

# Study of the Carbonation Mechanism in Natural Exposure and Accelerated Test Using Blast Furnace Slag

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# ABSTRACT

Using of admixtures is highly recommended for reducing environmental impact by cement industry. Using ground granulated blast furnace slag, which is a type of admixture, has lots of advantages, however there is concerned that the resistance to carbonation decrease. Progress of carbonation is one of standard for durability design in the structure, and checking for carbonation is essential. However, in many cases, carbonation is often judged using accelerated carbonation tests. In some research there is not much difference in carbonation depth between Ordinary Portland cement(OPC) and blast furnace cement in natural exposure. On the other hand, there is difference in the accelerated carbonation test, and the value of the carbonation rate coefficient for the ground granulated blast furnace slag becomes higher than OPC. Also in general, concrete carbonation produced calcite which is Calcium carbonate. However, in the hardened carbonated mortar, there is a different crystal structure from Calcium carbonate which is vaterite and aragonite. For this reason, it is thought that not only calcium hydroxide but also CSH accounts for near 70% of the hydration in concrete, are involved in carbonation.

In this study, carbonation test would be conducted on the hardened mortar made with ground granulated blast furnace slag. Curing period would also be changed to focused on the pores of the hardened mortar until carbonation is initiated. The study would also be focused on the carbonation mechanism that occurred as a result of chemical change during carbonation of hardened mortar when ground granulated blast furnace slag is used.



# 1. GENERAL INTRODUCTION

Deterioration prediction of cement hardened such as concrete is very important in prolonging the lifetime of structures. Regarding such deterioration prediction, since there are so many studies, deterioration phenomena under actual circumstances require a long period of time, so in the study of deterioration prediction based on many materials, promotion experiments using conditions are made. However, there remains a question as to whether the promotion condition simply advanced the condition of the real environment. This is because the deterioration conditions are different, it can be assumed that the reaction mechanism is different and the speed changes. For example, in the case of the acid resistance test, it is thought that the response mechanism under extremely severe conditions, such as the case where the reaction with acid rain etc. is performed under gentle conditions and the reaction under very severe conditions like different conditions of sewage. As a result, even in the test immersed in the same acid, the progress of deterioration of concrete differs depending on the composition because of its different pH. Regarding the carbonation of concrete, many researches have been conducted to estimate the speed at actual exposure from the accelerated test results, as testified by Uomoto-Takada, the carbonation coefficient rate is calculated from carbonic acid It is considered to be proportional to the square root of gas concentration. However, studies on differences in reaction mechanism under each carbon dioxide concentration condition are not much studied, and it is well known that it is difficult to compare reaction rates.

In general, the carbonation depth used for predicting deterioration can be obtained by measuring the color by spraying a phenolphthalein solution. However, in the case of accelerated curing test specimens, it is often observed that the carbonation area which appeared white immediately after spraying also turns reddish purple by time. Under present circumstances such explanation of the event is not done well. In addition, the authors observed the progress of carbonation from one side of cement paste by XRD, in the actual environment exposure and acceleration environment, there were differences in the formation situation of calcite and vaterite and the decrease in CH at low concentrations of CO<sub>2</sub>, calcite preferentially formed and vaterite predominantly formed at high concentrations of CO<sub>2</sub>. As described above, differences in carbonate were also observed from the past experiments, but the rate of carbonation is not only influenced by chemical composition but also greatly influences by the pore structure, so many researchers have found that pores. It was investigated volume, pore size distribution and so on.

In this study, it was focused on the difference of reaction mechanism in carbonation under real environment and accelerated environment and investigated the effect of hydrate composition on reaction rate. Changes in carbonation products and pore structures will be discussed separately, and in order to obtain cement hardened with different amounts of CH and C-S-H, ground granulated blast furnace slag which is often used as an effort to reduce environmental impact in recent years. The amount of addition was changed.

