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Experimental Investigation on Diffusion Coefficient of Carbon Dioxide for Sustainable Construction Materials

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ABSTRACT

In order to achieve sustainability of construction materials, it is necessary to assess the resistance of materials to deteriorating factors. Carbonation is one of the important deteriorating mechanisms which arise in concrete due to environmental conditions. The purpose of this study is to first develop a measurement device for carbon dioxide diffusion coefficients and then to identify the diffusion coefficients of carbon dioxide for various concrete mixtures. To this end, several series of tests have been conducted. The test results indicate that the diffusion coefficient of carbon dioxide increases with an increase of water-cement ratio. It is also shown that the diffusion coefficient of carbon dioxide decreases with the increase of relative humidity at the same water-cement ratio. The quantitative values of diffusivity of carbon dioxide for various concretes are presented. The present study may be used to predict realistically the carbonation process in concrete structures under environmental conditions and further to assess the service life of such structures.

INTRODUCTION

Recently, sustainability of construction materials has become a prime issue in construction industry of civil structures. In order to achieve sustainability, resistance of construction materials to deteriorating factors such as chlorides and carbonation is very important. Since carbonation is one of the important factors that affect durability of concrete structures, it is necessary to evaluate correctly the carbonation process of concrete structures. The carbonation of concrete may accelerate corrosion process by reducing alkalinity of concrete and by destroying passivity film of reinforcing bars [Houst & Wittmann 1986, Loo et al. 1994]. The corrosion of steel bar may cause cracking of concrete cover and loss of load bearing capacity of concrete structures [Oh et al. 2003a, 2003b, 2004].

It is known that the carbonation depth with time is affected not only by the concentration of carbon dioxide, but also by the diffusivity of carbon dioxide. Since the concentration of carbon dioxide is usually constant in a specific environment, it is clear that reasonable determination of carbon dioxide diffusivity is one of the most important factors to predict realistically the carbonation depth of concrete structures. However, the standard test method of measuring the diffusion coefficient of carbon dioxide has not been established yet. Therefore, a reasonable measurement system is necessary to determine the diffusivity of carbon dioxide. Therefore, the purpose of the present study is first to propose a realistic test

method to measure the carbon dioxide diffusivity of concrete and then to explore the effects of various factors on the diffusivity of carbon dioxide in concrete.

The diffusion coefficient of carbon dioxide is influenced by several factors such as mixture proportions of concrete, relative humidity, and some other factors. Therefore, the effects of mixture proportion and relative humidity on the diffusivity of carbon dioxide were explored in this study. The measurement device for gas diffusivity in concrete materials developed in this study and also measured diffusivity data may contribute significantly to the realistic assessment of durability of concrete structures.

GAS DIFFUSION AND MEASURING DEVICE

Diffusion process of gas

It is generally known that molecular diffusion through porous materials follows the Fick's 1st law and Fick's 2nd law [Chatterji1994]. When the pressures and the flow rates of both sides in Fig. 1 are the same and a steady state condition is achieved, the diffusion coefficient of gas A in Fig. 1 may be expressed as follows [Chatterji1994, Kropp & Hilsdorf 1995].

$$D_A = \frac{Q f_A L}{(1 - f_A) A} \quad (1)$$

Where D_A = diffusion coefficient of gas A (m^2/s), Q = flow rate of gas B (m^3/s), f_A = molar ratio of gas A in gas B, L = thickness of specimen (m), and A = cross sectional area (m^3).

Gas diffusion measurement system

A diffusivity measurement system was developed in the present study as shown in Fig. 1. It was designed to measure gas diffusion coefficient when gas diffusion through concrete specimen reaches steady state. The measurement of diffusivity of carbon dioxide can be estimated according to time from the measurement of carbon dioxide concentration in the nitrogen located in the right cell of Fig. 1. The diffusivity measurement device of Fig. 1 was also designed to explore the effects of relative humidity and specimen thickness which may influence the gas diffusivity of concrete materials. Namely, the measuring equipment was designed to measure gas diffusion coefficient by controlling relative humidity of inflow gas and also by controlling the thickness of specimen from 5mm to 50mm. Fig. 1 shows the schematic diagram for gas diffusion measurement system and Fig. 2 exhibits specifically the diffusion cell in the measurement system. It is important to maintain the equivalent pressure and flow rate of both sides of Fig. 1.
The test procedures for measurement of gas diffusivity in concrete are summarized as follows.

