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MOISTURE DISTRIBUTION, DIFFUSION COEFFICIENT AND SHRINKAGE OF CEMENT-BASED MATERIALS

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The purpose of this study was to develop a new method for obtaining diffusion, film and shrinkage coefficients of cement-based materials. These coefficients are required for the numerical simulation of the effect of shrinkage strain on the deformation of concrete structures using finite element method. An experimental approach for obtaining the relative humidity distribution in the specimen at arbitrary drying times and a numerical method for determining the material coefficients from the experimental data are proposed in this paper. Results show that this method can provide us with the diffusion coefficient not only as a function of moisture content but also as a function of relative humidity in cement-based materials.

Key Words: diffusion coefficient, relative humidity distribution, drying shrinkage strain, inverse analysis, sliced specimen

1. INTRODUCTION

Shrinkage strain develops with moisture loss from cement-based materials such as concrete. The strain therefore develops much quicker near the drying surface than in the center of concrete. If a concrete member consisted of separate elements whose thickness were infinitesimal and unrestrained, the relationship between the change of moisture and the deformation of each element would be linear[1)]. In a real concrete member, the strain due to quick drying near the surface as compared to the inside of the concrete produce tensile and compressive elastic strains due to eigenstresses. In order to simulate numerically the effect of drying on the deformation of concrete, we need the diffusion, film and shrinkage coefficients. By using both the diffusion coefficient and film coefficient, the moisture distribution at any arbitrary drying time can be obtained. The unrestrained drying shrinkage strain is calculated by multiplying the change of moisture by the shrinkage coefficient. Finally, the deformation can be calculated according to the principle of virtual work.

The diffusion coefficient of concrete is nonlinear and depends on the moisture content itself. If the diffusion coefficient can be expressed in terms of the relative humidity of the concrete, the analysis of drying is absolutely the same as that for the thermal case. The ambient relative humidity can be used as the boundary condition in a numerical analysis.

To obtain the nonlinear diffusion coefficient, various methods have been proposed by different researchers[2),3)]. However, they are for the nonlinear diffusion coefficient in terms of moisture content not in terms of relative humidity. To determine the diffusion coefficient as a function of the pore equilibrium relative humidity of concrete, the relationship between moisture content in concrete at hygral equilibrium and the relative humidity, i. e. the desorption isotherm, which varies with ambient temperature, must be known. The desorption isotherm determined by experiments is available for some cases for cement-based materials. But, it is not certain if they are available for any concrete. Indeed, it takes a lot of time to determine the desorption isotherms by experiment. A method of obtaining the diffusion coefficient as a function of relative humidity directly by experiment is desirable.

A useful and reliable method of obtaining the diffusion coefficient as a function of relative humidity is proposed in this paper. In the experiment, sliced specimens of 150x100x3 mm are used. Each specimen

Table 1 Mix proportions of mortar and concrete

Name of Mixture	Gmax (mm)	W/C (%)	s/a (%)	Unit weight per volume (kg/m³) W	C	S	G 4~8	G 8~16	*1
M240	4	50	100	240	480	1536	–	–	–
M200	4	50	100	200	400	1708	–	–	–
M160	4	50	100	156	320	1880	–	–	3.84
G8	8	50	50	200	400	854	880	–	–
G16	16	50	50	200	400	854	352	528	–

*1: Superplasticizer

Table 2 Mechanical properties of mortar and concrete

Name of Mixture	Age (days)		Comp. Strength (MPa)	Young's Modulus (GPa)	Flexural Strength (MPa)
M240	14		33.6	25.8	6.16
	120	Air	44.4	26.3	8.38
		Water	45.4	30.3	8.53
M200	14		32.1	24.8	5.10
	120	Air	41.5	27.4	7.84
		Water	44.3	29.3	8.33
M160	14		19.2	22.1	4.16
	120	Air	21.5	19.6	4.61
		Water	29.6	19.4	4.69
G8	14		42.0	31.8	6.74
	120	Air	48.3	36.7	10.1
		Water	52.8	39.5	10.3
G16	14		36.4	31.6	6.59
	120	Air	49.7	33.6	8.31
		Water	55.7	38.6	8.98

is prepared by piling up 11 slices and sealing the sides with aluminum sheet. The distribution of relative humidity is estimated by measuring the shrinkage strain on each slice at arbitrary drying times. An inverse analysis is then used to obtain the diffusion coefficient from the measured relative humidity distribution. The numerical approach proposed by us is based on the weighted residual method and on a nonlinear least squares method.

2. OUTLINE OF THE EXPERIMENT

(1) Mix proportions of mortar and concrete

In this research, normal Portland cement type-I (specific gravity: 3.12) was used. Fine aggregate was river sand (specific gravity: 2.62, water absorption: 1.93%). Coarse aggregate was river gravel (maximum size: 8 mm and 16 mm, specific gravity: 2.70, water absorption: 0.62%). Mix proportions of mortar and concrete are shown in **Table 1**. Mechanical properties of mortar and concrete are shown in **Table 2**. These data were obtained at ages of 14 days and 120 days. Two types of specimens were used for strength tests at the age of 120 days. One was cured in water for 120 days, the other was cured in water for 14 days and then in air for 106 days.

Table 3 Physical properties of mortar and concrete

Name of Mixture	Evaporable moisture $w/(S+w)$	Evaporable moisture w/S	Density (g/cm³)	Carbonation (mm)*
M240	8.94%	9.82%	2.12	3
M200	7.35%	7.93%	2.15	3
M160	6.59%	7.05%	1.99	10
G8	6.18%	6.59%	2.36	1
G16	6.20%	6.61%	2.32	1

w: evaporable moisture content (g)
S: solid part of mortar or concrete
*: all relative humidities

Table 4 Moisture loss referred to the moisture content at saturated condition (w'/w)

Name of Mixture	Relative humidity 45%	60%	75%
M240	69.1%	65.7%	51.7%
M200	68.8%	64.5%	53.0%
M160	77.4%	76.2%	68.4%
G8	62.6%	57.5%	51.9%
G16	64.4%	60.9%	51.5%

w': moisture loss measured in each relative humidity room

Table 3 shows the physical properties of the mortars and concretes studied. The evaporable moisture content was obtained from eight slices whose size was 150x100x3 mm. The specimens were dried at 100 C for 14 days after they were cured in water for 7 days and subsequently in a 100% relative humidity chamber for 7 days. All sliced specimens used in this study (150x100x3 mm) were sawed from cubes (150x150x150 mm). Carbonated thickness was measured by using phenolphthalein. These specimens were cured in water for 14 days and in air for 98 days. Three rooms of 45%, 60% and 75% relative humidity were used. In each room, the temperature was 20 C.

Table 4 shows the moisture loss when the mortar or concrete specimens reached equilibrium with the surrounding atmosphere. These data were obtained from the center three slices originally packed in the specimens prepared by piling up 11 slices (150x100x3 mm) and sealing the side with aluminium sheet. The specimens were stored for 98 days in each relative humidity room after they were cured in water for 7 days and subsequently in a 100% relative humidity chamber for 7 days. The values shown in this table are normalized with respect to the moisture content at saturated condition as shown in **Table 3**.

(2) Geometry of specimens

a) Specimens for moisture distribution

Fig.1 shows the sliced specimen for the determination of moisture distribution. The sliced specimen consists of 11 slices of the size of 150x100x3

