightarrow R6 \rightarrow R7 \rightarrow R8 \rightarrow R9 sample), the intensity of the portlandite reflections becomes weak until the peaks corresponding to portlandite are no longer observed in the XRD spectra. There seems to be no portlandite phase in the R8 (20% OPC + 80% GGBFS) or R9 (10% OPC + 90% GGBFS) samples (Figures 3(d) and 3(e)). For the R5, R6, and R7 samples, the intensity of the portlandite peaks decreases with increasing amount of Na_2CO_3 . This may indicate that the addition of Na_2CO_3 contributes to an increase in GGBFS activation, which then leads to an enhancement of the GGBFS pozzolanic reaction [24, 25]. As a result, it would be likely to form additional C-S-H. Kovtun et al. [26] reported on the Na_2CO_3 -activated slag concrete accelerated by different admixtures. They used NaOH, OPC, and a combination of silica fume and slaked lime as a chemical accelerator. They mentioned that slag hydration could be accelerated by C-S-H formed in the rapid cement hydration, which is attributed to the alkaline solution of Na_2CO_3 . For each sample in this study, it was observed that the formation of C-S-H was increased with increasing the addition of Na_2CO_3 . It could imply the rapid hydration reaction of C_3S and C_2S compounds in the OPC due to the use of Na_2CO_3 because the compounds play a role in generating C-S-H gel during the hydration reaction. It was identified that the peaks of C_3S and C_2S decrease as the amount of Na_2CO_3 increases.

Bernal et al. [16] reported that the early-age reaction in a CO_3^{2-} -activated binder occurs between CO_3^{2-} in pore solution and Ca^{2+} released by slag dissolution. They mentioned

TABLE 3: Relative strengths of samples with the Na_2CO_3 activator compared to the strength of samples without activation.

Sample ID	Age (d)	Na_2CO_3 content (wt.%)				
		1	2	3	4	5
R5	3	114.98	120.66	125.17	145.57	147.85
	7	105.61	119.28	121.68	125.13	137.29
	28	103.62	111.19	117.80	143.12	158.88
R6	3	112.07	117.04	122.83	151.23	128.96
	7	118.28	119.64	134.74	152.14	124.80
	28	113.36	135.05	153.53	159.60	156.40
R7	3	105.93	107.89	134.02	157.58	145.22
	7	115.00	118.75	135.22	147.86	150.71
	28	108.62	120.66	136.61	158.25	163.63
R8	3	103.06	105.42	126.21	141.14	164.29
	7	89.66	97.82	120.26	131.90	154.34
	28	91.33	108.71	121.55	139.43	168.00
R9	3	104.77	109.94	122.47	129.75	143.17
	7	100.00	106.74	119.21	137.36	144.83
	28	103.48	105.49	117.01	133.86	144.51

The results are listed relative to the strength of the sample without Na_2CO_3 , which was considered to have a value of 100%.

that enough Ca^{2+} could lead to consume CO_3^{2-} in the early age, thus resulting in the acceleration of slag hydration. They explained that the acceleration is because the slag hydration starts after the CO_3^{2-} is exhausted. In this study, OPC was used as a source of calcium in order to consume carbonate. It was observed that the calcite was formed at the age of 3 days in the R5 sample, and the calcite was increased with increasing the content of Na_2CO_3 . As a result, it can be seen that the use of Na_2CO_3 led to the formation of additional C-S-H and the generation of calcite, thus increasing the strength of the HVSC sample.

3.3. Water Absorption. Figure 4 shows the results of water absorption tests for the HVSC pastes with and without Na_2CO_3 . Samples with low strength could not give accurate water absorption results because the weak texture on the surface of the sample softened and came off during water immersion. Therefore, the water absorption was measured for samples at 28 days, when it could be accurately obtained for all samples.