(1) The measuring device should be installed indoors preserving temperature constant.

(2) The thickness and measurement area (or the diameter of measurement) of the specimen are measured by calipers.

(3) Install concrete specimen inside the diffusion cell as shown in Fig. 2**.**

(4) The carbon dioxide gas is injected into the one side of the specimen (left side) and the nitrogen gas into the other side (right side) of the specimen with same pressure constantly.

(5) The desired relative humidity of inflow gas should be set by controlling both the dry gas from silica gel and the saturated gas from saturator (see Fig. 1).

(6) When the concentration of carbon dioxide in the nitrogen gas (in right cell) becomes constant according to time, it is judged that the steady state has been reached. Then, one measures the concentration of carbon dioxide in the nitrogen gas.

(7) Calculate the diffusion coefficient based on Eq. (3).

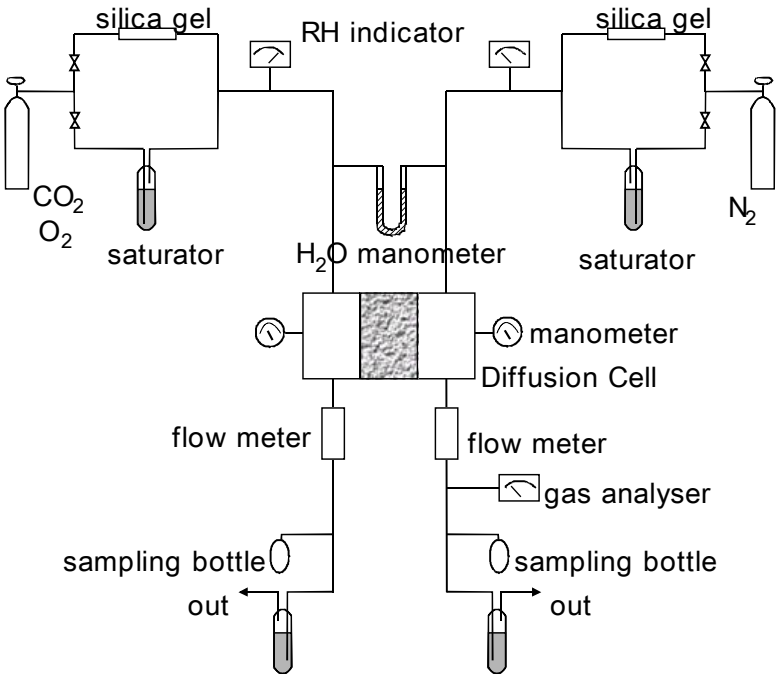


Fig. 1. Developed Gas Diffusion Measurement System

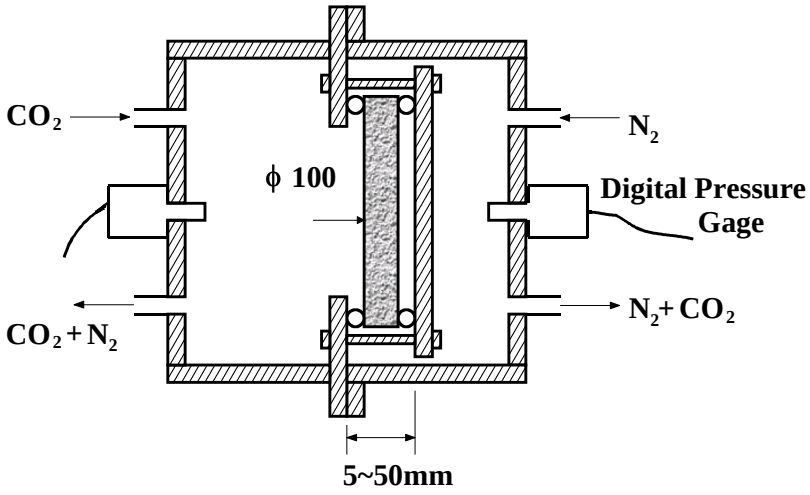


Fig. 2. Gas Diffusion Cell in Fig. 1

EXPERIMENTAL PROGRAM

Test Materials and Mixture Design. The absorption rates for fine and coarse aggregates were 2.18 and 0.94 percent, respectively. The specific gravities of fine and coarse aggregates used in this study were 2.56 and 2.60, respectively. The fineness moduli of fine and coarse aggregates were 2.85 and 6.51, respectively. Table 1 shows the mixture designs for concretes used in the present study.

Table 1. Mixture Design for Tested Concretes

W/C	Cement $kg/m^3(lb/yd^3)$	Water $kg/m^3(lb/yd^3)$	Fine agg. $kg/m^3(lb/yd^3)$	Coarse agg. $kg/m^3(lb/yd^3)$
0.50	315(531)	158(266)	748(1260)	1076(1813)
0.58	277(467)	161(271)	726(1223)	1117(1882)

Test Variables and Test Specimens. Major test variables here are water-to-cement ratio and relative humidity. The water-to-cement ratios considered in the present study were 0.50 and 0.58, respectively. The relative humidity ranged from 10 to 90%. The test materials were mixed by mechanical mixer and the molds were removed after 24 hours from mixing. Then, the test specimens were cured in a water tank with the temperature of 20±2℃ for 28 days. The diameter of test specimen was 100mm and thickness was 10mm. The 10mm thickness specimens were obtained by slicing the concrete cylinders of size 100mm X 200mm. The sides of specimens were coated with epoxy for one dimensional flow for diffusion tests.

