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# FACTORS AFFECTING CONCRETE CARBONATION RATIO

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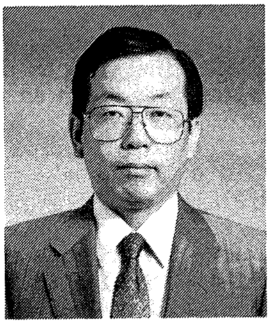

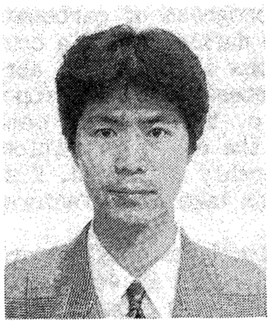

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## SYNOPSIS

This paper highlights carbonation as one of the major causes of concrete deterioration. Large amounts of field data were collected and accelerated carbonation tests were carried out in the laboratory. This led to many formulas describing carbonation rate. However, most of them contain water-cement ratio and time as the only parameters affecting carbonation. Intuitively, carbonation would seem to be affected by $CO_2$ concentration and environmental conditions, also. Hence, in this research, the effects of $CO_2$ concentration on the carbonation rate of concrete are clarified through experiments. Along with a comparative study of the data available in the literature led to the introduction of a new equation for the carbonation rate. This equation considers, as parameters, the concentration of $CO_2$ and the temperature in addition to the water-cement ratio. The equation is employed in correlating accelerated and natural carbonation, and good performance was verified.

keywords : durability, carbonation, carbon dioxide, concentration, temperature, water-cement ratio

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## 1. Introduction

The reinforcing bars in concrete structures are generally protected by the strong alkaline environment within the concrete; however, when carbon dioxide ($CO_2$) diffuses from the atmosphere into the concrete, the concrete itself is gradually carbonated from the surface inwards. When this extends to the vicinity of reinforcing bars in the interior, the bars become susceptible to corrosion. Hence, the progress of carbonation in concrete is extremely important when studying the durability of a concrete structure, and there have been numerous investigations in Japan and abroad on this subject. The effects of factors related to materials and mixproportions on carbonation rate, such as the water-cement ratio, the type of cement and aggregate, the type and dosage of admixtures, the curing conditions, and the degree of consolidation etc. have been considerably clarified. Kishitani's, Shirayama's, and Yoda's carbonation rate equations take these factors into account[1][2][3].

With conventional carbonation rate equations, the depth of carbonation below the concrete's surface is expressed by the equation given below; that is, the carbonation depth is proportional to the square root of elapsed time[4].

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

where, X=carbonation depth, t=elapsed time, and A=carbonation coefficient.

Equation (1) is called the Square-Root-t-Law, and is used quite generally. A is a coefficient experimentally obtained from various factors affecting the carbonation rate. The example of Kishitani's equation -the most commonly used- is shown here.

$$X = R(W/C - 0.25)\ /0.3\ (1.15 + 3\ W/C) \cdot \sqrt{t} \qquad (W/C \geqq 0.6)$$

$$X = R(4.6\ W/C - 1.76)\ /7.2\ \cdot \sqrt{t} \qquad (W/C \leqq 0.6)$$

where, X=carbonation depth (cm), W/C=water-cement ratio, t=period (years) of carbonation to X, and R is a constant determined by the type of cement, the type of aggregate, and the type of admixture (air-entraining agent, air-entraining-water-reducing agent).

Shirayama's equation has the same form as Kishitani's equation while Yoda's equation is also sometimes used. These carbonation rate formulas are based on Japanese environmental conditions, and since the $CO_2$ concentration in the atmosphere is constant at all times no $CO_2$ concentration factor is included; neither are the influences of temperature or humidity -other environmental factors- taken into account. Some research reports, however, have shown that when the temperature is varied, carbonation progresses more rapidly the higher the temperature[5][6]. Consequently, it can be said that the conventional carbonation rate formulas use experimentally evaluated coefficients, and give estimates of a general nature expressed as functions of the water-cement ratio. Since carbonation is a very slow process in a natural environment, accelerated carbonation tests with increased $CO_2$ concentration and higher temperatures are performed when evaluating the durability of concrete. However, in these accelerated tests carried out at various research institutions, test conditions such as $CO_2$ concentration, temperature, and humidity differ according to the individual institution, and no standard testing method has been established. Thus it can be said that the effects of environmental conditions, and particularly $CO_2$ concentration, temperature, and humidity, on the carbonation rate of concrete are not clear.

