- Technical Paper -

MECHANICAL PROPERTIES AND CARBONATION RESISTANCE OF SEA SAND CONCRETE CONTAINING GROUND GRANULATED BLAST FURNACE SLAG UNDER ACCELERATED CARBONATION

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ABSTRACT

This study aims to investigate the effect of accelerated carbonation on mechanical properties of sea sand concrete containing ground granulated blast furnace slag (GGBS). Carbonation depth was also measured. GGBS was used to partially replace OPC with a ratio of 45% by mass. The presence of chloride ion in non-desalted sea sand improved the mechanical properties and carbonation resistance of concretes regardless of GGBS replacement. Carbonation increased the compressive strength but decreased the modulus of elasticity of GGBS concrete using non-desalted and desalted sea sand. Keywords: Chloride ion, carbonation, blast furnace slag, compressive strength, modulus of elasticity

1. INTRODUCTION

Concrete is one of the most used construction materials on the earth with around 25 billion tons in 2016, resulting from the increase in demand of concrete for the development in building and infrastructure [1]. This increase leads to the overexploitation of river sand which is a common fine aggregate for concrete production. Under this circumstance, sea sand (SS) can be considered as an alternative material because it is very abundant at coastal areas. Sea sand is being exploited and used as aggregate for construction in UK, Netherlands, China. Although the exploitation of sea sand in Japan is now limited, it was reported that Japan was one of the most sea sand exploitation countries in the world with approximate volume of 39×10^6 m³ [1]. The utilization of SS not only is convenient in the transportation of material but also takes full advantage of local material, resulting in the reduction of marine construction cost. The direct use of SS without desalting for concrete production is attracting much more attention of researchers all over the world because there are some harmful ions in SS, e.g. prominent chloride ions, which can affect the mechanical properties and durability of reinforced concrete.

Some researchers have examined on the feasibility of SS as fine aggregate in concrete production. It was concluded that SS can be successfully used as a fine aggregate for plain concrete production when crushed limestone sand was partially substituted by SS from 15% up to 50% [2, 3]. Additionally, Chen et al. demonstrated that the sulphoaluminate cement concrete with SS has superior mechanical properties and sulfate resistance compared to ordinary Portland cement concrete with river sand because of the higher specific surface area of SS [3].

Ground granulated blast furnace slag (GGBS) is a by-product from the manufacture of iron and steel. It can improve the mechanical properties of concrete [4]. However, concrete containing GGBS was very sensitive to carbonation which is a major cause of reinforced concrete deterioration, especially when chloride ion is present in the concrete [5]. On the other hand, carbonation is a process of penetration of atmospheric carbon dioxide into cementitious materials, leading to the change in the microstructure of concrete [6]. Despite the fact that several studies regarding the use of SS for concrete production have been recently investigated, the combined effect of chloride ion and GGBS under the influence of carbonation on the mechanical properties and durability of concrete has not been studied sufficiently. More studies on the effects of carbonation on SS concrete containing GGBS are very necessary in order to provide more data as well as verify the feasibility of SS as construction material.

Therefore, the mechanical properties and carbonation resistance of SS concrete containing GGBS under accelerated carbonation were investigated in this study.

2. EXPERIMENTAL PROGRAM

2.1 Materials

The cementitious materials used in this study

Table 1 Chemical composition and physic	cal
properties of cementitious materials	

Cementitious		Chemical composition (% by mass)								
materials	CaO	SiO_2	Al_2O_3	Fe ₂ O ₃	MgO	SO_3	Na ₂ O	K_2O	Cl	LOI
OPC	64.79	19.89	5.19	3.07	1.26	1.95	0.31	0.36	0.006	2.65
GGBS	43.78	34.45	14.06	0.27	5.84	_	0.24	0.23	0.003	0.05
– Not measured										

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were ordinary Portland cement (OPC) and ground granulated blast furnace slag (GGBS) 4000 confirming to Japanese Industrial Standards JIS R5210 and JIS A 6206, respectively. The density and specific surface area of OPC and GGBS were 3.16 and 2.91 g/cm³, and 3340 and 4170 cm²/g, respectively. Main chemical compositions of these materials are shown in Table 1. The aggregate included coarse aggregate with the maximum size of 20 mm and two types of fine aggregates which were sea sands exploited from the Karatsu harbor in Saga prefecture in Japan. The physical properties of aggregates are presented in Table 2. Tap water was used for casting concrete specimens.

