- Technical Paper -

FUNDAMENTAL STUDY ON WATER DIFFUSION COEFFICIENT OF CEMENT BASED MATERIAL

Mao LIN*1, Hiroshi SASANO*2 and Ippei MARUYAMA*3

ABSTRACT
Drying experiments of both cement mortar and concrete specimens have been done to evaluate water movement behavior in cement based material. Boltzmann-Matano method is applied to the experimental data to calculate the water diffusion coefficient. Temperature dependency of water diffusion coefficient and effect of aggregate size on water diffusion coefficient are experimentally evaluated. As a result, temperature dependency of water diffusion coefficient is governed by power law and the aggregate size doesn't have much impact on water diffusion coefficient.
Keywords: Water diffusion coefficient, water movement, drying, high temperature, aggregate size

1. INTRODUCTION

Water movement in concrete structures is one of the most important topics for their durability and service life. Ordinary concrete, whose water to cement ratio is ranging from 0.40 to 0.60, contains much water than the water amount cement reaction demands, therefore, the water in concrete may evaporate to surrounding air if the environment of concrete structure is dry. This water loss firstly causes the insufficient cement reaction at the surface of concrete if the demoulding timing is not enough. The insufficient cement reaction yields coarse cover concrete which enhances penetration of substances relating to rebar corrosion. Continuous water evaporation is strongly related to the drying shrinkage and resultant induced stress and cracking of concrete. Shrinkage induced cracking could be through cracking or surface cracking. Though the roles of such cracking on the durability are not the same, they anyway jeopardize the durability of concrete. In addition to these effects, water content distribution in concrete member has an important impact on movement of chloride ions and carbon dioxides.

Based on this important knowledge, the study of water movement in concrete is still needed for design and maintenance of concrete structure for long-term service periods.

Experimental study of water movement is firstly developed by Sakata [1]. He used Boltzmann-Matano method for obtaining the relation of water content and water diffusion coefficient. The difficulty of water movement phenomenon in concrete is that it takes much time to obtain equilibrium than those of other porous materials. Therefore, compared to other methods, Boltzmann-Matano method has a benefit regarding cement based material research. The dependency of water diffusion coefficient on the water content is indicated by numerical approach of Bazant [2].

After Sakata's research, Akita and Fujiwara did comprehensive research on water movement [3] [4] [5]. They used different approaches to obtain the relationship between water content (or relative water content) and water diffusion coefficient, and obtained consistent results to those by Sakata. In addition to these results, they found the temperature dependency of water diffusion coefficient, water diffusion coefficient in very low water content region, and water diffusion coefficient of desorption and adsorption processes.

In the present research, effects of temperature and aggregate size on the water diffusion coefficient are focused and reproducibility by comparison with former researches is discussed.

2. EXPERIMENT

2.1 Temperature Dependency of Water Diffusion Coefficient in Cement Mortar

In this study, normal Portland cement (marked as N) is used. The properties of cement are shown in Table 1. The fine aggregate is standard sand. Its density is 2.64g/cm^3. The water absorption of the sand is 0.42% and the maximum grain size is 2mm. Water-cement ratio is 0.55. The cement to sand ratio is 1:1.8. The size of the specimen is 200×44×10mm. The specimens were demoulded 3 days after casting and cured in water in the room with temperature 20±1℃. To ensure stable pore structures of the cement mortar, the experiment of water diffusion coefficient evaluation started after the age of cement mortar specimens is more than 300 days. In that case, hydration process of mortar has fully

*1 Graduate Student, Graduate School of Environmental Studies, Nagoya University, JCI Member
*2 Graduate Student, Graduate School of Environmental Studies, Nagoya University, JCI Member
*3 Associate Professor, Graduate School of Environmental Studies, Nagoya University, JCI Member

Table 1 Cement properties

Cement Type	Density (g/cm^3)	Specific Surface (cm^2/g)	LOI (%)	Chemical Composition (mass %)								
				SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	K_2O	Cl^-
N	3.16	3110	0.64	21.8	4.49	2.90	63.9	1.84	2.26	0.20	0.38	0.007

completed, so the effect of hydration could be ignored.

In this experiment, 5 sides of specimens were sealed with aluminum tapes, while one side was not sealed (as shown in Fig. 1). The moisture diffusion becomes a one-dimension problem. The specimens were then placed in temperature and humidity control chamber for drying. The chamber temperatures were 20℃, 40℃ and 55℃ respectively. The 20℃ chamber used silica desiccant for drying and humidity was kept below 15%.