In this paper, having experimentally evaluated the effects of $CO_2$ concentration on carbonation rate in the case of plain concrete, we propose a carbonation rate equation. We used plain concrete because it has been reported that concrete with an air content of around 4% (using an air-entraining agent or air-entraining water reducing agent) has roughly the same carbonation rate as plain concrete; the susceptibility to carbonation resulting from the increased air content is offset by the change in density due to dispersion of the cement. This correlation commonly acknowledged today[7]. Using our equation, comparisons were made with test results obtained from various research institutions and a carbonation rate formula including the effects of temperature and water-cement ratio as well as $CO_2$ concentration was proposed. Results indicate that the correlation between accelerated carbonation at the various research institutions and the progress of carbonation under natural indoor conditions can be explained to a certain extent.

## 2. Outline of experiments

### 2.1 Materials and mix proportions of concrete

The cement used was ordinary portland cement; the coarse aggregate was crushed stone from Ryogami, Chichibu, Saitama Prepecture (SSD specific gravity: 2.69; FM: 6.80), and the fine aggregate was river sand (SSD specific gravity: 2.60; FM: 2.86). Plain concrete with a slump of 8cm and constant unit water content was used, while the water-cement ratio was varied between 0.70, 0.60, and 0.50. The mix proportions of the concrete are shown in Table 1.

Table 1 Mix Proportions of Concrete

| W/C | s/a (%) | Cement (kg/m³) | Water (kg/m³) | Coarse Agg. (kg/m³) | Fine Agg. (kg/m³) | Initial curing period |
|---|---|---|---|---|---|---|
| 0.70 | 49 | 263 | 184 | 978 | 908 | 0<br>and<br>5 days |
| 0.60 | 47 | 307 | 184 | 996 | 854 | |
| 0.50 | 45 | 368 | 184 | 1005 | 795 | |

### 2.2 Carbonation tests

The specimens were cylinders of diameter 10cm and height 20cm. After stripping 48 hours after casting, these were initially cured in water for periods of 0 to 5 days. After initial curing in water, the specimens were left standing in environments with $CO_2$ concentrations of 0.07% (natural indoor exposure), 1.0%, and 10% (accelerated carbonation), as measured in the laboratory under constant temperature (20℃) and relative humidity (55%). The carbonated depth was measured by splitting the specimens after compressive strength tests in a manner; this yielded fracture surfaces of 10× 20cm, which were immediately sprayed with 1% phenolphthalein solution. Any uncolored area was taken as carbonated. Measurements using calipers were taken at five points from each edge of the split surface, a total of 20 points, and the average of these was taken as the carbonated depth.

## 3. Experimental results and discussion

### 3.1 Compressive strength and weight loss ratio of concrete

Figure (1) shows the results of compressive strength tests with different $CO_2$ concentrations. Strength increases with higher $CO_2$ concentration and age. Specimens subjected to accelerated carbonation with $CO_2$ concentrations 1% and 10% had greater strength when compared with standard curing. Specimens subjected to natural indoor exposure, although undergoing slow carbonation, did not show an increase in strength due to carbonation; however, after the age of 8 weeks, their strength became roughly constant or decreased.

Figure (2) shows the results concerning weight loss of concrete due to differences in $CO_2$ concentration. In spite of identical temperature and humidity, lower weight loss was seen with higher $CO_2$ concentration. This is thought to be because diffusion of moisture within the specimens is inhibited when the compressive strength of carbonated portions increases at higher $CO_2$ concenrations, as shown in Fig.(1) [8].