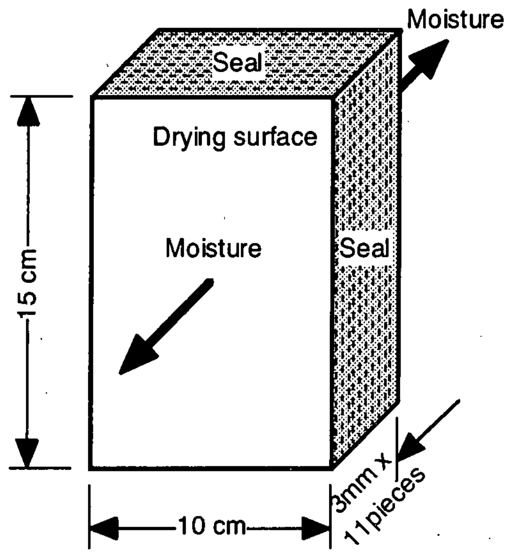

Fig.1 Sliced specimen for measuring moisture loss

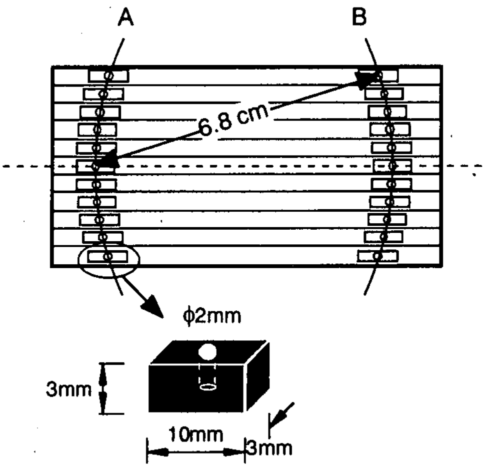

Fig.3 Point gauges on sliced specimens

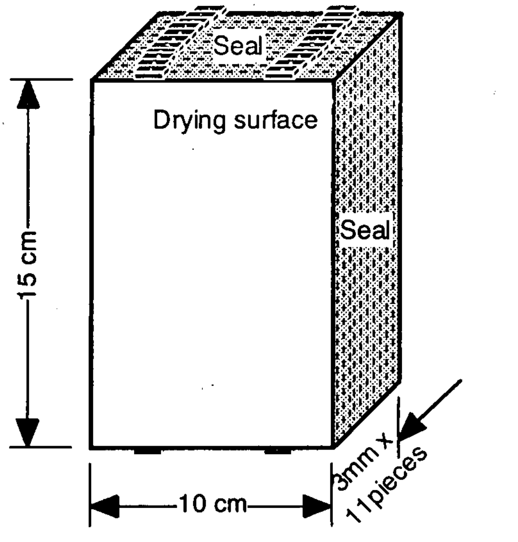

Fig.2 Sliced specimen for measuring shrinkage strains

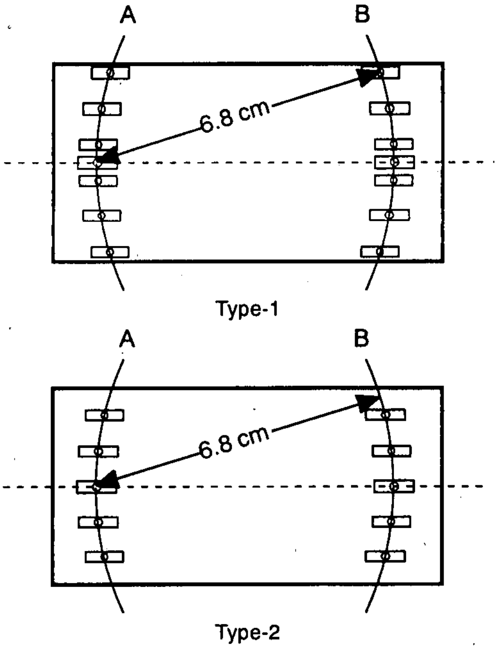

Fig.4 Point gauges on solid specimens

mm. Solid specimens of 150x100x33 mm are also used in series of the experiments. All surfaces, except the two parallel drying surfaces, of the specimens were sealed by aluminum sheet. Two solid specimens and nine sliced specimens were put into each room with a relative humidity of 45%, 60% and 75%, respectively. Altogether, 6 solid specimens and 27 sliced specimens were used for each type of mortar and concrete. The temperature of all rooms was 20 C. The start of drying was 14 days. The weight of each specimen was measured at the time of 0.5, 1, 2, 3, 5, 7, 10, 14, 21, 28, 35, 42, 56, 70 and 98 days after the start of drying. At the time of 0.5, 3, 7, 14, 28, 42, 56, 70 and 98 days after the start of drying, the aluminum sheet was removed from one sliced specimen in order to obtain the moisture loss of each slice. After moisture loss was measured, the specimen was discarded.

b) Specimens for shrinkage strain distribution

Fig.2 shows the sliced specimen for the investigation of the shrinkage strain. The sliced specimen consists of 11 slices (150x100x3 mm). **Fig.3** shows the position of the gauge points on the sliced specimens. The gauge points are put on the top and bottom surfaces in two circular rows. Row B is on the arc with the center gauge of the 6th layer in row A. The gauge points in row A are similar using the 6th slice of row B as center. The size of the solid specimens was 150x100x33 mm. **Fig.4** shows the position of gauge points on the solid specimens. There are two types of solid specimens. The difference is the position

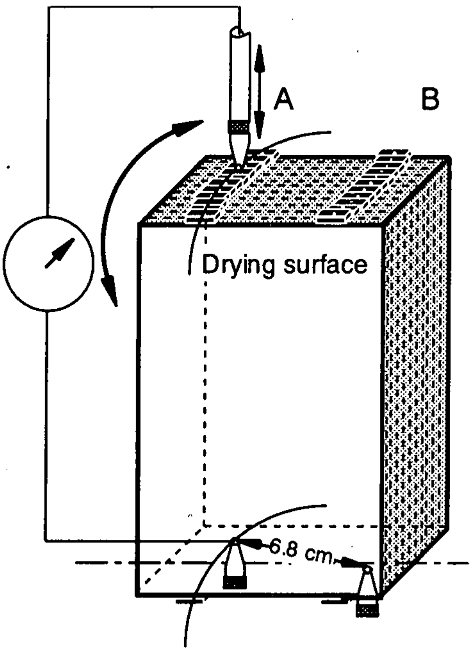

Fig.5 Measuring method of shrinkage strain

of gauge points. The placement of gauge points is the same as in the case of sliced specimens. The gauge points are, however, at every second layer on the solid specimen. The gauge points are made of brass and have a 2 mm hole and a size of 3x3x10 mm. Four surfaces of both types of specimens were sealed by aluminum sheet. The shrinkage strains were measured in the rooms with relative humidities of 45%, 60% and 75%, respectively. Drying started after the specimens were cured in water for 7 days and subsequently in a 100% relative humidity chamber for 7 days. The shrinkage strain of each specimen was measured at 0.5, 1, 2, 3, 5, 7, 10, 14, 21, 28, 35, 42, 56, 70 and 98 days after the start of drying.

(3) Moisture content related to the saturation value

The weights of the specimens were measured using an electronic balance with a minimum division of 1/100 g and a capacity of 5,000 g. With the weight $C(t)$ at drying time t and the weight $C'(t)$ after 14 days in oven, the moisture content $q(t)$ can be obtained from equation (1):

$$q(t)=\frac{w-w'(t)}{S+w-w'(t)}=\frac{C(t)-C'(t)}{C(t)} \quad (1)$$

where, S is the weight of solid part of concrete or mortar, i. e. $C'(t)$. $w'(t)$ is the weight of moisture lost. w is the weight of moisture content at saturated condition. If the moisture content at saturated condition is called p, then, the moisture lost per unit weight of concrete or mortar $H(t)$ can be calculated from equation (2).

$$H(t)=\frac{w'(t)}{S+w}=\frac{p-q(t)}{1-q(t)} \quad (2)$$

$$p=\frac{w}{S+w}=\frac{C(0)-C'(0)}{C(0)} \quad (3)$$

Where, $C(0)$ is the original weight of the specimen when drying time t is zero. $C'(0)$ is the weight of specimen dried in oven for 14 days. The moisture content $\omega(t)$ with respect to moisture content at saturated condition can be obtained from equation (4).

$$\omega(t)=1-\frac{w'(t)}{w}=1-\frac{1}{p}\times H(t) \quad (4)$$

(4) Measuring method for shrinkage strain

In **Fig.5**, the procedure of measurement of the shrinkage strain is displayed. The length change in the longitudinal direction was measured using a linear gauge with a minimum division of 1/1,000 mm. The specimen was supported by three pins that consist of the pointed head of the linear gauge and two supports. Dividing the change of length by the height of the specimen gives the shrinkage strain. When the shrinkage strain in row A is measured, one of two supports is fixed on the gauge point on the bottom of sixth slice in the row B. The second is put on the bottom gauge point of the slice to be measured. The tip of the linear gauge is placed on the top of the slice to be measured.

3. MOISTURE DISTRIBUTION

(1) Effect of gap in sliced specimen on moisture transfer

The diffusion coefficient of moisture transfer in air is approximately 218 mm^2/day at 20 C. It is about 50 or 100 times larger than that in the mortar or concrete[4]. This means that the transfer of moisture in the air is much easier than in mortar or concrete. However, the air layer between slices may act as an obstacle for the moisture transfer in the sliced specimen if the gaps' width is considerable.