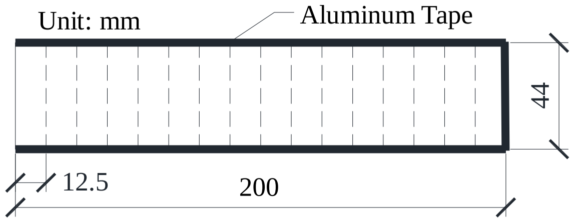

Fig. 1 Mortar specimens dimensions

To evaluate the water diffusion coefficient, the specimens were dried for 7, 14 and 28 days, respectively. Under each drying temperature, 3 specimens were used for each drying period. Water content distribution is based on mass loss of specimens over time at different depth from the drying surface. Fujiwara [4] proposed 3 drying approaches: 1) pre-cutting, 2) cutting after drying, 3) using specimens with different length. In this experiment, the 2nd approach cutting after drying was used. After drying, the specimens were cut into 16 pieces. Each piece was weighed after cutting, and then dried in the 105℃ chamber for 24 hours to get its absolute dry weight. The weight difference is the water content of each piece of mortar specimen. In this experiment, electronic scale with accuracy of 0.0001g was used for measuring weight.

Furthermore, to evaluate the sealing condition, total 6 sides sealed specimens were weighed over time. The weight changes of 28 days were 0.559%, 0.202% and 1.052% under the temperature 20℃, 40℃ and 55℃ respectively. The results show that aluminum tapes provide a good sealing condition.

The drying process of cement mortar is one-dimension diffusion problem and diffusion equation is applicable here. In this study, Boltzmann-Matano method is applied to the measured data. Water diffusion coefficient $D(w)$ (m^2/s) is the function of water content.

$$\lambda = \frac{x}{\sqrt{t}} \quad (1)$$

$$D(w) = -\frac{1}{2}\int_{w_0}^{w} \lambda dw \bigg/ \frac{\partial w}{\partial \lambda} \quad (2)$$

where,

λ : Boltzmann variable ($m/s^{1/2}$)
x : distance from drying surface (m)
t : drying time (s)
w_0 : saturated water content (g/m^3)
w : water content at any time (g/m^3)

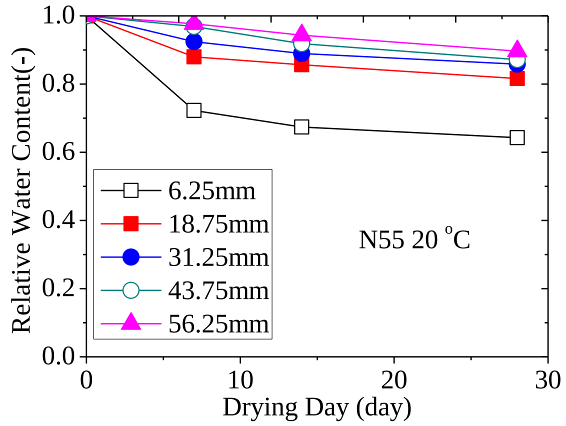

Fig. 2 Relative water content at 20℃

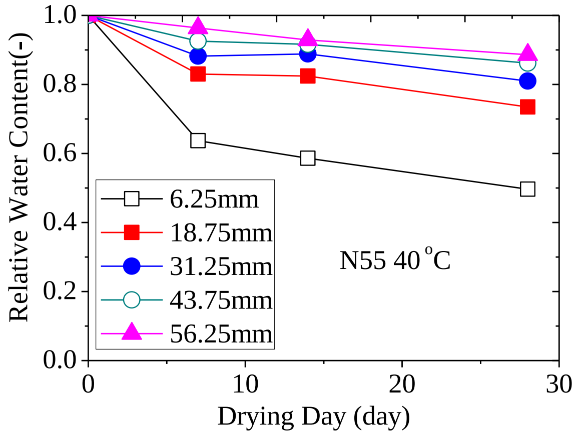

Fig. 3 Relative water content at 40℃

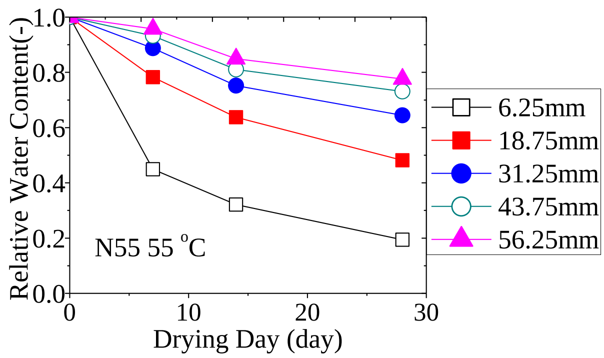

Fig. 4 Relative water content at 55℃

The relationship between λ and w could be obtained by the regression of the experiment data to solve the Eq. (2). According to the previous studies, the fitting function from Akita [3] is used for the regression in this study.