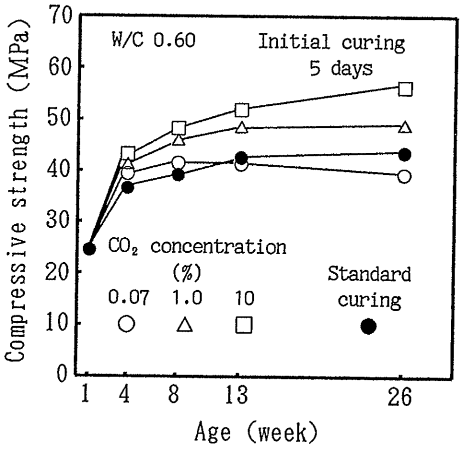

Fig.1 Compressive strength of concrete

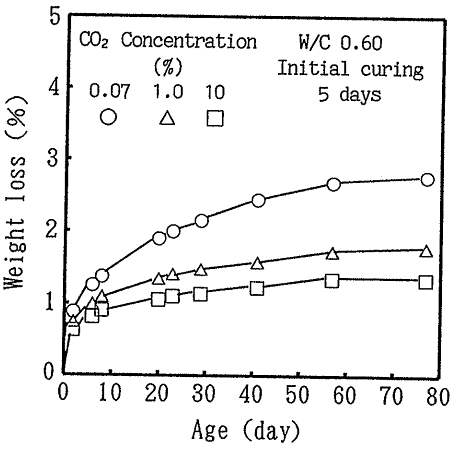

Fig.2 Weight of concrete

### 3.2 Carbonation depth

Time-dependent changes in carbonation depth by water-cement ratio for different $CO_2$ concenrations are shown in Fig.(3). As indicated by Eq.(1), the carbonation depth is proportional to the square root of elapsed time, and despite variations in $CO_2$ concentration, it was ascertained that carbonation depth was roughly in proportion to the square root of elapsed time. Further, for the same concentration, it can be seen that carbonation rate increases with higher water-cement ratio. The effect of water-cement ratio on carbonation rate is greater than usual because the conventional carbonation rate formulas are expressed in terms of functions of the water-cement ratio. Concerning the effects of initial curing, specimens not cured at all exhibited considerable progress of carbonation compared with those cured for 5 days in water; at the same age, the carbonation depth of specimens with zero initial curing was approximately 1.9 to 2.4 times that of those cured for 5 days. Further, it is known that when initial curing extends for more than 5 days, the effects of the curing period on carbonation depth are relatively small [9]. The water-cement

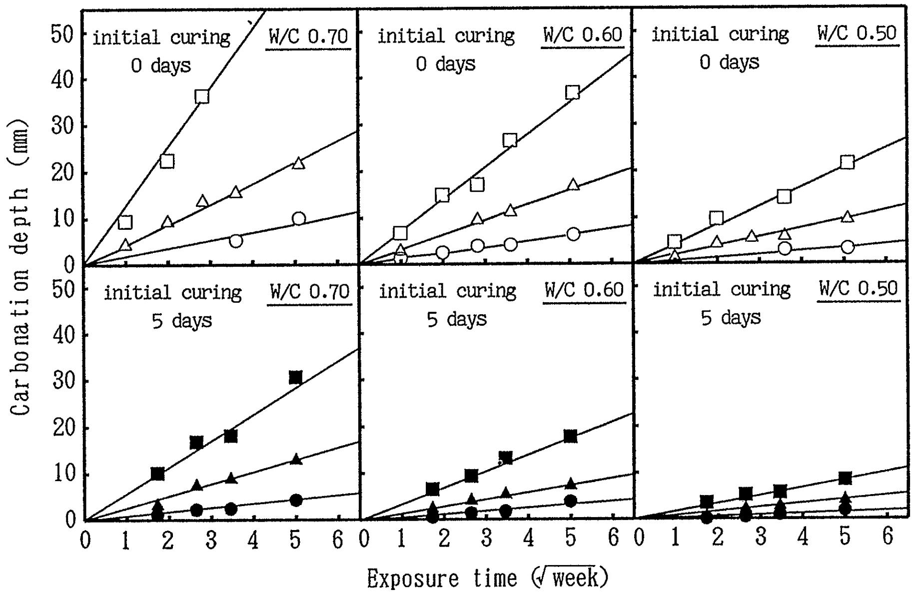

Fig.3 Progress of carbonation under various accelerated conditions ( $CO_2$: ○●: 0.07%, △▲: 1.0%, □■: 10% )

ratio determines the density of concrete and, as we know, it is difficult for $CO_2$ to diffuse in dense concrete structures.

Figure (3) shows the relationship between $CO_2$ concentration and carbonation coefficient for various concretes by the least squares method. Table 2 shows the level of carbonation . According to the table, the correlation coefficient is beter than 0.95 for all the concretes. On looking at the effects of $CO_2$ concentration, the time taken for carbonation to reach the same depth as in the case of accelerated tests with a $CO_2$ concentration of 10% is 33 to 49 times for speciments naturally exposed indoors, and 7 to 11 times for specimens accelerated at 1%.