Fig.6 shows the weight change of the solid specimen (150x100x33 mm) and the sliced specimen (150x100x3 mm x11slices) for mortar M240 in **Table 1**. The weight change expressed on the vertical axis was obtained by equation (5).

$$Weight\ change=\frac{C(0)-C(t)}{C(0)} \quad (5)$$

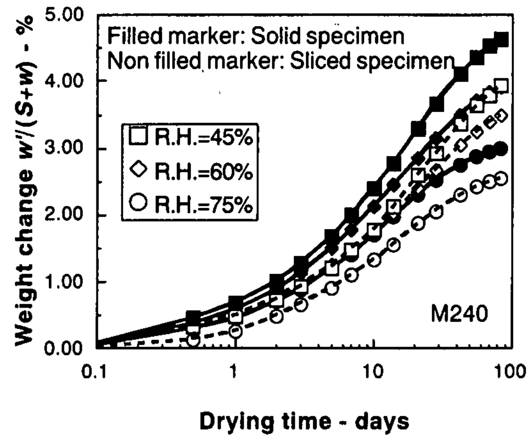

Fig.6 Weight change of solid and sliced specimen

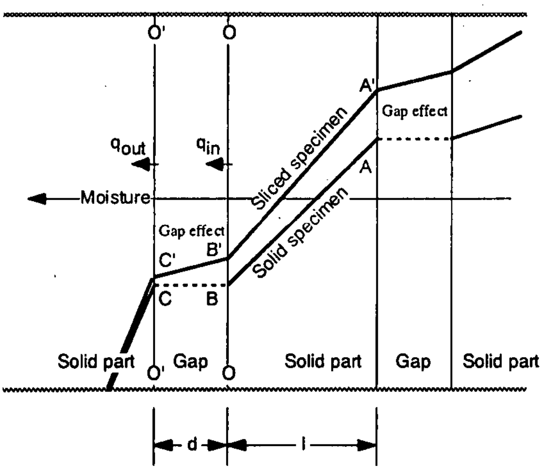

Fig.7 Effect of gap on moisture transfer

where, $C(0)$ is the weight of specimen when drying time is zero. $C(t)$ is the weight of specimen at drying time t. The solid marks represent the data obtained from the solid specimens. The open marks represent the data obtained from sliced specimens. Squares, lozenges and circles indicate different relative humidity rooms of 45%, 60% and 75%, respectively. The results clearly show the solid specimen loses moisture faster than the sliced specimen. This is due to the effect of gaps between each slice.

This can be explained by using **Fig.7**. The curve A'B'C' and curve ABC express the assumed moisture distribution in sliced specimens and in solid specimens, respectively. When q_{in} and q_{out} are the moisture flow per unit time from the surface O-O into air gap and that from air gap into the surface O'-O', respectively, q_{out} minus q_{in} is equal to the moisture change in the gap surrounded by OO and O'O'.

$$q_{out} - q_{in} = \frac{1}{2}\frac{\partial(\omega_{B'} + \omega_{C'})}{\partial t} \times d \quad (6)$$

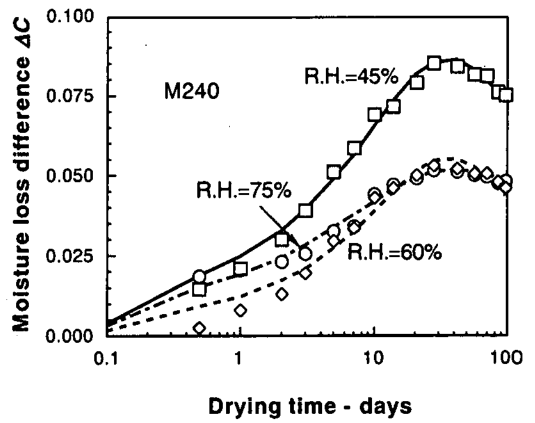

Fig.8 Moisture loss difference between sliced and solid specimen

where, $\omega_{B'}$ and $\omega_{C'}$ are moisture contents at the position B' and C', respectively; d is the thickness of the gap between each sliced specimen. If the thickness of gap is zero, q_{out} must be equal to q_{in}, that is, the moisture contents on the surface O-O and the surface O'-O' are the same such as ω_B and ω_C. Whereas, when d can not be ignored, the slope of moisture increases in the gap. With the slope of moisture in the gap and the diffusion coefficient D_{air} of air, the moisture flow q_{out} can be expressed by equation (7).

$$q_{out} = D_{air}\frac{\omega_{B'} - \omega_{C'}}{d} = q_{in} + \frac{1}{2}\frac{\partial(\omega_{B'} + \omega_{C'})}{\partial t} \times d \quad (7)$$

As q_{out} is bigger than q_{in}, $\omega_{B'}$ must be bigger than $\omega_{C'}$. The bigger the thickness of gap d, the bigger the difference between $\omega_{B'}$ and $\omega_{C'}$. Hence, the sliced specimen loses its moisture slower than the solid specimen (Q. E. D.).

Fig.8 shows the difference in moisture loss between the sliced and solid specimens. Squares, lozenges and circles represent results at 45%, 60% and 75% relative humidity, respectively. The difference of moisture loss is related to the saturated moisture content; equation (8).

$$\text{Moisture loss difference } \Delta C = \frac{1}{p} \times \left(\Delta C_{solid}(t) - \Delta C_{sliced}(t)\right) \quad (8)$$

$$\Delta C_{sliced}(t) = \frac{C_{sliced}(0) - C_{sliced}(t)}{C_{sliced}(0)} \quad (9)$$

$$\Delta C_{solid}(t) = \frac{C_{solid}(0) - C_{solid}(t)}{C_{solid}(0)} \quad (10)$$

where, p is the moisture content normalized with respect to the weight of specimen at saturated condition expressed by equation (3). $C_{sliced}(t)$ and $C_{solid}(t)$ are the weight of sliced and solid specimens at arbitrary drying time t, respectively. $C_{sliced}(0)$ and $C_{solid}(0)$ are the weight of sliced and solid specimens when drying time is zero. The moisture loss difference shown in **Fig.8** is due to the moisture transport disturbed by air gaps. The moisture loss difference has a peak and finally disappears as shown in **Fig.8**.

In this study, the moisture distribution obtained from a sliced specimen is used as a substitute for that of the solid specimen in order to obtain the diffusion coefficients. If the effect of a gap is ignored or the thickness of gaps is regarded as zero, then the moisture distribution in **Fig.7** decreases from A'B'C' to ABC. Before this substitution is applied, it has to be confirmed that the effect of gap on the moisture flow in sliced specimen is small or negligible.

(2) Moisture loss distribution

If the moisture loss of sliced specimen $g(x)$ at the position x is represented by a power expression, then equation (11) can be derived.

$$g(x) = a(x_{center} - x)^n + c \tag{11}$$

$$x_{center} = 16.5\,mm \tag{12}$$

where, $x(mm)$ is the distance from drying surface. a, n and c are constants determined from experimental data. Equations (11) and (12) describe the zero gradient of moisture loss at center of specimen due to the symmetry boundary condition. When $g(x)$ is the moisture loss per unit cross section, the total moisture loss of sliced specimen Q_{sliced} can be expressed by equation (13).

$$\begin{aligned}\frac{1}{2A} \times Q_{sliced} &= \int_{x_{surface}}^{x_{center}} g(x)dx \\ &= \frac{a}{n+1} x_{center}{}^{n+1} + x_{center} \times c = \sum_{i=1}^{6} l_i \times g_i\end{aligned} \tag{13}$$

where, A(=100x150 mm^2) is the drying surface. l_i is the thickness of the slice. The surface slice has i=1. The center slice has i=6. That is, l_i=3 mm when i=1 to 5. l_6=1.5 mm due to the symmetry boundary condition. g_i is the moisture loss of each slice. The moisture loss of surface slice and center slice can be represented approximately by equation (14) and equation (15), respectively.

$$g(x_{surface} = 0) = a \times x_{center}{}^n + c = \frac{g_1 - g_2}{3} \times 1.5 + g_1 \tag{14}$$

$$g(x_{center}) = c = g_6 \tag{15}$$

The constant c involved in $g(x)$ can be obtained by equation (15). From equation (13) and equation (14), equation (16) is derived for constant n. The constant a can be determined by equation (17).

$$n = \frac{\left(\frac{g_1 - g_2}{3} \times 1.5 + g_1 - g_6\right) \times x_{center}}{\sum_{i=1}^{6} l_i \times g_i - x_{center} \times g_6} - 1 \tag{16}$$

$$a = \frac{g(x_{surface}) - g_6}{x_{center}{}^n} \tag{17}$$

When $f(x)$ is the moisture loss within a solid specimen, the total moisture loss of solid specimens Q_{solid} can be expressed by equation (18).