2.2 Mixture proportions and preparation of specimens

Four mixture proportions with a constant water-to-cementitious material ratio of 0.50 were prepared as shown in Table 3. The GGBS was used to partially replace OPC with a ratio of 45% by mass.

All specimens were cast in cylindrical plastic moulds with $\phi 100 \times 200$ mm for compressive strength and modulus of elasticity tests. In addition, the prismatic specimens with dimensions of $100 \times 100 \times 400$ mm were also prepared for accelerated carbonation test. The top surface of all specimens after casting was sealed with aluminum tape in order to prevent water evaporation and carbonation, and then placed in controlled room at $20 \pm 2^{\circ}$ C for 28 days.

For mechanical properties test, after sealing for days, half of cylindrical specimens were 28 continuously kept under sealed condition. The others were demoulded and put in accelerated carbonation chamber until the age of 182 days. For accelerated carbonation, only one surface 100×400mm of prismatic specimens was exposed to CO_2 , whereas the other surfaces were covered with aluminum tape. The CO₂ concentration in the chamber was 5%, and the temperature and relative humidity were 20±2°C and 60±5%, respectively. Although the carbonated hydration products under such high CO₂ concentration might be different from that under the natural carbonation, the testing results can be expected more obviously under high CO₂ concentration. The specimens under sealed condition were denoted by "S", whereas the others under accelerated carbonation were denoted by "C".

2.3 Measurement

(1) Thermogravimetric analysis (TGA)

The amount of Ca(OH)₂ (CH) in concretes after sealing for 28 days were measured by TGA with heating from room temperature (~20) to 1000 °C at 10 °C/min under a nitrogen atmosphere. The sample was collected from the core of specimens after compressive strength test. The CH content in sample was determined based on the ignited mass of the sample and the mass loss from differential thermogravimetric analysis (DTG) curve between the initial and final temperatures of the corresponding DTG peaks. Then, the CH content in concrete was calculated after quantitatively measuring the amount of aggregate and paste in sample

Table 2 Physical properties of fine aggregates

Materials	Density	Water absorption	Chloride ion content
Materials	(g/cm ³)	(%)	(%)
Coarse aggregate (G)	2.62	0.7	-
Desalted sea sand (DSS)	2.60	1.3	0.0054
Non-desalted sea sand (NSS)	2.60	1.3	0.1968

Table 3 Mixture proportions of concretes							
Mixtures	OPC-DSS	OPC-NSS	GGBS45-DSS	GGBS45-NSS			
OPC (kg/m ³)	350	350	193	193			
Water (kg/m ³)	175	175	175	175			
GGBS (kg/m ³)	-	-	158	158			
DSS (kg/m ³)	847	-	841	_			
NSS (kg/m ³)	-	847	_	841			
$G_{\rm c}$ (kg/m ³)	966	966	960	960			

Table 4 CH content in concretes at 28 days						
Mixtures	OPC-DSS	OPC-NSS	GGBS45-DSS	GGBS45-NSS		
CH content (%)	15.43	15.39	7.22	7.08		

according to the methodology [7].

(2) Mechanical properties of hardened concrete

Mechanical properties of all concrete mixtures including compressive strength and modulus of elasticity were determined according to JIS A 1108 and JIS A 1149, respectively. Uniaxial compression test was performed on cylindrical specimens at the ages of 28, 91 and 182 days. The value was obtained by the average of three measurements.

(3) Carbonation depth

At the designated ages, approximately 50-mm sample was split from the prismatic specimen along the longitudinal direction. The carbonation depth was measured by spraying phenolphthalein solution onto the freshly broken surfaces. The value was the average of ten measurements. On the other hand, the remaining part of the prismatic specimen was kept back in chamber after sealing with aluminum tape on the freshly broken surface for testing at later ages. The carbonation depth of all specimens was measured at 91 and 182 days.