# 2. COMPARING THE PROGRESS OF CARBONATION IN A REAL AND ACCELERATED ENVIRONMENT

In order to prepare samples with different amounts of CH and C-S-H, mortar was prepared by changing the blast furnace slag substitution rate, and the curing condition was changed by changing the curing condition. Carbonation was carried out in a real environment and an accelerated environment, and the progress of carbonation was investigated.

### 2.1 Preparation and use of samples materials and specimen specifications

Table 1 shows the chemical analysis results of ordinary Portland cement used in this study and ground granulated blast furnace slag. Table 2 also shows the mix proportion of the mortar used in the experiment. The fine aggregate used was Kimitsu-shi from Chiba prefecture (F.M. 2.57, surface dry density  $2.62g/cm^3$ , water absorption rate 1.48%). In this study, it was focused on how the hydrate composition relates to the carbonation mechanism, so carbonation test was carried out by changing the blended amount of ground granulated blast furnace slag by 6 blending ranging from low substitution to high substitution. Here, B100 is 100% of blast furnace slag, but in order to promote hardening, calcium hydroxide (15%) is added as a stimulant. The specimen size of the mortar specimen was  $40 \times 40 \times 160$ mm, the water binding ratio W/B was constant at 50% for all formulations and the mass ratio of water: binder: fine aggregate was 0.5:1:3. The implanted mortar prevented



moisture escape by covering the upper surface with a wrap film in an environment of 20°C. It was demolding on the next day without curing, and the sealing curing was carried out for 7 days and 28 days, and three kinds of specimens under this curing condition were subjected to the carbonation test. For accelerated carbonation test only 56 days of sealing curing was added. The reason for changing the period of curing days is to change the amount of hydrate in the mortar and the pore structure because of the difference in the progress of the hydration reaction in the specimen.

### Table 1. Chemical components of materials

Cement	Density [g/cm <sup>3</sup> ]	Specific surface area	Contents(mass%)											
		[cm <sup>2</sup> /g]	SiO <sub>2</sub>	$AI_2O_3$	$\rm Fe_2O_3$	CaO	MgO	$SO_3$	Na <sub>2</sub> O	$K_2O$	TiO <sub>2</sub>	$P_2O_5$	MnO	CI
OPC	3.16	3500	21.28	5.09	3.15	65.36	1.01	2.01	0.32	0.41	0.25	0.14	0.1	0.006
BFS	2.91	4290	33.42	13.43	-	43.29	6.02	2.62	0.27	0.3	0.6	0.01	0.18	0.004

Table 2. Mix proportion for binders

	W/D(0/)	S/C	Ratio of Binder (weight %)				
	W/B (%)	S/C	OPC	BFS			
B0			100	0			
B20		2	80	20			
B50	50		50	50			
B70	30	3	30	70			
B90			10	90			
B100			0	100			

# 2.2 Carbonation tests

The top and carbonated as two open faces. The test specimen was allowed to stand still in an outdoor environment which not affected by rain, and was allowed to stand in an accelerated carbonation test apparatus (temperature:  $20^{\circ}$ C, RH 60%, CO<sub>2</sub> concentration: 5%). The carbonation depth was measured according to JIS standards, split at each age of carbonating material, sprayed with a phenolphthalein solution, measured 8 points from the surface to a portion that turned reddish purple, and the average value was taken as the carbonation depth.

The carbonation coefficient rate was calculated using equation (1) on the assumption that the carbonation reaction follows the square root rule.

$$X = A\sqrt{t}$$
(1)

Where, X: carbonation depth (mm), t: carbonation period (year), A: carbonation coefficient rate

### 2.3 Compressive strength tests

After completion of curing, compressive strength test of mortar at the time of carbonation start (sealing curing 7, 28, 56days) was carried out according to "Physical test method of cement (JIS R 5201-2015)".