Table 2 Carbonation coefficient in study

| Initial curing period (day) | W/C | Carbonation coefficient (mm/√week) | | |
|---|---|---|---|---|
| | | $CO_2$ concentration (%) | | |
| | | 0.07 (indoor) | 1.0 | 10 |
| 0 | 0.70 | 1.78<br>(0.977) | 4.63<br>(0.995) | 11.0<br>(0.996) |
| | 0.60 | 1.23<br>(0.994) | 3.29<br>(0.997) | 7.05<br>(0.994) |
| | 0.50 | 0.67<br>(0.972) | 1.87<br>(0.978) | 4.14<br>(0.988) |
| 5 | 0.70 | 0.79<br>(0.988) | 2.62<br>(0.983) | 5.51<br>(0.985) |
| | 0.60 | 0.62<br>(0.924) | 1.60<br>(0.999) | 3.61<br>(0.998) |
| | 0.50 | 0.27<br>(0.824) | 0.82<br>(0.959) | 1.72<br>(0.992) |

(     )=Coefficient of correlation

Figure (4) shows the relationship between carbonation coefficient and concentration of $CO_2$ under natural indoor exposure. This coefficient is more or less constant regardless of the water-cement ratio and the initial curing conditions. This means that the acceleration is constant at all times, regardless of the water-cement ratio, initial curing conditions, and type of concrete. Since these are experimental results for fixed temperature and humidity conditions, it may be said that this ratio is effect of the $CO_2$ concentration on carbonation rate.

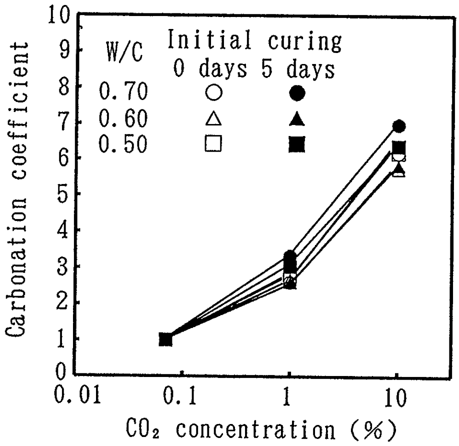

Fig.4 Relationship between concentration of $CO_2$ and carbonation coefficient

## 4. Influence of various factors on carbonation rate

### 4.1 Effect of $CO_2$ concentration on concrete carbonation rate

The following equation is valid if it is assumed that carbonation occurs due to diffusion of a $CO_2$ concentration (C) steadily through the concrete[10][11].

$$dX/dt = k \cdot C/X \quad \cdots\cdots\cdots\cdots (2)$$

where, X=carbonation depth, t=elapsed time, k=coeficient of proportionality, and C=$CO_2$ concentration. On solving this diferential equation, the follwing equation applies when time is 0 (t=0), with carbonation depth as X=0:

$$X = k\sqrt{C \cdot t} \quad \cdots\cdots\cdots\cdots (3)$$

The conventional carbonation rate formula is for natural conditions. Since the concentration in the atmosphere is constant value at all times, no term for $CO_2$ concentration is included. The conventional formula results if (C) is made a constant in Eq.(3). On this basis, where the properties of the concrete do not vary according to differences in $CO_2$ concentration, it is considered that the carbonation depth of concrete is proportional to $\sqrt{C \cdot t}$ and the proportional coeficient k, when the depth for natural indoor exposure is taken as 1. The results are shown in Fig.(5). As shown by this figure, the value of k is not constant and becomes smaller with higher $CO_2$ concentration. This is because the quality of the concrete through which $CO_2$ has passed varies due to differences in $CO_2$ concentration; this can be seen in the compressive strength results in Fig.(1) and the results of weight loss measurements in Fig.(2).

This led us to hypothesize that the value of k, as a term dependent environmental conditions, could be expressed in terms of $CO_2$ concentration ($k_{CO2}$), temperature ($k_T$), and humidity ($k_H$), plas a term dependent on materials and mix proportions, such as water-cement ratio, ($k_W$).