(4) Scanning electron microscopy (SEM)

SEM analysis was used to strengthen the arguments provided in discussions. For conducting SEM test, the samples from the core of specimen under sealed condition were collected, whereas samples from carbonated part of prismatic fragment were collected to represent for carbonated area. Then, the samples were carefully crushed into approximate size of 5 mm and immersed in acetone and dried for 24 h before testing. After coating with platinum, an acceleration voltage of 15kV and magnification of 500 were used to characterize the morphology of samples.

3. RESULTS AND DISCUSSIONS

3.1 CH content in 28-day concrete

The CH content in concretes after sealing for 28 days is shown in Table 4. According to the results, the amount of CH in OPC concretes was two times higher than that in GGBS concretes. The lower CH content in GGBS concretes could be related to the dilution effect when GGBS was used to replace a part of OPC. In

addition, this decrease could be also attributed to the effect of hydraulicity of GGBS which consumed the CH in order to produce C–S–H gel [4, 8]. Moreover, the CH content in NSS concretes was slightly lower than DSS concretes, especially for GGBS mixture. The reduction in CH content can be explained by Eq. (1), which is the interaction between tricalcium aluminate (C₃A), Ca(OH)₂ and NaCl [9], leading to the formation of Friedel's salt (C₃A.CaCl₂.10H₂O).

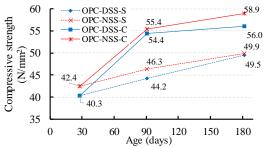
$$C_{3}A + Ca(OH)_{2} + 2NaCl + 10H_{2}O \rightarrow$$

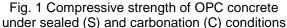
$$C_{3}A.CaCl_{2}.10H_{2}O + 2NaOH \qquad (1)$$

3.2 Compressive strength of concrete

The compressive strengths of the concretes under sealed and carbonation conditions are presented in Figs. 1 and 2. It is evident that all of the concretes exhibited an increase in the compressive strength with time regardless of sealed or carbonation condition. This increase is attributed to the continuous hydration of cement and hydraulicity of GGBS under sealed condition and carbonation. According to Figs. 1 and 2, the specimens using NSS generally achieved the higher compressive strength than those using DSS regardless of curing durations and conditions. For instance, the compressive strength of OPC concrete specimens using NSS was higher than those using DSS by 4.8 and 0.8% for sealed condition, and 1.8 and 5.2% for accelerated carbonation condition at the ages of 91 and 182 days, respectively (see Fig. 1). Similarly, as seen in Fig. 2, GGBS-NSS concrete showed higher compressive strength under sealed condition from 5.4 to 6.7% and that under carbonation condition approximately 2.2% in comparison with GGBS-DSS concrete at 91 and 182 days. It indicated that NSS could perform better in improvement of the compressive strength than DSS. This improvement can be attributed to the presence of chloride ion in NSS. The chloride ion in NSS can be bound by products of cement hydration and secondary hydration, resulting in the formation of Friedel's salt [9]. Friedel's salt can make concrete denser, leading to enhance the compressive strength of concrete.

On the other hand, the carbonation condition noticeably affected the strength development of concretes. It was found that the compressive strength of concretes under carbonation condition was higher than that of concretes under sealed condition irrespective of ages and GGBS replacement. Under sealed condition, the compressive strength of OPC concretes increased with the approximate ratio of 10% from 28 to 91 days. Meanwhile, the increase ratio of compressive strength of OPC concretes was 30% under accelerated carbonation. Moreover, the compressive strength of OPC concretes under accelerated carbonation was eventually higher from 13.1 to 18.0% in comparison to that of OPC concretes under sealed condition at 182 days. It can be concluded that the carbonation increased significantly the compressive strength of OPC concretes. Additionally, accelerated carbonation in chamber might reduce the moisture of specimens. Therefore, the decrease in moisture content led to the increase in compressive strength of concrete [10]. It is