### 2.4 Total pore volume

At the time of performing the compressive strength test, the porosity was determined by the Archimedes method. The sample size is  $40 \times 40 \times 20$  mm. As a pre-treatment, hydration reaction was stopped by subjecting acetone to vacuum saturation treatment. In the test, the sample was placed in water and saturated in vacuum for 2 hours to measure the mass in water and the saturated water mass. Thereafter, it was dried in an oven drying at  $40^{\circ}$ C for about 1 week to obtain an absolute dry weight. From the obtained results, the pore ratio (vol%) was calculated using the equation (2).

$$P = (S - A) / (S - M) \times 100$$
(2)

Here, P: Porosity (%), S: saturated water mass, A: absolute dry mass, M: mass in water



### 3. EXPERIMENTAL RESULTS AND DISCUSSION

### 3.1 Relationship between porosity and strength

Figure 1 shows the relationship between the total pore volume obtained by mortar with different substitution rate of ground granulated blast furnace slag and compressive strength. The relationship between the porosity and compressive strength was different in the specimens of 7 days sealing and 28 days sealing curing. At the same porosity, the compression strength was lower in the case of curing for 28 days. It is considered that the drying is not sufficient with respect to the size of the cured, and the pore may be measured too little.



### Figure 1. The relationship between porosity and compressive strength of mortars cured for 7 and 28days

#### 3.2 Compare the carbonation test under real environment and accelerated environment

Figure 2 shows the relationship between the carbonation period and the carbonation depth of specimens with different curing conditions. It was found that the carbonation reaction progressed considerably even in a real environment, in a specimen with a short curing period, that is, a hydration reaction did not proceed. This tendency was remarkable especially in specimens with high ground granulated blast furnace slag replacement rate. The results of the carbonation in the accelerated environment without curing seem to be disturbed in the long-term carbonation period in which the carbonation has been particularly deeply carbonized is because the size of the test specimen is smaller than the carbonation period It seems that it got smaller and an error occurred in the measurement.

From these results, carbonation coefficients rate is obtained by assuming that they follow the square root rule, respectively, and are shown in Figure 3 and 4 As the substitution rate increases, the carbonation coefficient rate does not proportionally increase, but it becomes remarkably large when the substitution rate exceeds 70%. The difference in curing period in the accelerated test was considerably close in the sealing regimen on 7 days and 28 days. The reason why the carbonation coefficient rate decreases by applying curing is that as the period of carbonation becomes longer, the carbonation depth becomes larger than the influence range of the curing, so that the curing at the carbonation rate It is thought that the influence becomes small.





Figure 2. Changes in the carbonation depth on different curing periods and concentration of carbon dioxide

[Carbonation conditions] (a), (c), (e): Natural conditions, (b), (d), (f): accelerated conditions [Curing conditions] (a), (b): No curing, (c), (d): curing for 7days and (e), (f): cured for 28days



Figure 3. Coefficient of carbonation rate with mixing ratio: under natural condition





# Figure 4. Coefficient of carbonation rate with mixing ratio: under accelerated condition

# 3.3 Relationship between carbonation coefficient rate under real environment and accelerated environment

Prediction of long-term carbonation deterioration of cement hardened. carbonation prediction is often performed by converting the carbonation coefficient rate obtained from the test result performed in the advanced condition into the carbonation coefficient rate under the real environment. Here, as the prediction expression, the Uomoto-Takada's equation (3), and the relational expression is expressed as a function of the CO<sub>2</sub> concentration in the carbonation environment.

$$kc^* = (2.804 - 0.847 \log_{10} 0 (C)) * C^{0.5}$$
 (3)

Where, Kc \*: Carbonation coefficient rate C: Carbon dioxide concentration

As a result, the coefficient (ka) of the accelerated environment (concentration 5%) is calculated to be 7.68 times the coefficient (kn) of the real environment (concentration 0.04%). Consideration of carbonation process