$$\frac{1}{2A} \times Q_{solid} = \int_{x_{surface}}^{x_{center}} f(x)dx = \sum_{i=1}^{6} l_i \times g_i + \frac{L}{2} \times \Delta C \tag{18}$$

where, ΔC is the moisture loss difference between a solid specimen and a sliced specimen as shown in **Fig.8**. L is the thickness of the specimen, i. e. 33 mm. The moisture loss at the drying surface of a solid specimen $f(x_{surface})$ can be determined by using the moisture loss of a surface layer of a sliced specimen $g(x_{surface})$ and the moisture loss difference on the drying surfaces between solid and sliced specimen Δh_s. Δh_s is calculated from the film coefficient H_F and $d\Delta C/dt$ as follows.

$$\begin{aligned}\Delta h_s &= \frac{1}{H_F} \times \frac{1}{2A} \times \left(\frac{dQ_{solid}}{dt} - \frac{dQ_{sliced}}{dt}\right) \\ &= \frac{1}{H_F} \times \frac{L}{2} \times \frac{d\Delta C}{dt}\end{aligned} \tag{19}$$

$d\Delta C/dt$ is the rate of moisture loss difference between a solid specimen and a sliced specimen. Δh_s is zero when $d\Delta C/dt$ is equal to zero. Δh_s remains zero and can never become negative because the rate of moisture loss of a sliced specimen is never greater than that of a solid specimen. Equation (19) is derived from equation (20) and equation (21).

$$\frac{1}{2A} \times \frac{dQ_{sliced}}{dt} = H_F\left(g(x_{surface}, t_\infty) - g(x_{surface}, t)\right) \tag{20}$$

$$\frac{1}{2A} \times \frac{dQ_{solid}}{dt} = H_F\left(f(x_{surface}, t_\infty) - f(x_{surface}, t)\right) \tag{21}$$

where, $g(x_{surface}, t_{\infty})$ and $f(x_{surface}, t_{\infty})$ are the moisture loss of surface layer of sliced and solid specimens at ultimate drying time, respectively. These two values must be the same. The film coefficient H_F expresses the relationship between q_t and $\omega_{surface}$-ω_a. q_t is the moisture that passes through the unit area of drying surface per unit time. $\omega_{surface}$ is the moisture loss through the drying surface at arbitrary drying time. ω_a is the moisture loss when the relative humidity of the cement-based material is equal to that of the surrounding atmosphere.

$$q_t = H_F \times \left(\omega_a - \omega_{surface}\right) = \frac{1}{2A} \times \frac{dQ_{sliced}}{dt} \qquad (22)$$

Fig.9 shows the relationship between q_t and $\omega_{surface}$-ω_a of the sliced specimens. It is independent of the ambient relative humidity. The film coefficient of this material is 2.43 mm/day.

When $f(x)$ is expressed by equation (23), the moisture loss through the drying surface of the solid specimen can be expressed by equation (24).

$$f(x) = \alpha\left(x_{center} - x\right)^{\beta} + \gamma \qquad (23)$$

$$f(x_{surface}) = \alpha \times x_{center}{}^{\beta} + \gamma = g(x_{surface}) + \Delta h_s \qquad (24)$$

Moisture content and the gradient of moisture distribution of solid specimens are smaller than those of sliced specimens because the moisture transfer in sliced specimens is disturbed by air gaps. If the gradients of moisture distributions at the drying surfaces of sliced and solid specimens are considered to be identical, the following equation can be derived. Under this hyphothesis, the difference of moisture content at the center of each specimen is maximum.

$$\frac{df(x_{surface})}{dx} = \alpha \times \beta \times x_{center}{}^{\beta-1} = \frac{dg(x_{surface})}{dx} \qquad (25)$$

By using equations (18), (24) and (25), constants α, β and γ can be obtained as follows.

$$\beta = \frac{x_{center}{}^{2} \times \frac{dg(x_{surface})}{dx}}{\left(g(x_{surface}) + \Delta h_s\right) \times x_{center} - \sum_{i=1}^{6} l_i \times g_i - \frac{L}{2} \times \Delta C} - 1 \qquad (26)$$

$$\alpha = \frac{\frac{dg(x_{surface})}{dx}}{\beta \times x_{center}{}^{\beta-1}} \qquad (27)$$

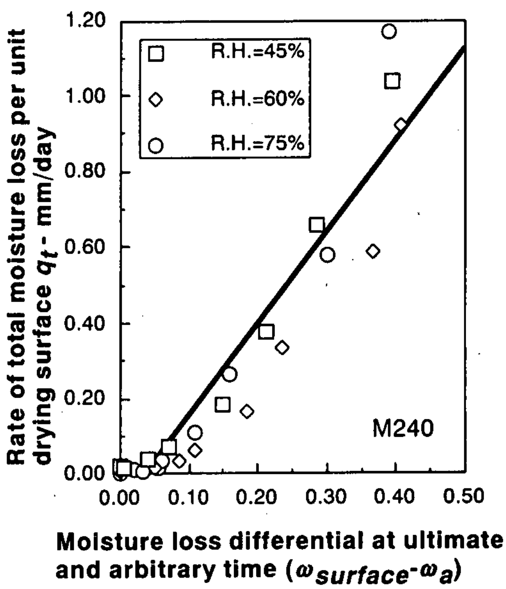

Fig.9 Relationship between q_t and ω_a-$\omega_{surface}$

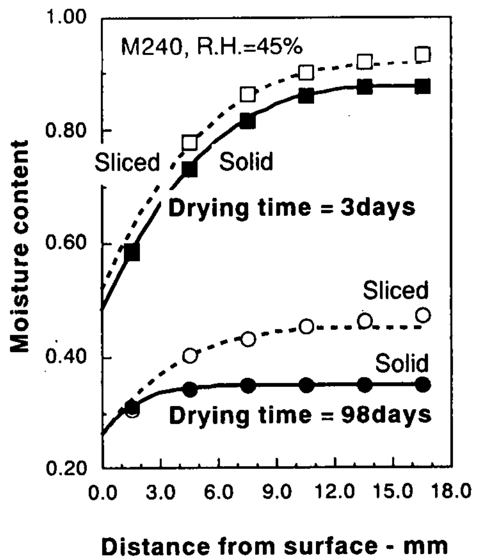

Fig.10 Moisture distribution of solid and sliced specimen

$$\gamma = g(x_{surface}) + \Delta h_s - \alpha \times x_{center}{}^{\beta} \qquad (28)$$

Fig.10 shows the moisture distribution of sliced and solid specimens. The moisture loss shown in this figure is normalized with respect to the moisture content at saturated condition. The drying times are 3 days and 98 days. Open circles and squares are the experimental results obtained from sliced specimens. The solid circles and squares are the estimated moisture distributions of solid specimens by equation (23). It is evident that the moisture distribution in sliced specimens can be represented by a power expression.

From the comparison of moisture distributions of sliced specimen and solid specimen, it is confirmed that the effect of the gap between slices seems to be small, nevertheless the moisture distribution in solid specimens is estimated on the hypothesis which promotes the difference of moisture distribution between sliced and solid specimen.

(3) Diffusion coefficient as a function of moisture content

Equation (29) is the diffusion equation in one dimension.

$$\frac{\partial \omega(x,t)}{\partial t} = \frac{\partial}{\partial x}\left(D(\omega) \times \frac{\partial \omega(x,t)}{\partial x}\right) \tag{29}$$

where, $\omega(x,t)$ is the moisture content. $D(\omega)$ is the diffusion coefficient which is a function of moisture content $\omega(x,t)$. x and t are distance in the direction of specimen's thickness and drying time, respectively. Equation (29) must be satisfied everywhere in the specimen. Equation (30) holds with respect to an arbitrary function $F(x,t)$.

$$\int_{x_{surface}}^{x_{center}} F(x,t)\left\{\frac{\partial}{\partial x}\left(D(\omega) \times \frac{\partial \omega(x,t)}{\partial x}\right) - \frac{\partial \omega(x,t)}{\partial t}\right\}dx = 0 \tag{30}$$

By means of the formula of partial integration, the first term of equation (30) is rewritten as follows.

$$\begin{aligned}&\int_{x_{surface}}^{x_{center}} F \times \frac{\partial}{\partial x}\left(D \times \frac{\partial \omega}{\partial x}\right)dx \\ &= \left[F \times D \times \frac{\partial \omega}{\partial x}\right]_{x_{surface}}^{x_{center}} - \int_{x_{surface}}^{x_{center}} \frac{\partial F}{\partial x} \times D \times \frac{\partial \omega}{\partial x}dx\end{aligned} \tag{31}$$

Substituting equation (31) into equation (30), equation (32) is derived.