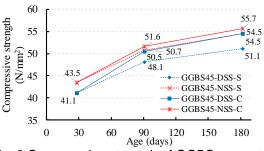
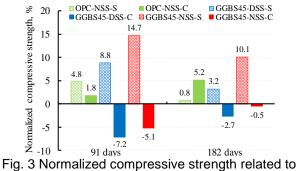
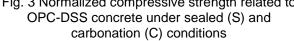


Fig. 2 Compressive strength of GGBS concrete under sealed (S) and carbonation (C) conditions





worthily mentioned that the exposure conditions affect significantly the compressive strength development of OPC and GGBS concretes. Fig. 3 shows the normalized compressive strength of all concrete specimens compared to that of OPC-DSS concrete under sealed and carbonation conditions. The compressive strength of GGBS concretes was higher from 3.2 to 10.1% than that of OPC-DSS concrete under sealed condition at 182 days. This is probably due to the hydraulicity of GGBS. CH which is produced during the hydration of cement reduces the bonding between aggregate and cement matrix. Therefore, for GGBS specimens under prolonger sealed curing, CH is transformed into secondary C-S-H gel as a result of hydraulicity, resulting in the refinement of pore system of concrete and the improvement of compressive strength [4]. On the contrary, the compressive strength of GGBS concretes was lower from 0.5 to 2.7% in comparison to that of OPC-DSS concretes under accelerated carbonation condition at 182 days. When concrete is exposed to carbon dioxide, the carbonation of CH and C-S-H seems to occur simultaneously following reactions (2) and (3):

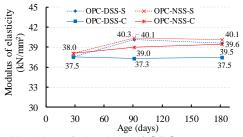
$$\begin{array}{ll} Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O & (2) \\ (CaO)_x(SiO_2)(H_2O) + xCO_2 \rightarrow xCaCO_3 + \\ SiO_2(H_2O)_t + (z-t)H_2O & (3) \end{array}$$

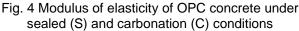
The precipitation of CaCO₃ leads to the reduction in porosity owing to the positive difference of molar volume between hydration products (CH and C-S-H) and CaCO₃ [6], resulting in the increase in the compressive strength of concretes. Therefore, the compressive strength of concretes under carbonation was higher than that of concretes under sealed condition, as seen in Figs. 1 and 2. This result is agreement with the report of Jerga [11]. On the other hand, according to Table 4, the amount of CH in GGBS concretes was significantly lower than that in OPC concretes because of the reduction of cement content and consequence of hydraulicity of GGBS. Therefore, the amount of CaCO₃ precipitation in OPC concretes was much higher than that of GGBS concretes. As a consequence, the compressive strength of OPC concretes was higher than that of GGBS concretes under carbonation condition.

3.3 Modulus of elasticity of concrete

The modulus of elasticity of all mixtures are plotted in Figs. 4 and 5. Against the results of the compressive strength, the modulus of elasticity of both OPC and GGBS concretes under sealed condition was higher than that of concretes under carbonation at the age of 182 days. In particular, for OPC concretes, the modulus of elasticity increased by approximately 5.5% for both cases of concretes using DSS and NSS under sealed condition from 28 to 182 days. The modulus of elasticity of NSS concrete slightly increased by 1.5% while that of DSS concrete stabilized after 182 days under accelerated carbonation condition. In contrast, the carbonation process led to the reduction of modulus of elasticity of GGBS concretes from 2.8 to 8.4% at 182 days. Meanwhile, the modulus of elasticity of GGBS concretes noticeably increased over 10% under sealed condition from 28 to 182 days. Similar to the tendency of the compressive strength, the modulus of elasticity of GGBS concretes was higher from 6.1 to 6.8% than that of OPC concretes under sealed condition at 182 days (see Fig. 6). Nevertheless, the result was completely reversed under carbonation condition. At 182 days, the modulus of elasticity of GGBS concretes was remarkably lower from 6.7 to 8.8% in comparison with OPC concretes under carbonation. In addition, the modulus of elasticity of OPC-NSS concrete was higher than that of OPC-DSS concrete regardless of ages and conditions. Meanwhile, the modulus of elasticity of GGBS concretes in both cases of DSS and NSS was only higher under sealed condition but was significantly lower under carbonation in comparison with that of OPC-DSS irrespective of ages.