For concrete specimens, diffusivity tests for carbon dioxide were also performed for completely-carbonated specimens in order to explore the effect of carbonation on the diffusivity. The carbonation of concrete specimen was done artificially by using a carbonation chamber. For the purpose of measuring the diffusion coefficient according to relative humidity, the specimens were placed in vacuum desiccators which remain in constant relative humidity until the specimens approach constant weight. This process was done according to the guideline ASTM E104 (ASTM 1996).

ANALYSIS OF TEST RESULTS

Determination of Time to Steady State Conditions. Fig. 3 shows the volume fractions in percent of carbon dioxide passed through non-carbonated concrete specimens according to time for water-to-cement ratio w/c=0.5. Fig. 3(a) represents the case of relative humidity RH=10% and Fig. 3(b) the case of relative humidity RH=75%, respectively. It can be seen from Fig. 3 that the diffusion of carbon dioxide reaches the steady state within about five hours after exposure for non-carbonated concrete specimens.

Fig. 4 shows the volume fractions of carbon dioxide passed through carbonated concrete specimens according to time. It is again seen from Fig. 4 that the diffusion of carbon dioxide reaches the steady state within about two hours after exposure for carbonated concrete specimens. It is noted here that the time to steady state condition is somewhat shorter for carbonated concrete specimens than for the case of non-carbonated concrete specimens.

Influence of water to cement ratio

Fig. 5 shows the influence of water to cement ratio on the diffusion coefficient of carbon dioxide in concrete. The test results indicate that the diffusion coefficient increases with an increase of water to cement ratio. This is especially more pronounced when relative humidity is very small as shown in Fig. 5.

However, the ratio of increase of diffusion coefficients according to water to cement ratio becomes small when the relative humidity is high. The reason may be that the variation of pore structures caused by the change of water to cement ratio has little effect on the diffusion of carbon dioxide in concrete under high humidity because most of pores are filled with water when relative humidity is high.

Influence of Relative Humidity. Fig. 6 shows the variation of diffusion coefficients according to relative humidity for concrete specimens with different water-cement ratios. It is shown that the diffusion coefficient of carbon dioxide decreases with an increase of relative humidity for all concrete specimens.