$$\begin{aligned}&\int_{x_{surface}}^{x_{center}} D \times \frac{\partial F}{\partial x} \times \frac{\partial \omega}{\partial x}dx \\ &= \left[F \times D \times \frac{\partial \omega}{\partial x}\right]_{x_{surface}}^{x_{center}} - \int_{x_{surface}}^{x_{center}} F \times \frac{\partial \omega}{\partial t}dx\end{aligned} \tag{32}$$

From the boundary conditions, equations (33) and (34) are derived.

$$q_t = -D \times \frac{\partial \omega(x_{surface},t)}{\partial x} \tag{33}$$

$$\frac{\partial \omega(x_{center},t)}{\partial x} = 0 \tag{34}$$

q_t is the moisture content which passes through a unit of the drying surface area per unit of time. Taking the boundary conditions expressed by equation (33) and equation (34) into consideration, equation (32) can be rewritten as equation (35).

$$\begin{aligned}&\int_{x_{surface}}^{x_{center}} D \times \frac{\partial F}{\partial x} \times \frac{\partial \omega}{\partial x}dx \\ &= F(x_{surface},t) \times q_t - \int_{x_{surface}}^{x_{center}} F \times \frac{\partial \omega}{\partial t}dx\end{aligned} \tag{35}$$

Now, $\omega(x,t)$ is chosen for the arbitrary function $F(x,t)$. Equation (35) is rewritten in equation (36).

$$\begin{aligned}&\int_{x_{surface}}^{x_{center}} D \times \left(\frac{\partial \omega}{\partial x}\right)^2 dx \\ &= \omega(x_{surface},t) \times q_t - \int_{x_{surface}}^{x_{center}} \omega \times \frac{\partial \omega}{\partial t}dx\end{aligned} \tag{36}$$

Furthermore, when $\omega(x,t)$ is expressed by the moisture content of each layer $\omega_1(t)$, $\omega_2(t)$, ..., $\omega_i(t)$, ... and $\omega_6(t)$, the moisture passing through the unit drying surface area per unit time q_t is expressed in equation (37).

$$q_t = \sum_{i=1}^{6} \frac{d\omega_i(t)}{dt} \times l_i \tag{37}$$

Equation (36) can be rewritten as equation (38).

$$\begin{aligned}&\sum_{i=1}^{6} D(\omega_i) \times \left(\frac{d\omega_i}{dx}\right)^2 \times l_i \\ &= \omega(x_{surface},t)\sum_{i=1}^{6} \frac{d\omega_i}{dt} \times l_i - \sum_{i=1}^{6} \omega_i \times \frac{d\omega_i}{dt} \times l_i\end{aligned} \tag{38}$$

where, $d\omega_i/dx$ is the gradient of moisture distribution at the center $x=x_i$ of each layer. The position x_i which represents ω_i is as follows: x_1=1.5 mm, x_2=4.5 mm, x_3=7.5 mm, x_4=10.5 mm, x_5=13.5 mm and x_6=16.5 mm. If the diffusion coefficient is expressed by an exponential equation, that is,

$$D(\omega) = a \times e^{b(1-\omega)} \tag{39}$$

then, equation (38) can be rewritten as equation (40).

$$\begin{aligned}&a\sum_{i=1}^{6}\left(e^{b(1-\omega_i)} \times \left(\frac{d\omega_i}{dx}\right)^2 \times l_i\right) \\ &= \omega(x_{surface},t) \times \sum_{i=1}^{6} \frac{d\omega_i}{dt} \times l_i - \sum_{i=1}^{6} \omega_i \times \frac{d\omega_i}{dt} \times l_i\end{aligned} \tag{40}$$

Furthermore, as equation (40) must be satisfied at any drying time, equation (41) is derived.

$$a\sum_{t=0}^{t=98}\sum_{i=1}^{6} e^{b(1-\omega_i)}\times\left(\frac{d\omega_i}{dx}\right)^2\times l_i = \sum_{t=0}^{t=98}\left(\omega(x_{surface},t)\sum_{i=1}^{6}\frac{d\omega_i}{dt}\times l_i - \sum_{i=1}^{6}\omega_i\times\frac{d\omega_i}{dt}\times l_i\right) \quad (41)$$

Now, $\omega(x_{surface},t)$, $d\omega_i/dx$ and $d\omega_i/dt$ are known because the moisture distribution has been determined by the experimental data. Therefore, every factor in equation (41) except constants a and b are known. By using one of the least square methods, constant a and b can be determined. The modified Marquart method has been used as the least square method in this study. The diffusion coefficient D_{sliced} and D_{solid} of concrete G16 in **Table 1** are obtained by using the experimental data of sliced specimens and by using the assumed moisture distributions of solid specimens calculated by equation (23), respectively.

$$D_{sliced} = 6.47e^{-3.23(1-\omega)} \quad (42)$$

$$D_{solid} = 9.15e^{-3.35(1-\omega)} \quad (43)$$

The smaller the diffusion coefficient, the slower the moisture loss. It is evident from a comparison of equation (42) and equation (43) that the diffusion coefficient of sliced specimens is smaller than that of solid specimens. It is obvious that the proposed numerical method has exactly translated the fact that the moisture transfer in sliced specimen is slower than that in solid specimen.

(4) Effect of air gaps on moisture transfer

Fig.11 shows the average moisture loss of 3 cylindrical specimens with different diameters for mix G16. Analogous specimens were made using the other mixes and were obtained similar results. The circles, lozenges and squares are the experimental results obtained from the cylinders with diameter 50, 80 and 150 mm, respectively. The height of the cylinders of diameter 50, 80 and 150 mm is 100, 100 and 150 mm, respectively. These results were obtained in the 45% relative humidity room. The top and the bottom surfaces were sealed with resin in order to prevent moisture transfer. **Fig.12** shows the moisture loss of the cylindrical and rectangular specimens measured in the 45% and 75% relative humidity rooms. Circles are the experimental data of the cylinder. Squares are the experimental data of the prism. The data expressed by open marks and solid marks were measured in the 45% and 75% relative humidity rooms, respectively. The dimensions of cylindrical and rectangular

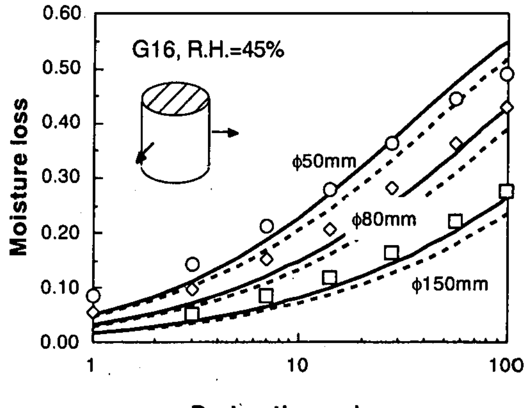

Fig.11 Moisture loss of cylinders with different diameter

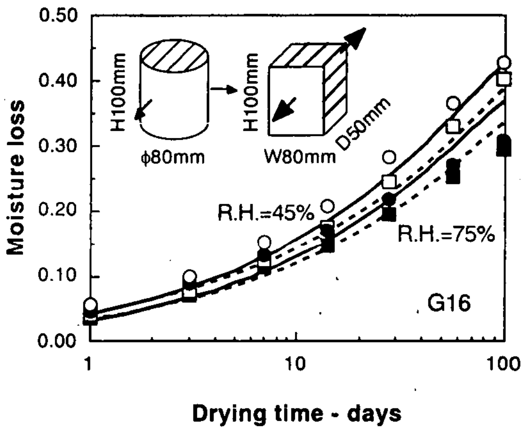

Fig.12 Moisture loss of (○) cylindrical and (□) rectangular specimens in 45% and 75% relative humidity

specimens are ϕ80x100 mm and 50x80x100 mm, respectively. The top and the bottom of the cylinder were sealed by resin. The four sides of the rectangular specimen were sealed by aluminum sheet. The drying surface was 80x100 mm. The curves were calculated by the finite element method. The broken line was calculated with the diffusion coefficient D_{sliced} as expressed by equation (42). The solid line was calculated with the diffusion coefficient D_{solid} as expressed by equation (43). It is clear from each figure that the difference between the calculated values with D_{sliced} and D_{solid} is very small and that the calculated curves fit the experimental data well. The effect of gaps between each slice on the moisture transfer can be considered as negligible.