The correlation between compressive strength and modulus of elasticity under sealed condition as well as carbonation condition was shown in Fig. 7. It is obvious that all moduli of elasticity obtained from experiment were approximately 20% higher than the recommendation of Japan Society of Civil Engineering





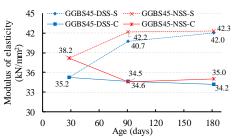


Fig. 5 Modulus of elasticity of GGBS concrete under sealed (S) and carbonation (C) conditions

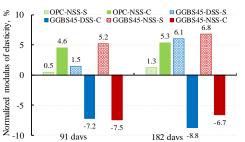


Fig. 6 Normalized modulus of elasticity related to OPC-DSS concrete under sealed (S) and carbonation (C) conditions

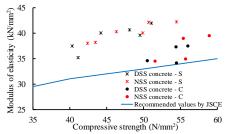


Fig. 7 Relationship between compressive strength and modulus of elasticity of concretes

JSCE 2017, except for carbonation condition. In addition, the modulus of elasticity of all concretes under sealed condition was higher than that of concretes under carbonation. These results of elastic modulus of OPC concretes are contrary with the report of previous studies [10, 12]. Chang et al. [12] demonstrated that the carbonation increased the modulus of elasticity of OPC concretes by 17.1%. The disagreement can be attributed to the difference in a water-to-cementitious materials ratio and carbon dioxide concentration between previous study and this study. In addition, the moisture of concrete specimen was reduced under accelerated carbonation. Shoukry at al. [10] reported that the moisture had an impact on the modulus of elasticity of concrete and the modulus of

elasticity of concrete increased with the moisture content decreasing. Therefore, it is necessary to conduct further experiment in order to explain the reason.

Generally, the main product of GGBS concrete is C–S–H while the productions of cement hydration are CH and C–S–H [4, 8]. According to the reference [6], when concrete is carbonated, both of reactions as shown in Eqs. (2) and (3) take place simultaneously. According to Table 4, the quantity of CH is dominant in OPC concretes. It means that the reaction in Eq. (2) seems to occur with priority over that in Eq. (3). However, the reaction (3) becomes more pronounced when concrete contains GGBS. The lower CH content as well as higher C-S-H content is attributed to vulnerably carbonate [8]. Therefore, the carbonation of C-S-H is believed to induce the formation of microcracks [13]. Consequently, the modulus of elasticity of GGBS concretes under carbonation significantly decreased compared to the corresponding specimens under sealed condition. This result is consistent with the conclusion of previous study [14].

Normally, it is well known that the formation of microcracks leads to the decrease in compressive strength and modulus of elasticity of concrete. When concrete is carbonated, the precipitation of CaCO₃ results in porosity clogging [6]. However, the occurrence of microcracks takes place simultaneously due to carbonation or drying shrinkage. It is assumed that the porosity clogging predominated microcracks. In other words, the microcracks were satisfactorily compensated by precipitation of CaCO₃. Therefore, the compressive strength of concretes still increased. On the other hand, because of extensive occurrence of microcracks during carbonation, the increase in deformation was significant for GGBS concretes. The changes in deformation were not satisfactorily compensated by CaCO₃ precipitation, leading to the decrease in modulus of elasticity of GGBS concretes.