$$w = w_0\left(1 + f - \frac{a}{(\lambda/2+b)^2}\right) \quad (3)$$

where, a, b, f are regression constants derived from the regression of experimental data by least square fit. In this study, relative water content R ($R=w/w_0$) is used.

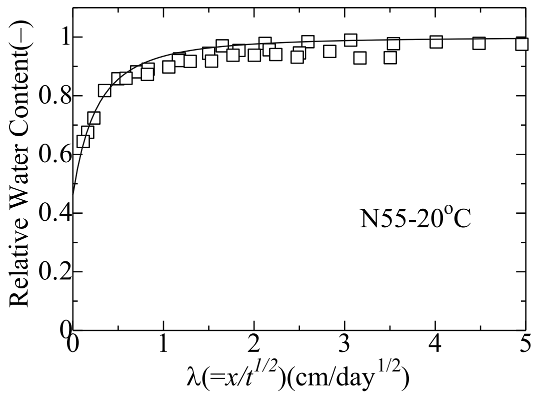

Fig. 5 Relationship of R and λ at 20℃

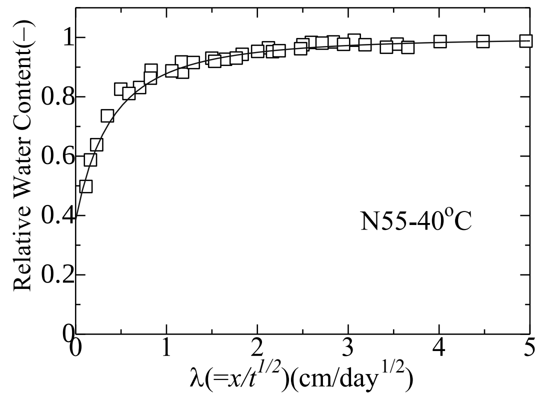

Fig. 6 Relationship of R and λ at 40℃

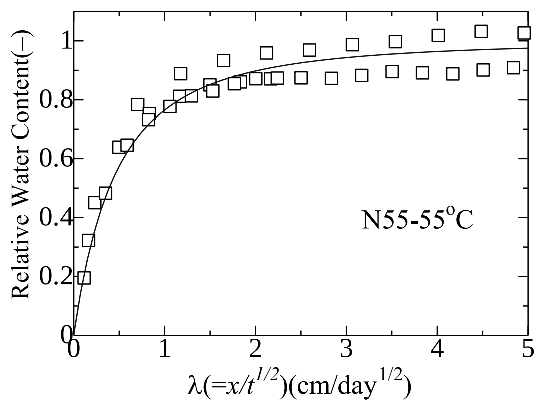

Fig. 7 Relationship of R and λ at 55℃

Fig.2~4 show the relative water content change of the specimens over time at different distance from the drying surface under the ambient temperature of 20℃, 40℃ and 55℃ respectively. It is clear that the drying process is fast at 6.25mm from the drying surface and slows down with increasing distance from the drying surface. The temperature dependency of drying process is also obvious. The specimens dried at higher temperature have much more water loss.

Boltzmann-Matano method is used to calculate the water diffusion coefficient. The relationship of relative water content R and Boltzmann variable λ is obtained by regression of the experimental data using the fitting function Eq. (3). Fig. 5~7 show the experimental data and the regression curve. The results of regression are listed in Table 2.