4. RELATIVE HUMIDITY DISTRIBUTION

(1) Relative humidity in mortar and concrete

Fig.13 shows the measured shrinkage strains of each layer of a sliced specimen. Squares, lozenges, circles,

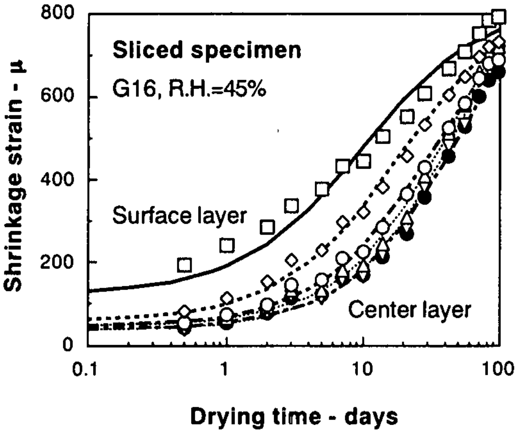

Fig.13 Shrinkage strain measured at each layer of a sliced specimen

triangles, reverse triangles and solid circles are the data obtained from the drying surface layer, the second layer and so forth to the center layer. The curves drawn in this figure are obtained by regression analysis using equation (44).

$$Shrinkage\ strain = c_0 + \frac{c_2 \times t}{c_1 + t} \tag{44}$$

where, t is drying time. c_0, c_1 and c_2 are unknown factors which are determined by the least square method. As the shrinkage strain of very thin specimens develops approximately linearly with the change of relative humidity of a specimen, the relative humidity of the sliced specimen can be expressed by using c_1 or c_0 and c_2 as shown in equation (45).

$$Relative\ humidity\ of\ specimen = 1 - \frac{(1-R.H.)\times t}{c_1 + t}$$
$$= 1-(1-R.H.)\times \frac{Shrinkage\ strain - c_0}{c_2} \tag{45}$$

where $R.H.$ is the relative humidity in the surrounding atmosphere. Equation (45) expresses that the relative humidity of each thin sliced specimen is 100% when drying time is equal to zero and that relative humidity in specimen is equivalent to that of atmosphere when the shrinkage strain is reached ultimate value, that is, c_0 plus c_2. The change of relative humidity of specimen is expressed by using the coefficient which expresses the development of shrinkage strain.

(2) Diffusion coefficient as a function of relative humidity

Substituting the term of moisture content ω_i in equation (41) for the term of relative humidity h_i of cement-based materials, equation (41) may be rewritten as equation (46).

$$a\sum_{t=0}^{t=98}\sum_{i=1}^{6} e^{b(1-h_i)} \times \left(\frac{dh_i}{dx}\right)^2 \times l_i$$
$$= \sum_{t=0}^{t=98}\left(h(x_{surface},t)\sum_{i=1}^{6}\frac{dh_i}{dt}\times l_i - \sum_{i=1}^{6} h_i \times \frac{dh_i}{dt}\times l_i\right) \tag{46}$$

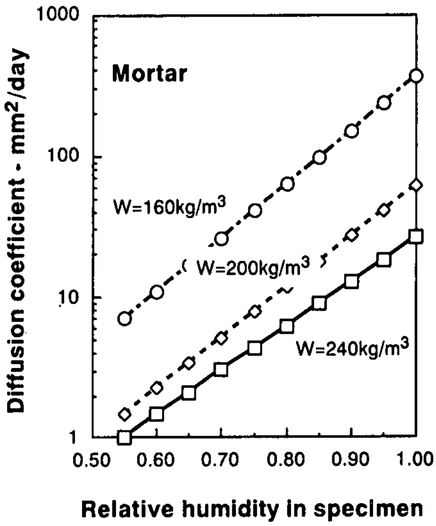

Fig.14 Diffusion coefficient of mortars M240, M200 and M160

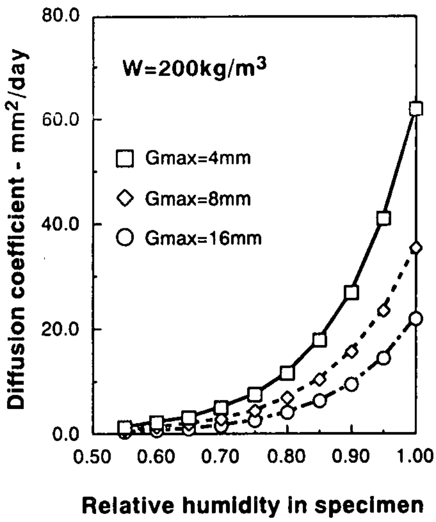

Fig.15 Diffusion coefficient of M200, G8 and G16

Each diffusion coefficient of M240, M200, M160, G8 and G16 has been determined by using equation (46). To obtain these diffusion coefficients, all the measured data from the specimen for 45%, 60% and 75% relative humidity were used. Results are given in equations (47) to (51).

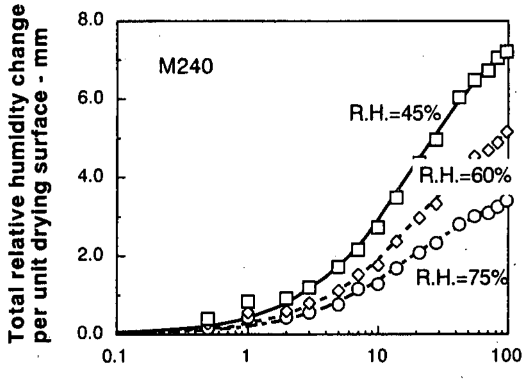

Fig.16 Total relative humidity change of M240

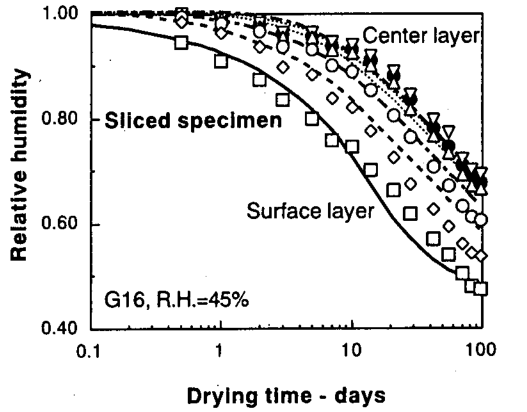

Fig.18 Comparison of calculated value and experimental data for the relative humidity as function of dring time

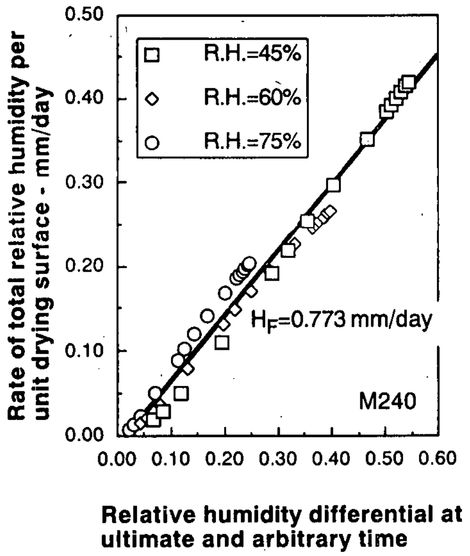

Fig.17 Film coefficient of M240

M240: $$D(h) = 26.3e^{-7.21(1-h)} \tag{47}$$

M200: $$D(h) = 62.0e^{-8.29(1-h)} \tag{48}$$

M160: $$D(h) = 363e^{-8.77(1-h)} \tag{49}$$

G8: $$D(h) = 35.4e^{-8.15(1-h)} \tag{50}$$

G16: $$D(h) = 22.0e^{-8.26(1-h)} \tag{51}$$

Fig.14 and **Fig.15** show the relationship between the diffusion coefficient and the relative humidity of mortar and concrete as expressed by equations (47) to (51). It is confirmed that the bigger the maximum size of aggregate, the smaller the diffusion coefficient. The results obtained by the proposed method are in agreement with usual conception of moisture transfer in concrete due to drying.

(3) Film coefficient

Fig.16 shows half the total relative humidity $Q/2$ of mortar M240 with respect to the drying time. The total relative humidity Q can be calculated by equation (52).

$$Q = \sum_{i=1}^{11} (1 - h_i) \times l_i \tag{52}$$

The film coefficient H_F expresses the relationship between q_t and $(h_{surface} - h_a)$. q_t is the time differentiation of $Q/2$. $h_{surface}$ is the relative humidity on the drying surface. h_a is the relative humidity of the surrounding atmosphere. The bigger the film coefficient, the more important becomes the moisture transfer of the drying surface.

$$H_F = \frac{q_t}{h_{surface} - h_a} \tag{53}$$

$$q_t = \frac{1}{2} \times \frac{dQ}{dt} \tag{54}$$

Fig.17 shows the relationship between q_t and $(h_{surface} - h_a)$ of M240. As is evident from this figure, the effect of relative humidity of the surrounding atmosphere on the film coefficient is very small. The film coefficient H_F has been regressed by equation (53). For mixes M240, M200, M160, G8 and G16, they are 0.773, 1.082, 2.502, 0.726 and 0.541 mm/day, respectively. It is confirmed that the bigger the maximum aggregate size, the smaller the film coefficient. Valid results for film coefficients have been obtained, too.