As water absorption can be partially affected by cracks and pores in the samples [27], a sample with low water absorption is likely to imply that it has a relatively dense matrix. Increasing water absorption is thus associated with a reduction in strength. Previous studies [5, 23] reported that the water absorption of HVSC decreased with increasing replacement level of GGBFS in slag-cement systems. In contrast, Chen and Brouwer [28] noted that with increasing slag content in slag-OPC mixtures, the total porosity also increased and suggested that this could be due to the low degree of slag hydration in OPC. It can be deduced from their study [28] that as the slag content increases, the water absorption would increase due to the increase of total porosity. This result is close to our findings. This study showed that with increasing amount of GGBFS, the water absorption of HVSC pastes without Na_2CO_3 increased, and the

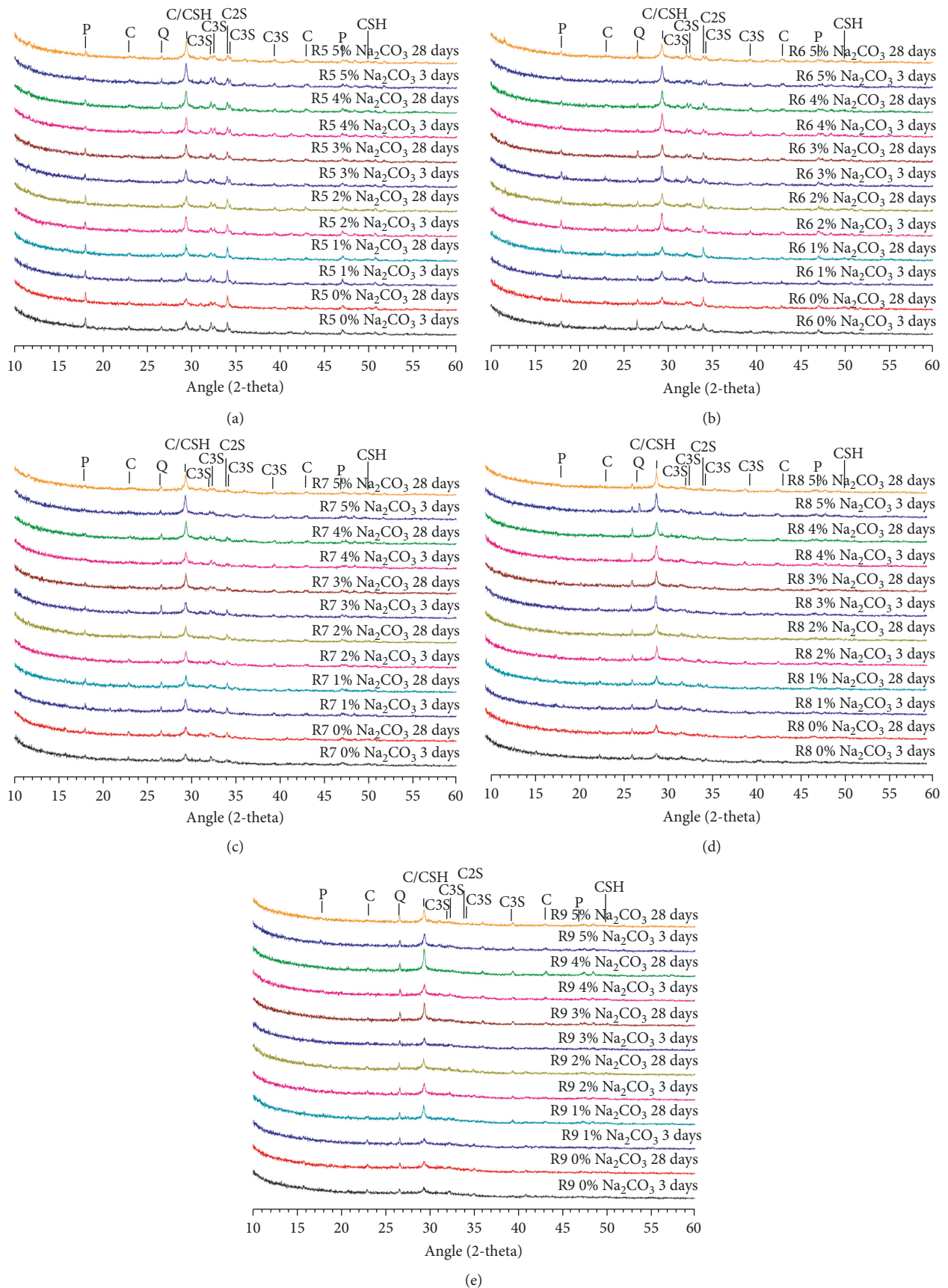


FIGURE 3: X-ray diffraction spectra for Na₂CO₃-activated HVSC pastes with different GGBFS replacement levels: (a) R5, (b) R6, (c) R7, (d) R8, and (e) R9 samples. Peak positions are indicated for portlandite (P); calcite (C); quartz (Q); C–S–H (CSH); alite (C₃S); belite (C₂S).