Table 2 Constants of Eq. (3)

Temperature	a	b	f
20℃	0.03859	0.2678	0.0001
40℃	0.09866	0.4004	0.0001
55℃	0.2213	0.4712	0.0001

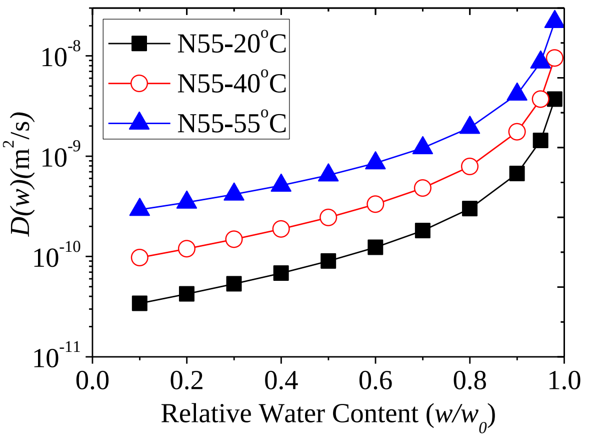

Fig. 8 Water diffusion coefficient of mortar

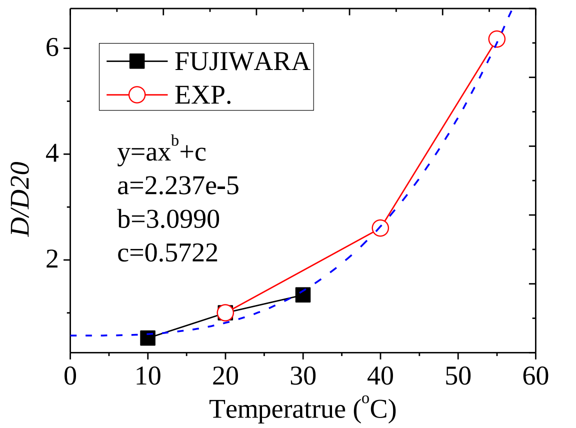

Fig. 9 Relation between temperature and *D/D20*

Based on the relation function of λ *and* R obtained by regression, the water diffusion coefficient $D(w)$ was calculated by Eq. (2). The results are shown in Fig. 8. High water content corresponds with large diffusion coefficient. The diffusion coefficient increases sharply when the relative water content is larger than 90%. The diffusion coefficient at low water content range is comparatively constant. This tendency is

Table 3 Mix proportion of concrete

Water-Cement Ratio	Fine Aggregate Ratio	Mass (kg/m^3)			
		Water	Cement	Sand	Coarse Aggregate
0.55	0.43	185	336	783	1026

almost the same as the research of Sakata [1] and Akita [3]. The water diffusion coefficient is not only affected by water content but also affected by temperature. The higher the temperature is, the larger the diffusion coefficient is.

The relative water diffusion coefficient *D/D20* of this experiment at the relative water content of 90% is compared to Fujiwara's paper [5], where *D20* is the water diffusion coefficient at 20℃. The result is shown in Fig. 9. Temperature dependency of water diffusion coefficient has been confirmed. The temperature has more influence on water diffusion coefficient in high temperature range. The relationship of temperature and D/D20 is governed by a power-law function by fitting the experiment data. The function is shown in Fig. 9.

2.2 Effect of Aggregate Size on Water Diffusion Coefficient

In this experiment, normal Portland cement is used. The coarse aggregate is diorite crushed stone. The density in saturated surface-dry condition is 2.95g/cm^3, and the water absorption is 0.48%. The maximum size of coarse aggregate is 20 mm. The fine aggregate is crushed sand. The water absorption of the sand is 2.34%, and the maximum grain size is 5mm. The mix proportion is listed in Table 3. The size of concrete specimens is ϕ 100×300mm. The specimens were demoulded 2 days after casting and then cured in water. The concrete drying experiment started at the age of 100 days. In the drying experiment, the temperature is 20℃ and the humidity is 40%.

In the drying experiment of concrete specimens, the approach of precutting is used. According to Fujiwara [4], precutting has a little lower speed of drying, but the tendency of water content distribution from the drying surface is almost the same. Furthermore, the cutting surface has very small effect on water movement inside concrete specimens. As the water loss is larger near the drying surface and much less deep inside the specimens, the specimens were cut into 6 segments with different length while the segments near the drying surface have smaller thickness (as shown in Fig. 10).