# 4. CONSIDERATION OF CARBONATION PROCESS

### 4.1 Observation of cleavage plane of carbonated cured

Figure 5 shows the coloration condition when spraying the phenolphthalein solution on the split face. Here, representative carbonation results of the real environment at 7 days of age and the carbonation result of the accelerated environment at the time when the same carbonation depth is reached are shown without curing of B0, B50, B70, B90. A major feature here is that under the accelerated environment of B0, which is thought to have a large amount of CH production, a pink zone clearly appears. Such a phenomenon is often observed under an accelerated environment in a cement hardened having a large amount of OPC. It was confirmed that in the measurement results of the authors, CH remained in the carbonation area where phenolphthalein no longer coloring under accelerated conditions. This implies that the front of the carbonation layer is moving to a deeper position before CH is all carbonated. On the other hand, such a zone hardly appears in a cement hardened (B90) in which the mixing ratio of ground granulated blast furnace slag is large, in which the amount of OPC is small, that is, the amount of CH generated is small. This is considered to be caused by the absence of CH remaining in the carbonation area.

# 4.2 On the reactivity of carbonation

The progress of carbonation was considered to be due to the diffusion of carbon dioxide, and the conditions under which the carbonated front was advanced in the cement hardened were examined. In this study, CH and C-S-H were taken into consideration for the amount of hydrate to be carbonated in the cement hardened of its influence. CH was converted to CaCO<sub>3</sub> by carbonation, and C-S-H was converted to SiO<sub>2</sub> gel and CaCO<sub>3</sub> by carbonation. Focusing on CaCO<sub>3</sub>, [CO<sub>3</sub><sup>2-</sup>] supplied to the front is assumed to generate CaCO<sub>3</sub> by reaction with CH component in CH and C-S-H. First, the properties of CH and C-S-H are organized. As to the potential reactivity with CH and C-S-H in [CO<sub>3</sub><sup>2-</sup>], the solubilities of CH and C-S-H are determined and the ion product of [Ca<sup>2+</sup>] and the environment





### Figure 5. Observation of fractured surfaces colored by sprayed phenolphthalein solutions

 $[CO_3^{2^2}]$  becomes larger than the solubility product (Ksp) of CaCO<sub>3</sub> It is considered that the reaction proceeds. According to the calculation from the thermodynamic data, the solubility product (Ksp) of CaCO<sub>3</sub> is 4.50×10<sup>-9</sup>. The solubilities of CH and C-S-H vary with pH, but they are 0.0125mol/L and 0.003mol/L, respectively, with respect to water. Therefore, the lowest  $[CO_3^{2^2}]$  at which CaCO<sub>3</sub> precipitates can be calculated as  $3.60\times10^{-7}$ mol/L and  $1.50\times10^{-6}$ mol/L, respectively, that is, C-S-H has better  $[CO_3^{2^2}]$  is about 20 times stronger, it can be said that the reaction does not start. Here, the reaction equations of dissolution used for calculation is as follows.

$$CaCO_3 = Ca^{2+} + CO_3^{2-}$$
 (4)

$$Ca(OH)_2 = Ca^{2+}+2OH^-$$
(5)

$$C_{2}SH_{1.17} + 1.83H_{2}O = 2Ca^{2+} + H_{3}SiO_{4} + 3OH^{-}$$
(6)

In the real environment, it is assumed that the  $[CO_3^{2-}]$  supply rate is smaller than the reaction rate of CH, and "Process in which CH reacts constantly when CH exists and C-S-H reacts when CH disappears" is conceivable. On the other hand, in the accelerated environment, since the supply rate of  $[CO_3^{2-}]$  is superior to the reaction rate of CH, "a process in which both CH and C-S-H react as well as being exposed to an accelerated environment" can be considered.

Considering the size of each particle, the size is about several  $\mu$ m. It is often observed from the results of SEM and the like, and from the data of gas adsorption, CH is several nm thick overlapping of the crystal structure unit sheets It can be thought of as a sheet with something. Each reaction follows the diffusion rule (Jander's equation), It is inversely proportional to the cube of the magnitude of the particle radius, and it is considered that the particle diameter absolutely influences the velocity coefficient.