It is interesting to note here that the decrease of diffusion coefficient of carbon dioxide according to relative humidity becomes smaller when the water to cement ratio becomes lower. This reason may be explained such that the open pores, which make it possible to diffuse gas, are relatively small due to dense matrix of concrete with low water to cement ratio and thus the influence of relative humidity on diffusion coefficient becomes small when the water to cement ratio is low.

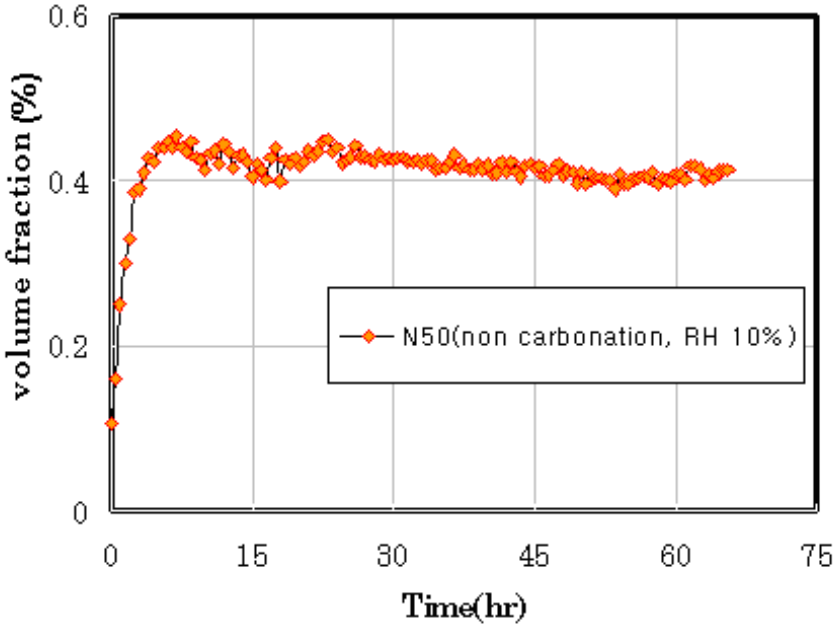
Influence of Carbonation. Fig. 7 shows the comparison of diffusion coefficients for non-carbonated and carbonated concrete specimens for water to cement ratio of 0.58. It can be seen from Fig. 7 that the diffusion coefficients of carbon dioxide of carbonated concrete are slightly lower than those of non-carbonated concrete. The present study indicates that the effect of carbonation on diffusion coefficient is small for concrete with relatively high water to cement ratio. This may be due to relatively large pore structures for concrete with high water to cement ratio. Fig. 7 also indicates that the effect of relative humidity on diffusion coefficient of carbon dioxide is very much similar for both non-carbonated and carbonated concrete specimens.

SUMMARY AND CONCLUSIONS

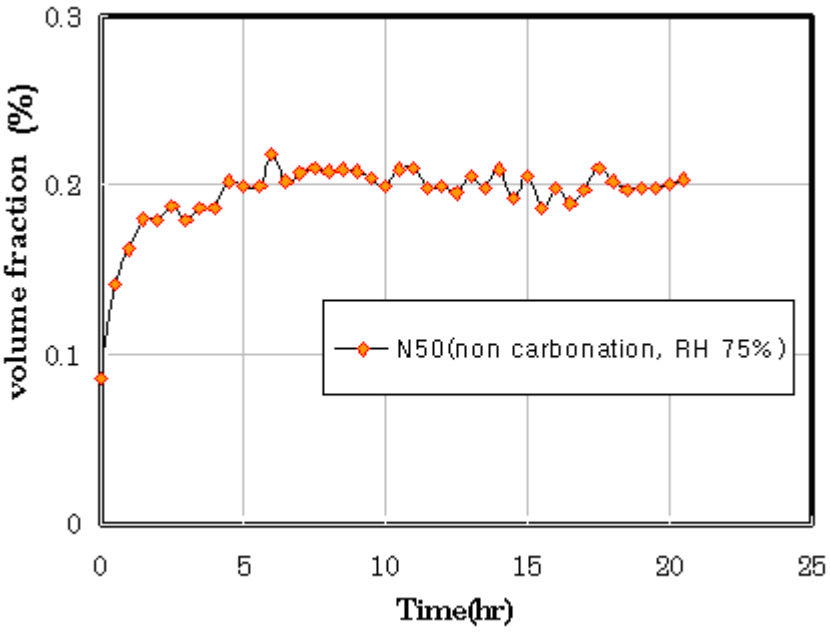
The purpose of the present study is to explore the diffusion coefficient of carbon dioxide in concrete. For this purpose, a realistic measurement system for gas diffusion coefficient was developed in the present study. The following conclusions were drawn from this study.