3 specimens were used in this experiment. The specimens were cut into 6 segments and their saturated weights in surface dry condition were measured. Those segments of specimens were then combined for drying. The side and bottom of the column were sealed with aluminum tapes and the top surface was exposed to surroundings (as shown in Fig. 10). The weight of each segments were measured by electronic scale with accuracy of 0.01g over 1, 3, 7, 14 and 27 days, respectively. In the end of the experiment, those specimens were dried in 105℃ chamber for 7-10 days to get their absolute dry mass. Then the water content over time at different depth of specimens could be calculated. Fig. 11 shows the relative water content change of the concrete specimens. The tendency is almost the same as the cement mortar drying experiment. At the depth of 5mm from the drying surface, the water loss is the most drastic. After the depth of 55mm, the drying process becomes rather slow.

Boltzmann-Matano method is used to calculate the water diffusion coefficient. The relationship of relative water content R and Boltzmann variable λ is obtained by regression of the experimental data using the fitting function Eq. (3). The result is shown in Fig. 12. And then water diffusion coefficient at different water content is calculated by Eq. (2). Then the water diffusion coefficient of these concrete specimens was compared to the results of Sakata [1] and Akita [6]. The result is shown in Fig. 13. The drying curve of this experiment is similar to Akita's experiment and also in the same magnitude of Sakata's experiment. It shows that the water diffusion coefficient in this experiment has the same tendency with the results of Sakata and Akita.