### 4.3 Model of carbonation progression under real and accelerated environment

In the real environment, the supply of  $[CO_3^{2-}]$  is gentle, so it is imaginable that the supply of momentary  $[CO_3^{2-}]$  at the front is small,  $[CO_3^{2-}]$  reacts with CH, but C-S-H cannot react. Then, in the reaction at the carbonated front, CH reacts first, it is consumed, and when  $[CO_3^{2-}]$  is supplied, C-S-H begins carbonating. When C-S-H is carbonated,  $[CO_3^{2-}]$  is supplied to a deeper part and the front moves forward. Under accelerated environment, the supply amount of  $[CO_3^{2-}]$  is large, and from the early stage, the concentration of  $[CO_3^{2-}]$  can be carbonated at the same time with CH at the same time as C-S-H. Therefore, CH and C-S-H both react, but its rate is high crystallinity, CH with large particle size is much slower than C-S-H with very small crystal size, and even if CH does not react, C-S-H reacts. It is conceivable to scoff, in which case the carbonated front moves forward. In other words, the reaction rate (forward speed of the coloration zone by phenolphthalein) is influenced by the amount of (CH+C-S-H) in the real environment, and is largely affected mainly by the amount of C-S-H in the accelerated environment it is conceivable that.

Figure 6 shows an image of the progress of carbonation front in real environment and accelerated environment. Under real circumstances, CH reacts first and C-S-H reacts after consumption when the







Figure 7. CO<sub>2</sub> distribution in carbonated zone under natural condition



Figure 6. Schematic process of carbonation in front zone (Comparing natural condition and accelerated condition)

Figure 8. CO<sub>2</sub> distribution in carbonated zone under accelerated condition

supply amount (concentration) of  $[CO_3^{2-}]$  is low (low). Under the accelerated environment, since the amount of  $[CO_3^{2-}]$  supplied is high,  $[CO_3^{2-}]$  of the environment increases, both CH and C-S-H react.

As a result, an image of  $CO_2$  concentration distribution in the carbonated layer is shown in Figure 7 and 8. In the real environment, the amount of  $CO_2$  corresponding to the same (CH+C-S-H) amount is distributed to the vicinity of the front, and in the case of acceleration conditions, although carbonation of CH occurs, it is considered that the amount of  $CO_2$  corresponding to C-S-H is distributed.

### 4.4 Calculation amount of CH and C-S-H in cement hardened

Based on the above-mentioned idea, among the hydrates, the target components of carbonation are considered to be the amount of CaO in CH and C-S-H in the cured product was determined. The author's previous research result was used as the amount of CH production. On the other hand, the amount of C-S-H was obtained from the research results of the authors from the reaction rate of C<sub>3</sub>S in OPC and the reaction rate of ground granulated blast furnace slag. In the study reported previously be the author, the amount of C<sub>3</sub>S was measured by Rietveld and the amount of CH was determined by TG-DTA.  $C_2S$  was assumed to be unreacted because it was considered in the range up to 28 days of age at this time, and it was assumed that only  $C_3S$  reacted.



Age	Nm	OPC	BFS	amount ofC₃S	hydration ratio of OPC	C-S-H from OPC	CaO in BFS	reaction ratio of BFS	C-S-H from BFS	Total amount of CaO from C-S-H	Total amount of CaO in CH	CaO from (C-S-H+CH)
1d	B0	100	0	60	0.55	14.59	0.00	0.00	0.00	14.59	9.73	24.32
	B20	80	20	48	0.55	11.67	8.60	0.54	4.60	16.27	7.78	24.05
	B50	50	50	30	0.55	7.29	21.50	0.18	3.83	11.12	4.86	15.99
	B70	30	70	18	0.55	4.38	30.10	0.09	2.86	7.23	2.92	10.15
7d	B0	100	0	60	0.94	24.93	0.00	0.00	0.00	24.93	16.62	41.56
	B20	80	20	48	0.94	19.95	8.60	0.59	5.11	25.06	13.30	38.35
	B50	50	50	30	0.94	12.47	21.50	0.42	9.03	21.49	8.31	29.80
	B70	30	70	18	0.94	7.48	30.10	0.25	7.46	14.94	4.99	19.93
28d	B0	100	0	60	1.00	26.53	0.00	0.00	0.00	26.53	17.68	44.21
	B20	80	20	48	1.00	21.22	8.60	0.94	8.11	29.33	14.15	43.48
	B50	50	50	30	1.00	13.26	21.50	0.54	11.55	24.82	8.84	33.66
	B70	30	70	18	1.00	7.96	30.10	0.37	11.16	19.12	5.31	24.43