(1) The diffusion of carbon dioxide reached the steady state within about five hours after exposure for non-carbonated concrete specimens and within about two hours after exposure for carbonated concrete specimens.

(2)The present test results indicate that the diffusion coefficient of carbon dioxide increases with an increase of water to cement ratio. This is especially more pronounced when relative humidity is small.

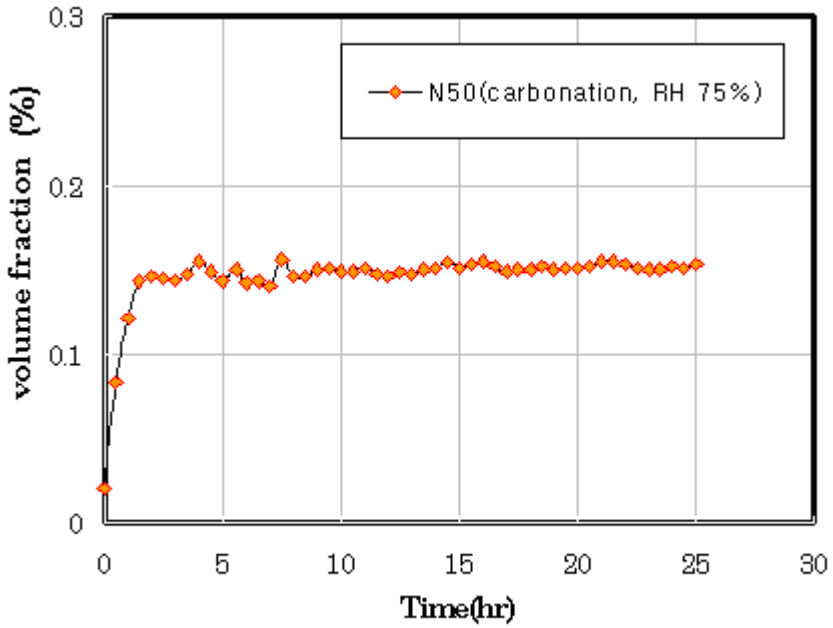


(a) Non-carbonated concrete specimen (w/c=0.5, RH=10%)

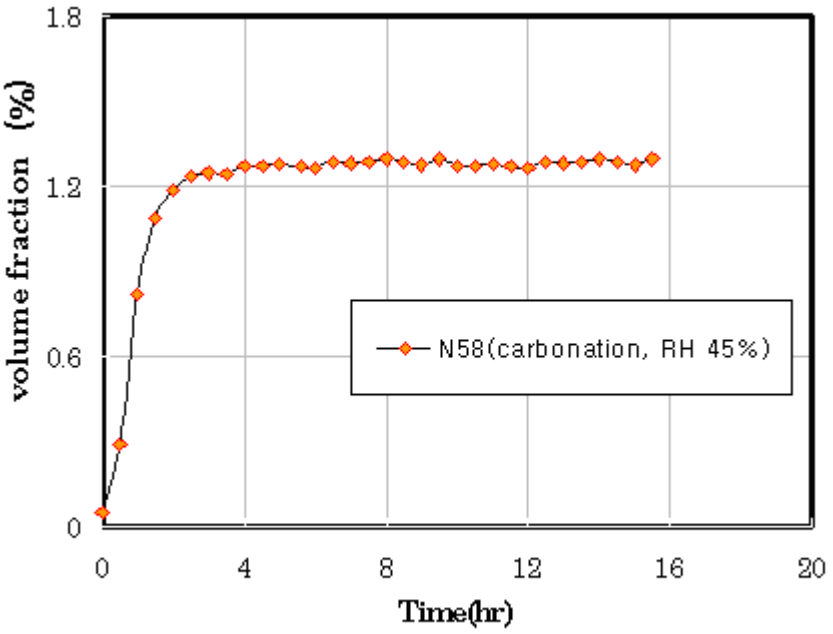


(b) Non-carbonated concrete specimen (w/c=0.5, RH=75%)