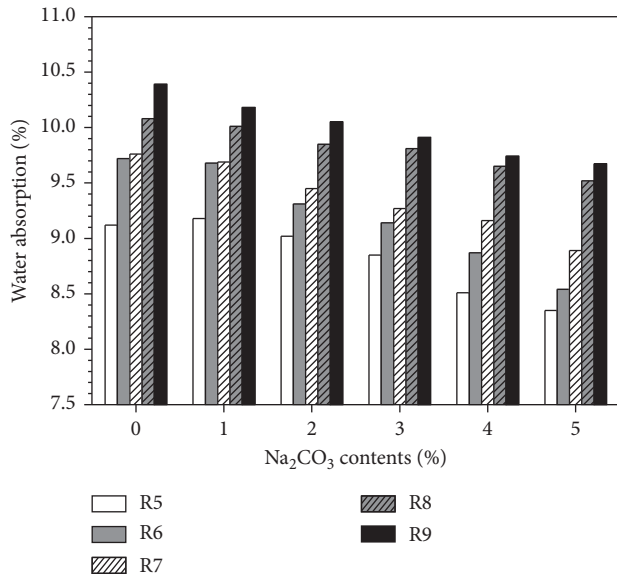
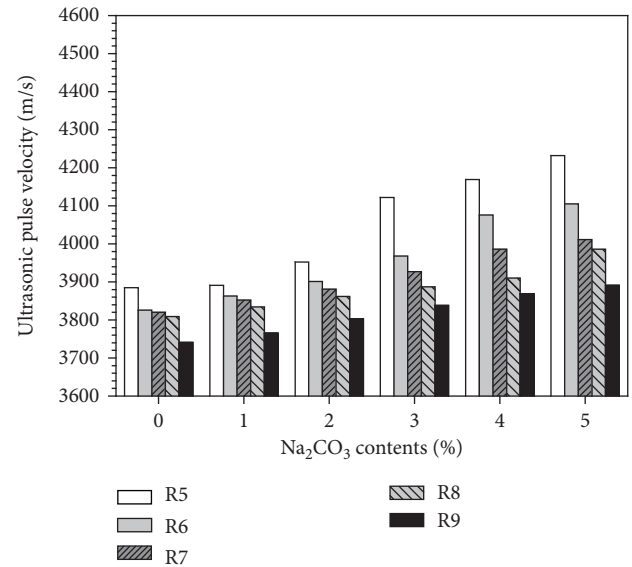


FIGURE 4: Water absorption of HVSC pastes containing Na_2CO_3 .

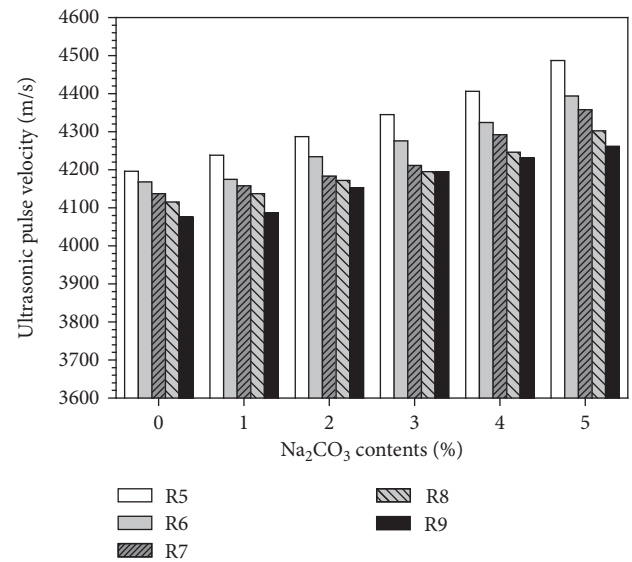
compressive strengths decreased. With respect to the amount of added Na_2CO_3 , as the amount of Na_2CO_3 increases, the water absorption decreased, implying that the strength would be higher with the amount of added Na_2CO_3 .