Table 3. Calculation for CaO from C-S-H and CH on different cement types (Unit: weight%)

Therefore, it is assumed that the amount of CaO reacted by multiplying the hydration conversion ratio and the molecular weight of CaO/C<sub>3</sub>S to the C<sub>3</sub>S content in the binder. Subsequently, those which became CH and the one which became C-S-H with respect to the total amount of CaO were calculated by considering the C/S ratio. C/S of C-S-H is assumed to be 1.8. Therefore, the reaction of C<sub>3</sub>S in OPC is assumed to be the equation (7).

$$C_3S+H_2O=C_{1.8}SH_x+1.2CH$$
 (7)

Calculation of the amount of CaO in C-S-H produced by the ground granulated blast furnace slag is based on Table 1 and calculates the amount of CaO contained in the ground granulated blast furnace slag for each age for ground granulated blast furnace slag by the reaction rate of the reaction. As for the reaction rate of ground granulated blast furnace slag, the result calculated for each substitution rate by using the salicylic acid acetone methanol method of the previous study was used.

C-S-H from GGBFS = Content of CaO 
$$\times$$
 reaction rate (8)

The sum of the above calculations is the amount of CaO forming CH and C-S-H at each material age for each substitution rate. Table 3 shows the sum of the CaO contents of CH and C-S-H from the calculated OPC and C-S-H from GGBFS, respectively.

### 4.5 Relationship between hydrate composition and carbonation coefficient rate

As mentioned in the discussion of the carbonation process, the carbonation rate and the hydrate composition are thought to be highly related. Therefore, it was investigated the relationship with the carbonation coefficient rate using the amount of CaO contained as CH and C-S-H obtained in 4.3. Figure 9 shows the relationship between CaO content and carbonation coefficient rate. This figure shows the results obtained in the real environment and the accelerated environment, respectively. In both cases, it was found that there was a very good correlation between the CaO amount as the hydrate amount and the carbonation coefficient rate, regardless of the mixing ratio and the curing period. Also, when look at the reliability factor (R2 factor), it can be found that there is a better correlation in the accelerated environment than in the real environment.





### Figure 9. Relationship between carbonation coefficients rate and the amounts of CaO

### 5. CONCLUSION

For this study, it was investigated the carbonation mechanism of cement hardened, especially the difference between the real environment and the process under accelerated environment. The obtained results are shown below.

(1) Calcium hydroxide and calcium silicate hydrate were taken up as subjects of carbonation, and their solubility products were examined. As a result, under real environment, since the supply of  $CO_2$  is gentle, CH is first carbonated, then C-S-H is carbonated, and the front advances as both are consumed. On the other hand, in the accelerated environment, since the supply of  $CO_2$  is fast, carbonization of both CH and C-S-H from the beginning and carbonation of C-S-H with finer particle size result in carbonation front moving forward while leaving uncarbonated CH.

(2) Based on the above idea, it was calculated the carbonation coefficient rate of mortar with substitution rate of ground granulated blast furnace slag and curing condition changed

As a result of investigating the relation with CH and C-S-H amount, it was found that there is a very good correlation with the amount of CaO contained in the hydrate irrespective of the substitution rate and the curing condition.

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