Fig. 3. Volume Fractions in Percent of Carbon Dioxide passed through Non-carbonated Concrete Specimens According to Time.



(a) Carbonated concrete specimen (w/c=0.5, RH=75%)



(b) Carbonated concrete specimen (w/c=0.58, RH=45%)

Fig. 4. Volume fractions of carbon dioxide passed through carbonated concrete specimens according to time.

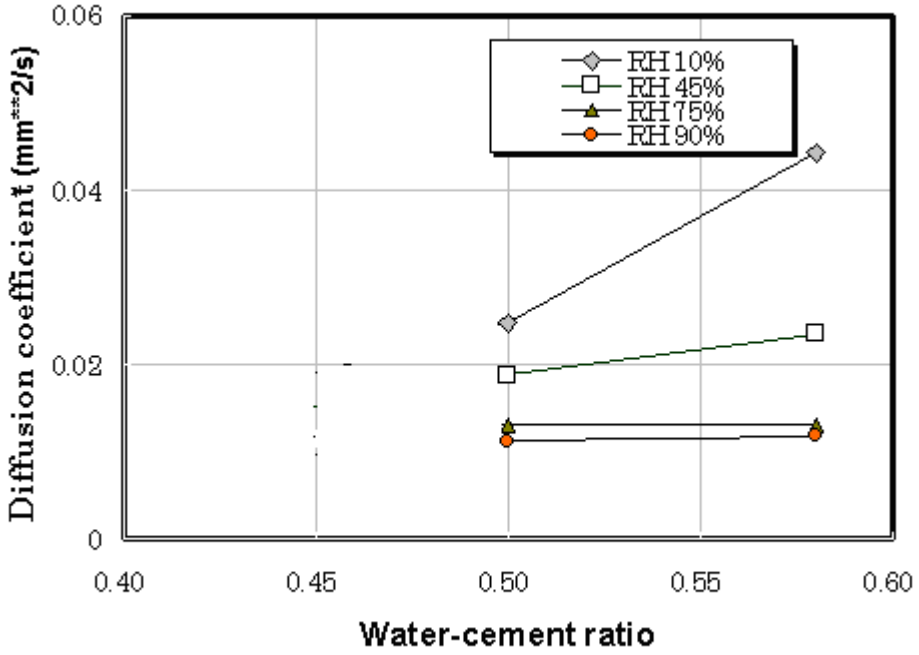


Fig. 5. Comparison of Diffusion Coefficients According to Water to Cement Ratio

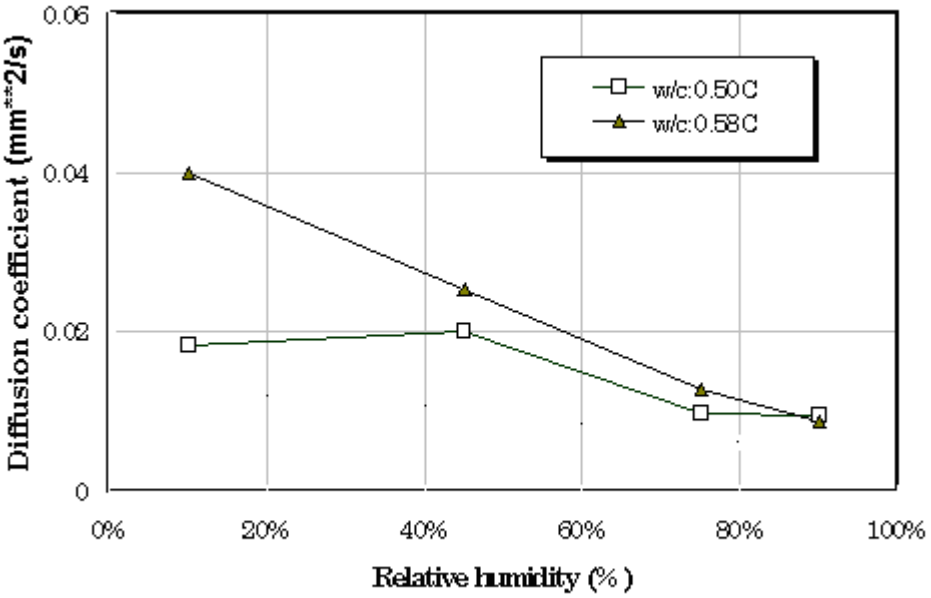


Fig. 6. Comparison of Diffusion Coefficients for Concrete Specimens according to relative humidity

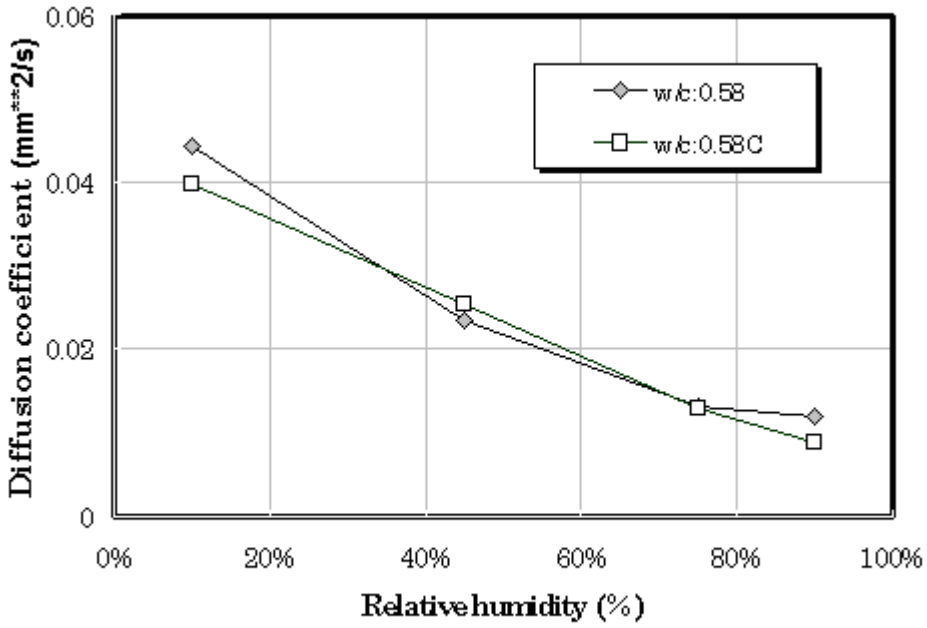


Fig. 7. Comparison of diffusion coefficients for non-carbonated and carbonated concrete specimens

(3) The ratio of increase of diffusion coefficients according to water to cement ratio becomes small when the relative humidity is high. This is probably due to the fact that the variation of pore structures caused by the change of water to cement ratio has little effect on the diffusion of carbon dioxide in concrete under high relative humidity because most of pores are filled with water when relative humidity is high.

(4) The present study indicates that the diffusion coefficient of carbon dioxide decreases with an increase of relative humidity for all concrete specimens. However, the decrease of diffusion coefficient of carbon dioxide according to relative humidity becomes smaller when the water to cement ratio becomes lower. The reason may be due to the fact that the open pores, which make it possible to diffuse gas, are relatively small due to dense matrix of concrete with low water to cement ratio and thus the influence of relative humidity on diffusion coefficient becomes small when the water to cement ratio is low.

(5) It is shown that the diffusion coefficients of carbon dioxide of carbonated concrete are somewhat lower than those of non-carbonated concrete. The present study also indicates that the effect of carbonation on diffusion coefficient is small for concrete with relatively high water to cement ratio.

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