Mo et al. [29] reported that the formation of CaCO_3 in the steel slag pastes is attributed to the microstructure densification, thereby improving the strength of the pastes. In Figure 3, the R5 sample had the highest calcite peak compared with other samples. It can indicate that the R5 sample has a denser microstructure than other samples, which would result in the reduction of water absorption. It can be seen that the R5 sample gives lower water absorption than other samples. Note that the reaction products were portlandite and C-S-H as well as calcite. Therefore, it is clear overall that water absorption of samples would not be influenced only by the formation of calcite. It could be seen that the combined action of calcite and C-S-H gel has an effect on reducing water absorption by decreasing the overall porosity of the matrix.

3.4. Ultrasonic Pulse Velocity (UPV). UPV results for HVSC samples at the ages of 3 and 28 days are shown in Figure 5. UPV can be used to predict the quality of hardened samples, as UPV is related to the strength of the samples [22, 30]. Higher UPV values indicate higher strength. This study showed that UPV, for each Na_2CO_3 content, decreased with increasing GGBFS content (R5 \rightarrow R6 \rightarrow R7 \rightarrow R8 \rightarrow R9 sample). It can be mentioned that as the amount of GGBFS increases, the quality of matrix in samples would be comparatively lower, which could cause decrease in strength. When Na_2CO_3 was added to the HVSC sample, the UPV increased with increasing Na_2CO_3 content for all mixtures. In addition, the UPV values tended to increase with curing age for all samples. An increase in UPV implies that more reaction products were produced in the sample during the



(a)



(b)

FIGURE 5: Ultrasonic pulse velocity (UPV) for HVSC pastes containing Na_2CO_3 at (a) 3 days and (b) 28 days.

activation and hydration of the binder, and thus, the sample matrix would be denser, resulting in the improvement of strength.

Considering the density of reaction products, calcite has 2.71 g/cm^3 and portlandite has 2.44 g/cm^3 [29]. It is clear that the density of calcite is higher than that of portlandite. It is expected that the density of a reaction product could be related to the microstructure densification in the sample. The XRD result showed that the higher the content of OPC and the higher the concentration of Na_2CO_3 , the greater the amount of calcite formed. The increase in calcite with higher density than other reaction products under the same circumstances could imply that HVSC density would be increased. It would lead to the improvement of microstructure densification and the increase in the UPV.

Figure 6 shows the relationship between UPV and water absorption for all samples at 28 days. It is found that the UPV is related to the water absorption of the hardened samples: the higher the UPV, the lower the water absorption. This suggests that the addition of Na_2CO_3 is effective for increasing the densification of HVSC samples. It is also confirmed in Figure 6 that the sample with the highest UPV and lowest water absorption at 28 days is the R5 sample with 5% Na_2CO_3 .

3.5. *Shrinkage*. Shrinkage generally occurs at an early age [31], and the maximum shrinkage rate occurs during the first 7 days of aging [32]. In this study, the shrinkage of HVSC samples with varying Na_2CO_3 contents was measured for 35 days, in accordance with ASTM C157 [20]. Figure 7 shows the shrinkage results after 35 days of aging.

As previously noted, increasing the amount of GGBFS resulted in an increase in water absorption and a decrease in UPV, as the mechanical properties of Na_2CO_3 -activated HVSC pastes are related to the quality of hardened samples by the amount of GGBFS. However, drying shrinkage of HVSC samples has a different tendency from UPV or water absorption. R5 and R6 samples increase with increasing the content of Na_2CO_3 . The R7 sample shows similar dry shrinkage regardless of Na_2CO_3 content. R8 and R9 samples decrease the drying shrinkage with increasing Na_2CO_3 content. Duran Atiş et al. [33] studied the influence of activators on the dry shrinkage of alkali-activated slag mortar and showed that a Na_2CO_3 -activated slag mortar with an 8% Na concentration had greater shrinkage than that with 4% Na at 28 days. At the same time, the sample strength with 8% Na was higher than that with 4% Na. Some studies [31, 34–37] have found that the activator dosage, porosity, pore size distribution, and curing conditions can influence the dry shrinkage of alkali-activated slag. In this study, the different drying shrinkage tendencies based on the R7 sample can be considered as follows: because the drying shrinkage of samples was measured only up to 35 days, the results can be different from long-term age shrinkage. In the cases of R5 and R6 samples, the drying shrinkage was high. It would be due to the high OPC content in the HVSC sample. On the contrary, R8 and R9 samples would be relatively slow in reactivity due to the low OPC content. Therefore, the samples were still undergoing hydration reaction, and the shrinkage would not be completed. In the case of the R7 sample, there was little change in drying shrinkage. It seems that the hydration reaction of OPC and GGBFS was simultaneously proceeding. It can indicate the importance to make the measurement until the long-term age.