3.4 Carbonation depth

The results of carbonation depth of all concrete specimens are presented in Fig 8. The percentage in the figure shows the difference of carbonation depth compared to OPC-DSS at the same ages. It can be seen that the NSS concretes showed slightly lower carbonation depth than DSS concretes regardless of ages and GGBS replacement. For OPC concrete, carbonation depth reduced by 5.3 and 8.3%. Meanwhile the carbonation depth of GGBS concrete could decrease by 12.9 and 5.4% at 91 and 182 days, respectively. The reason was attributed to the formation of Friedel's salt which was abovementioned in section 3.2. Friedel's salt can make concrete denser, leading to enhance the carbonation resistance. On the other hand, it is evident that the carbonation depth of concrete containing GGBS was much higher than that of the corresponding OPC concrete. Even when NSS was used to make GGBS concrete, its carbonation depth was also dramatically higher than that of OPC-DSS concrete up to 60.6 and 56.4% at 91 and 182 days, respectively. This is due to the lower CH content in GGBS concretes which was confirmed by the result in Table 4 [15]. Fig. 9 presents the interaction among GGBS replacement, compressive strength and carbonation based on the above discussion at the age of 182 days in comparison with OPC concretes. After 182 days of sealed condition, 45% of GGBS replacement slightly increased the compressive strength by 3-9%, but significantly increased the carbonation depth up to 65-71%. On the other hand, the increase in carbonation depth led to the

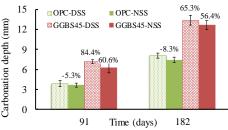


Fig 8. Carbonation depth of OPC and GGBS concretes

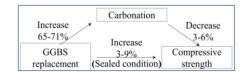


Fig. 9 The interaction among GGBS replacement, carbonation and compressive strength

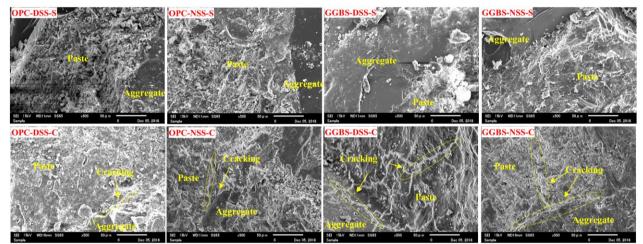


Fig 10. SEM images of OPC and GGBS concretes under sealed (S) and carbonation (C) conditions

reduction of compressive strength of concrete under carbonation by 3-6%. Therefore, regarding the durability of concrete using NSS, it is very necessary to consider carefully whether to use GGBS for replacing OPC because the carbonation can change the chloride ion behavior, leading to increase the possibility of steel bar corrosion [5, 9].

3.5 Scanning electron microscopy (SEM)

Fig. 10 shows the SEM images of OPC and concretes under sealed and carbonation GGBS conditions. GGBS concretes exhibited a denser microstructure than OPC concretes under sealed condition. The additional formation of C-S-H owing to the hydraulicity of GGBS filled up the large harmful pores, resulting in enhancement of the mechanical properties of concretes. On the other hand, the interfacial transition zone (ITZ) could not be easily distinguished as a consequence of carbonation. The large precipitation of CaCO₃ under carbonation made the microstructure of concretes significantly denser than that under sealed condition. However, it should be noticed that the microcracks also appeared around especially GGBS aggregate, concretes under carbonation. The microcrack was also observed by SEM [16]. Because the intention is to focus on an area where microcracks occurred, the magnification was small (×500). As a result, it was slightly difficult to see microcracks from Fig. 10. The occurrence of microcracks was attributed to drying or carbonation shrinkage [6].

4. CONCLUSIONS

- (1) The presence of chloride ion in NSS improved the compressive strength of concretes regardless of GGBS replacement and exposure conditions.
- (2) GGBS concretes exhibited higher compressive strength than OPC concretes under sealed condition. However, the contrast tendency was demonstrated under carbonation condition.
- (3) The chloride ion introduced by NSS could increase the carbonation resistance of concrete. Meanwhile, the GGBS replacement caused the reduction of the carbonation resistance of concrete, even in the case of NSS.
- (4) The carbonation significantly affected the modulus of elasticity of GGBS concrete. While hydraulicity of GGBS could continuously contribute to the increase in the modulus of elasticity of GGBS concretes, the carbonation decreased the modulus of elasticity of GGBS concretes, resulting from carbonation or drying shrinkage.

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