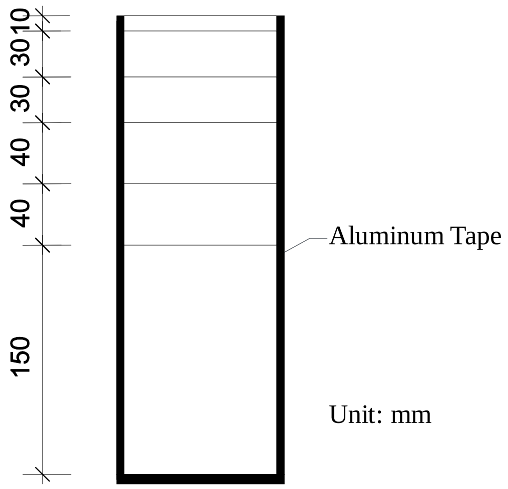

Fig. 10 Concrete specimens dimensions

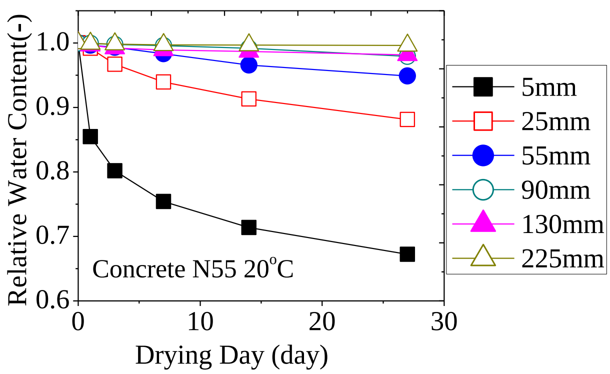

Fig. 11 Relative water content of concrete

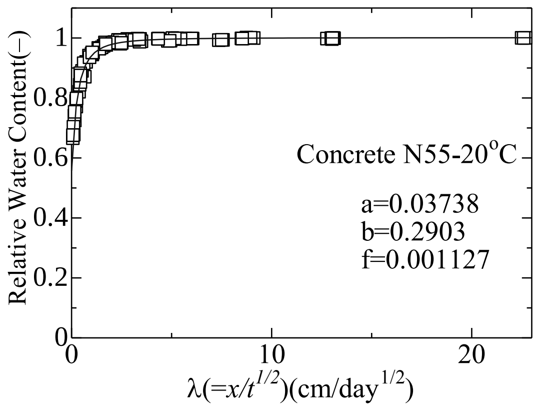

Fig. 12 Relationship of *R* and *λ* of concrete

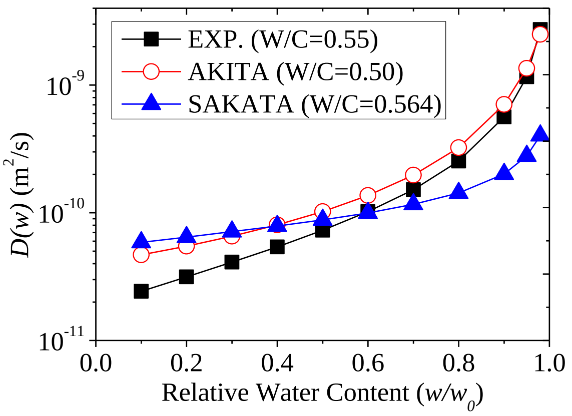

Fig. 13 Water diffusion coefficient of concrete

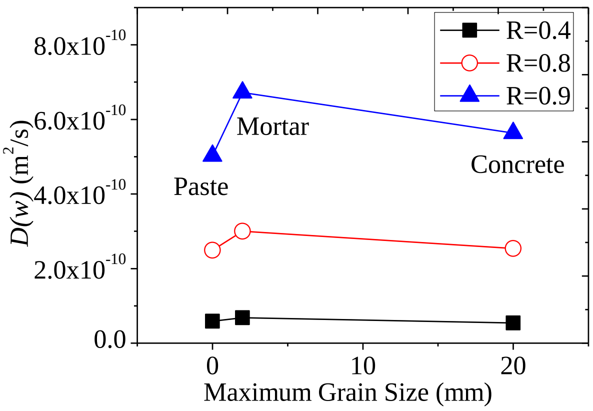

Fig. 14 Relation of maximum grain size and *D(w)*

The influence of aggregate size on water diffusion coefficient is also evaluated. The concrete drying experiment is compared to cement paste [7] and mortar specimens drying experiment which have the same water-cement ratio (*W/C*=0.55) and the same drying temperature (20℃). Those types of cement based material have different aggregate sizes. Concrete has the maximum grain size of 20mm, while the maximum grain size of mortar and paste is 2mm and 0mm, respectively. The relation of *D(w)* and the maximum grain size at different relative water content is shown in Fig. 14. In this comparison, mortar has the largest water diffusion coefficient, while concrete and paste are similar. Clear relationship between aggregate size and water diffusion coefficient has not been observed in this study. It is well known that interfacial transition zone, which is including micro-cracking around aggregate, induces water transfer. But when the large size of aggregate is used in concrete, the pass of water movement is expanded, and resultantly, water diffusion coefficient of concrete becomes smaller than that of mortar.

3. CONCLUSIONS

In this study, drying experiment of both cement mortar and concrete has been conducted.

(1) According to the drying experiment of cement mortar specimens under different ambient temperature, temperature dependency of water diffusion coefficient (WDC) is confirmed. The higher temperature shows larger WDC. The temperature dependency of WDC is governed by power law.

(2) The WDC curve, which is a function of relative water content, obtained in the present drying experiment of concrete reproduces the existing research data.

(3) In the limit of the present research, there is no aggregate size dependency of WDC generally. The WDC of mortar is the largest and concrete is the second. Paste sample shows the minimum value of water diffusion coefficient. The increase of WDC in mortar is explained by the more interfacial transition zone (ITZ) than those in cement paste. Concrete has the same ITZ effect, but due to the size of aggregate grain, the water diffusion pass is extended. This could be the reason that the WDC of concrete is less than that of mortar.

REFERENCES

[1] Sakata, K., "A Study on Moisture Diffusion in Drying and Drying Shrinkage of Concrete", Cement and Concrete Research, Vol. 13, Mar. 1983, pp. 216-224

[2] Bazant, Z.P. and Najjar, L.J., "Nonlinear Water Diffusion in Nonsaturated Concrete", Materiaux et Constructions, Vol.5, 1972, pp.3-20

[3] Akita, H., Fujiwara, T. and Ozaka, Y., "Water Movement within Mortar due to Drying and Wetting", Proceedings of JSCE, No. 420, 1990, pp. 61-69 (In Japanese)

[4] Fujiwara, T., Sato, K. and Konishi, T., "Water Movement within Concrete due to Drying and Wetting", Review of 42 General Meeting, CAJ, 1988, pp. 427-430 (In Japanese)

[5] Fujiwara, T., Kayaba, M. and Akita, H., "Water Movement within Concrete due to Drying under Various Temperature Conditions", CAJ Proceedings of Cement & Concrete, No.46, 1992, pp.428-433 (In Japanese)

[6] Akita, H., Fujiwara, T., and Ozaka, Y., "An Analytical Method of Moisture Transfer within Concrete due to Drying", Proceedings of JSCE, No. 490, 1994, pp.101-110 (In Japanese)

[7] Maruyama, I., Igarashi, G. and Kishi, N., "Fundamental Study on Water Transfer in Portland Cement Paste", Journal of Structural and Construction Engineering. Transactions of AIJ, No. 668, vol.76, Oct. 2011, pp.1737-1744 (In Japanese)