4. Conclusions

The following conclusions were obtained from the results of this experimental investigation:

The HVSC samples with the Na_2CO_3 activator generally produced greater strength than those without Na_2CO_3 at early and later ages, regardless of the amount of added Na_2CO_3 . It was identified that Na_2CO_3 was effective for

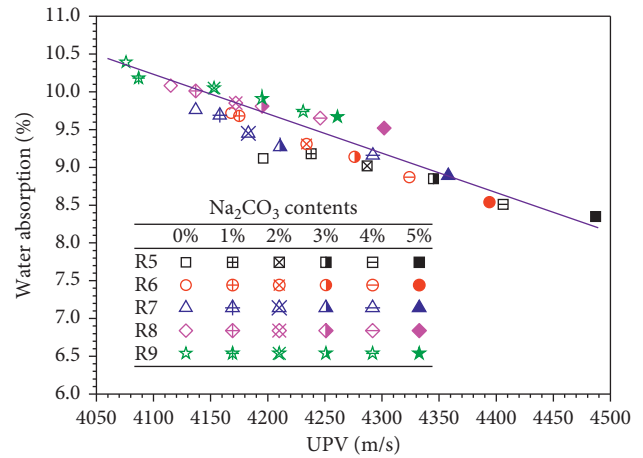


FIGURE 6: Relationship between water absorption and UPV for all samples containing Na_2CO_3 .

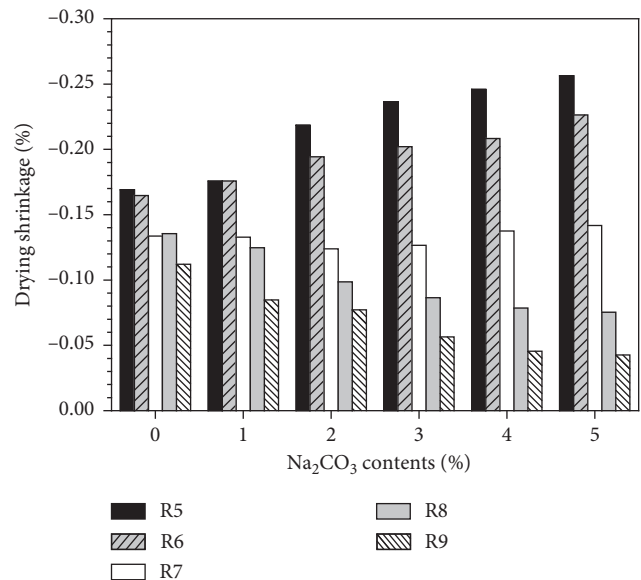


FIGURE 7: Drying shrinkage for HVSC pastes containing Na_2CO_3 .

increasing the HVSC sample strength at all ages. With respect to the amount of added Na_2CO_3 , the strength of HVSC samples increased with increasing Na_2CO_3 content. The compressive strength results indicated that the optimum combination for the early and later ages was the R5 sample with 5% Na_2CO_3 .

The CO_3^{2-} ion of Na_2CO_3 forms calcite with Ca^{2+} released by GGBFS dissolution. It indicated that the addition of Na_2CO_3 in the HVSC sample contributes to an increase in GGBFS activation, which then leads to an enhancement of the GGBFS pozzolanic reaction. As the amount of Na_2CO_3 increases, the peaks of calcite and C–S–H in HVSC samples increased.

Additional C–S–H formation due to GGBFS pozzolanic reaction and the generation of calcite would lead to the improvement of microstructure densification in the HVSC sample. The HVSC samples with Na_2CO_3 resulted in a decrease in water absorption and an increase in UPV, comparing to

the HVSC without Na_2CO_3 . The drying shrinkage of HVSC samples should be taken into consideration of the long-term measurement and GGBFS hydration degree.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

This research was supported by Basic Science Research Program through the National Research Foundation of Korea (NRF) funded by the Ministry of Science, ICT and Future Planning (NRF-2014R1A1A3052424) and the Ministry of Education (NRF-2015R1D1A3A01019583 and NRF-2017R1D1A1B03034700).

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