(4) Precision of the proposed method

Fig.18 shows the comparison of experimental data of relative humidity in each slice with the calculated data using the diffusion coefficient and the film coefficient described in the previous section. The curves in this figure are the calculated values. The marks are the experimental data obtained from concrete G16 in the 45% relative humidity room. It is evident from this figure that the calculated values fit the experimental data very well.

5. DEFORMATION DUE TO DRYING

(1) Shrinkage coefficient

The shrinkage coefficient expresses the strain change on function of the moisture or relative humidity change. If the thickness of specimen is so thin that the moisture distribution can be regarded to be constant, the shrinkage coefficient may be obtained directly. However, the deformation of a very thin specimen is strongly affected by carbonation as shown in **Fig.19**. The shrinkage strain and weight change in this figure were measured on 100x150x3 mm prisms for 28 days. The horizontal axis is the weight change of specimens divided by the weight before drying started. This value is positive when moisture is lost. Within one week after drying, the weight of each specimen in all three relative humidities increased even though it dried. This is the effect of carbonation. Hence it is impossible to determine the shrinkage coefficient of a very thin specimen except in the special condition of a carbon dioxide free environment. A method of obtaining the shrinkage coefficient without the effect of carbon dioxide needs to be established[5].

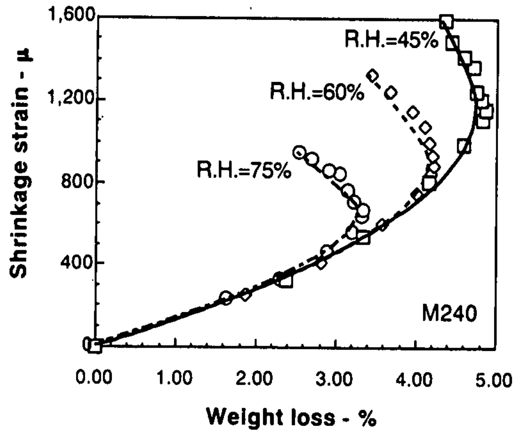

Fig.19 Shrinkage strain of very thin specimen

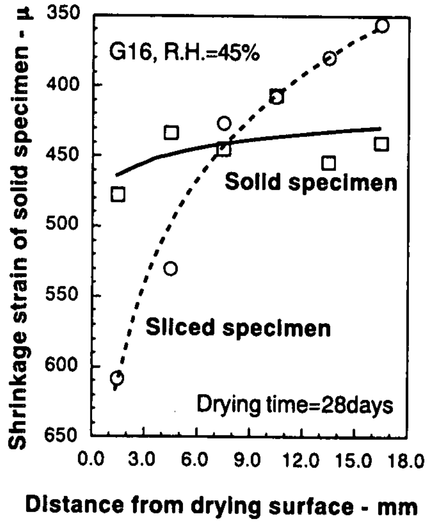

Fig.20 Shrinkage strain on sliced and solid specimen

Fig.20 shows the shrinkage strain measured at the solid and sliced specimen. The deformation of sliced specimens due to drying increases almost proportionally to relative humidity change because each layer is unrestrained. Whereas the deformation of solid specimens is nearly independent of position due to eigenstresses which modify the deformation to ensure compatibility. It is clear that Bernoulli's hypothesis, plane sections remain plane, holds for the deformation due to drying of this size of solid specimens. The stress condition of solid specimens can be expressed as follows.

$$\sigma_y = \sigma_z = f(x) \tag{55}$$

$$\sigma_x = \tau_{xy} = \tau_{yz} = \tau_{zx} = 0 \tag{56}$$

x indicates the position in direction of the thickness. As shrinkage distribution $Sh(x)$ within a solid specimen changes along the x-axis, each strain is expressed as follows.

$$\varepsilon_y = \varepsilon_z = \frac{f(x)}{E}(1-\nu) + Sh(x) \tag{57}$$

$$\varepsilon_x = -\frac{2\nu f(x)}{E} + Sh(x) \tag{58}$$

$$\gamma_{xy} = \gamma_{yz} = \gamma_{zx} = 0 \tag{59}$$

where, ν is the Poisson ratio and E is Young's modulus. Equation (60) is obtained by substituting equations (57), (58) and (59) into the condition of compatibility expressed by equations (61), (62) and (63).

$$\frac{d^2}{dx^2}\left\{f + \frac{E}{1-\nu}Sh(x)\right\} = 0 \tag{60}$$

$$\frac{\partial^2 \varepsilon_x}{\partial y^2}+\frac{\partial^2 \varepsilon_y}{\partial x^2}=\frac{\partial^2 \gamma_{xy}}{\partial x \partial y} \quad (61)$$

$$\frac{\partial^2 \varepsilon_y}{\partial z^2}+\frac{\partial^2 \varepsilon_z}{\partial y^2}=\frac{\partial^2 \gamma_{yz}}{\partial y \partial z} \quad (62)$$

$$\frac{\partial^2 \varepsilon_z}{\partial x^2}+\frac{\partial^2 \varepsilon_x}{\partial z^2}=\frac{\partial^2 \gamma_{zx}}{\partial z \partial x} \quad (63)$$

Equation (64) which expresses the stresses σ_y and σ_z can be obtained by solving the differential equation (60).

$$\sigma_y=\sigma_z=f(x)=-\frac{E}{1-\nu}Sh(x)+C_1+C_2x \quad (64)$$

where C_1 and C_2 are constants. They are obtained under the condition that the resultant stress and moment on the edge of a specimen are equal to zero, that is,

$$\int_{-L'}^{L'}\sigma_y dx=\int_{-L'}^{L'}\sigma_z x dx=0 \quad (65)$$

where L'=16.5 mm is half the thickness of a specimen. By using equation (65), constant C_1 and C_2 can be determined and equation (66) is obtained.

$$\sigma_y=\sigma_z$$
$$=\frac{E}{1-\nu}\left\{-Sh(x)+\frac{1}{2L'}\int_{-L'}^{L'}Sh(x)dx+\frac{3x}{2L'^3}\int_{-L'}^{L'}Sh(x)xdx\right\} \quad (66)$$

$$N_T=\int_{-L'}^{L'}E\times Sh(x)dx \quad (67)$$

$$M_T=\int_{-L'}^{L'}E\times Sh(x)\times xdx \quad (68)$$

When equations (67) and (68) are used, equation (66) can be rewritten as equation (69). The stress distribution can then be expressed by equations (69) and (70).

$$\sigma_y=\sigma_z=\frac{1}{1-\nu}\left\{-E\times Sh(x)+\frac{1}{2L'}N_T+\frac{3x}{2L'^3}M_T\right\} \quad (69)$$

$$\sigma_x=\tau_{xy}=\tau_{yz}=\tau_{zx}=0 \quad (70)$$

The strain distribution can be expressed from equations (71), (72) and (73).

$$\varepsilon_y=\varepsilon_z=\frac{1}{E}\left\{\frac{1}{2L'}N_T+\frac{3x}{2L'^3}M_T\right\} \quad (71)$$

$$\varepsilon_x=-\frac{2\nu}{(1-\nu)E}\left\{\frac{1}{2L'}N_T+\frac{3x}{2L'^3}M_T\right\}+\left(\frac{1+\nu}{1-\nu}\right)\times Sh(x) \quad (72)$$

$$\gamma_{xy}=\gamma_{yz}=\gamma_{zx}=0 \quad (73)$$

If the Young's modulus at each position of the x-axis is the same, then N_T represented by equation (67) and M_T represented by equation (68) can be rewritten as follows.

$$N_T=l\times E\times(2Sh_1+2Sh_2+2Sh_3+2Sh_4+2Sh_5+Sh_6) \quad (74)$$

$$M_T=0 \quad (75)$$

where Sh_i is the shrinkage strain which corresponds to i-th layer with the relative humidity change. Equations (74) and (75) take advantage of symmetrical boundary condition. Equation (71) can be rewritten by equations (74) and (75) as follows.

$$\varepsilon_i(t)=\frac{l}{L}\times\sum_{i=1}^{11}Sh_i(t) \quad (76)$$

where $\varepsilon_i(t)$ is the shrinkage strain measured at the position i on solid specimens. As l=3 mm and L=33 mm, l/L=1/11. Equation (76) means that the deformation of this size of solid specimen due to drying is independent of the position to be measured.

When the relationship between shrinkage strain and relative humidity of specimen is expressed by the power expression to take the effect of carbonation and creep due to eigenstresses into consideration, equation (78) is derived.

$$Sh_i(t)=a\times(1-h_i(t))^b \quad (77)$$

$$\varepsilon_{avg.}(t)=\frac{1}{11}\times a\sum_{i=1}^{11}(1-h_i(t))^b \quad (78)$$

where, $\varepsilon_{avg.}(t)$ is the average deformation measured on solid specimen.

$$\varepsilon_{avg.}(t)=\frac{1}{11}\sum_{i=1}^{11}\varepsilon_i(t) \quad (79)$$

As equation (78) must be satisfied at any drying time, equation (80) can be derived:

$$\sum_{t=0}^{t=98}\varepsilon_{avg.}(t)=\frac{1}{11}\times a\sum_{t=0}^{t=98}\sum_{i=1}^{11}(1-h_i(t))^b \quad (80)$$

where, a and b are unknown factors determined by

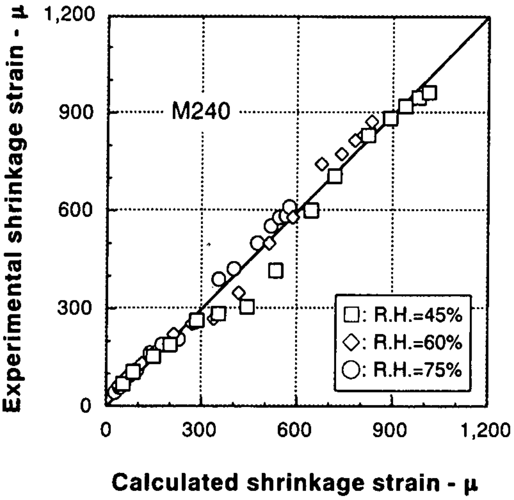

Fig.21 Precision of analysis results of M240

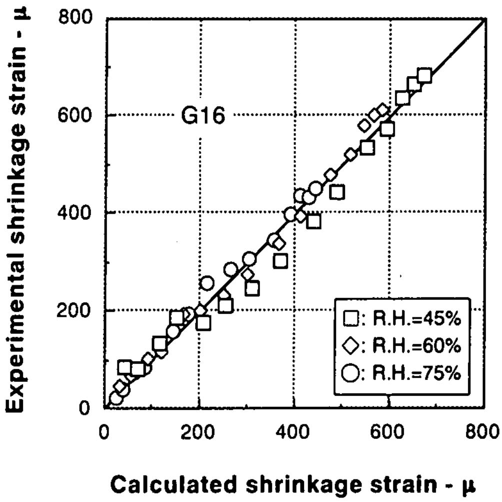

Fig.22 Precision of analysis results of G16

the least square method. The relationship between shrinkage strain and relative humidity of M240, M200, M160, G8 and G16 obtained from equation (80) can be summarized by equations (81), (82), (83), (84) and (85), respectively. All data measured in the 45%, 60% and 75% relative humidity rooms were used to obtain these equations.

M240: $$Sh(h) = 2{,}170(1-h(t))^{0.918} \quad (81)$$

M200: $$Sh(h) = 1{,}840(1-h(t))^{0.933} \quad (82)$$

M160: $$Sh(h) = 1{,}530(1-h(t))^{0.744} \quad (83)$$

G8: $$Sh(h) = 1{,}320(1-h(t))^{0.770} \quad (84)$$

G16: $$Sh(h) = 1{,}370(1-h(t))^{0.749} \quad (85)$$

It is confirmed that the higher the water content of mortar, the bigger becomes the shrinkage strain and that the bigger the maximum size of aggregate, the smaller will be the shrinkage strain. The result shows that the experimental approach and numerical method proposed in this study give valid values.

(2) Precision of the proposed method

Fig.21 and **Fig.22** show the comparison of experimental shrinkage strain of solid specimens with the calculated value. The diffusion, film and shrinkage coefficient obtained by the proposed method were used. Squares, lozenges and circles express the data measured in the 45%, 60% and 75% relative humidity rooms, respectively. In both figures, the calculated data fit well the experimental data. It is found that the relationship between shrinkage strain and relative humidity expressed in equations (81) to (85) are adequate.

6. VERIFICATION OF THE PROPOSED METHOD

Fig.23 and **Fig.24** show the deformation of cylinders due to drying for the M240 and G16 mixes (one cylinder only). The other mixes were also investigated and gave similar results. Circles and squares are the experimental data measured at the center and the edge of the cylindrical specimens, respectively. The diameter and the height of cylindrical specimens are both 150 mm. The top and bottom of the specimens were sealed by resin to prevent moisture transfer. These data were measured in the 45% relative humidity room. The curves in these figures were calculated by the finite element method by using the diffusion, film and shrinkage coefficients described in the previous sections. The results show that the calculated values fit the experimental data very well, and demonstrate the validity and the applicability of the proposed method.

7. CONCLUSIONS

An experimental approach and a numerical method were proposed to obtain the diffusion, film and shrinkage coefficients of cement-based materials. The advantage of the proposed method is the direct determination of the diffusion coefficient as function of relative humidity of cement-based materials without desorption isotherms. The results obtained from the

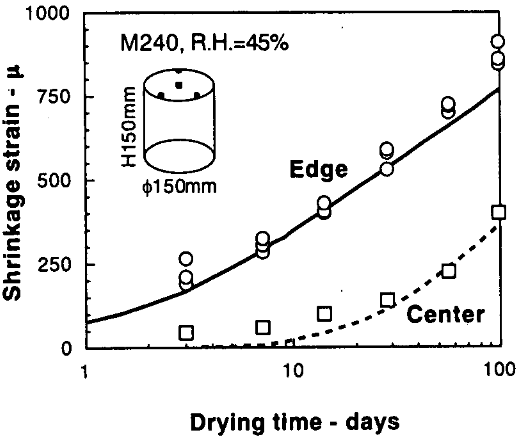

Fig.23 Shrinkage strain of cylinder specimen (M240)

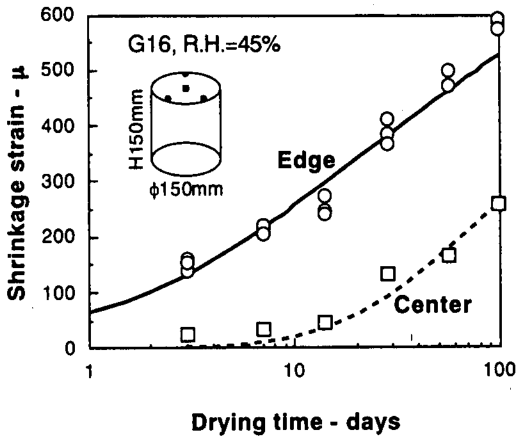

Fig.24 Shrinkage strain of cylinder specimen (G16)

experiments with sliced specimens confirm that the influence of the gaps between slices on the transfer of moisture can be neglected. Therefore, the diffusion coefficient and film coefficient as function of relative humidity can be determined by the sliced specimens without the consideration of the effect of gaps. A new numerical method to obtain the diffusion coefficient from the moisture distribution or relative humidity distribution is based on the weighted residual method combined with a nonlinear least square method. The shrinkage coefficient was obtained from solid specimens. The thickness of the specimen satisfies Bernoulli's hypothesis. It was shown that the relationship between shrinkage strain and relative humidity of solid specimens could be expressed by a power law. Three mortars with different mix proportions and two concretes with aggregates of different maximum size were used. The values of the diffusion, film and shrinkage coefficients can be obtained in all types of cement-based materials. The results demonstrate the applicability of the method proposed in this contribution.

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コンクリート中の水分分布の変化に伴う変形挙動と その数値解析のための諸係数の決定方法の提案

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本論文は，コンクリートの乾燥によって生じる変形挙動を数値解析する際に必要な拡散係数，フィルム係数および収縮係数を合理的に求めるための新たな手法の提案を行ったものである．コンクリート中の水分量自身に依存する拡散係数は，水分量のみならず湿度の関数としても求めることが可能である．本論文で提案する手法では薄くスライスした供試体を用いて湿度分布を求めている．従って，スライス供試体間の空気層がコンクリート中の水分移動に及ぼす影響についても考察を行い，その影響が極めて小さいことを示し本論文で提案する手法の妥当性を論じた．また，実験結果を基に逆解析によって拡散係数および収縮係数を求めるための数学的な手法の提案も行った．