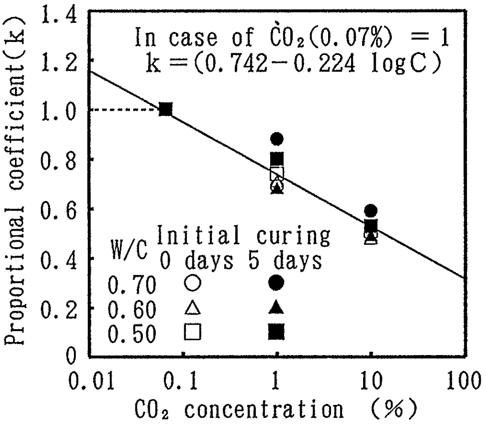

Fig.5 Relationship between concentration and proportional coefficient(k)

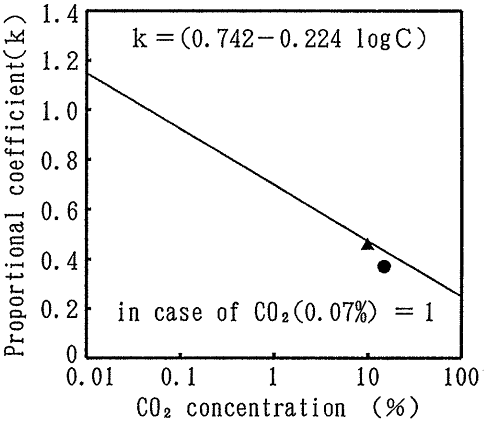

Fig.6 Proportional coefficient from results of experiments elsewhere

$$k = k_{CO2} \cdot k_T \cdot k_H \cdot k_W \quad \cdots\cdots\cdots\cdots (4)$$

Here, with regard to $k_T$ , $k_H$ , and $k_W$ , these are constant values since the conditions are constant. On the other hand, since $k_{CO2}$ is not constant for the reasons given earlier, its value was expressed by the empirical equation relating it to the $CO_2$ concentration.

where, $k$=1 when the $CO_2$ concentration $C$ is 0.07%.

$$k_{CO2} = (0.742 - 0.224 \log C) \quad \cdots\cdots\cdots\cdots (5)$$

Adopting the above relationship, the carbonation rate formula becomes the following:

$$X = (0.742 - 0.224 \log C)\ A\sqrt{(C/0.07) \cdot t}$$

$$= (2.804 - 0.847 \log C)\ A\sqrt{C \cdot t} \quad \cdots\cdots\cdots\cdots (6)$$

where, $X$=carbonation depth (mm), $C$=$CO_2$ concentration (%), $t$=elapsed time (weeks), and $A$=carbonation coefficient under natural indoor exposure conditions (in this case, $C$=0.07%)

In past accelerated carbonation tests, the part of the proportional coefficient dependent on $CO_2$ concentration was incorporated in the coefficient $A$ given in Eq.(6), so it was unclear how many times faster the accelerated tests proceeded compared with actual conditions. Further, Eq.(6) is a formula for the case of indoor natural exposure -$CO_2$ concentration 0.07%- and the time required for carbonation to reach the same depth as in an outdoor environment ($CO_2$ concentration 0.0351%[12]) is approximately 1.8 times. However, this difference is strictly dependent on $CO_2$ concentration only, the assumption being that temperature and humidity, the other environmental factors, are the same as in accelerated carbonation tests. Actually, concrete in exposed outdoors will be subject to variations in temperature and humidity, as well as repeated wetting and drying due to rain and other factors, and it is known that progress of carbonation is even more restrained in these circumstances[13]. Furthermore, it is natural to assume that these effects will differ according to regional characteristics. However, the carbonation coefficients obtained experimentally which consider both the effects of differences in $CO_2$ concentration between inside and outside and repeated wetting and drying, it is believed that the effects of $CO_2$ concentration have at least been made clear. Fig.(6) shows a comparison with the results of the few past experiments in which indoor exposure and accelerated tests with identical materials were carried out. As the figure shows, the results fall roughly on the curve of the experimental equation proposed by the authors[4][5].

### 4.2 Prediction of carbonation rate in indoor exposure according to past accelerated carbonation tests

Accelerated carbonation tests performed at various research institutions (a total of 34 establishments) are shown in Fig.(7) and Table 3 gives the conditions for these carbonation tests[4]-[9],[13]-[40]. Water-cement ratio is plotted on the abscissa and carbonation coefficients -carbonation depth divided by the square root of elapsed time- are on the ordinate. The test conditions range from a $CO_2$ concentration representing indoor exposure conditions ($CO_2$ concentration of about 0.1%) to 100%, as shown in Table 2. With regard to temperature, this was varied over the range 10 ℃ to 40℃, while relative humidity was in the range 40 to 60%, where carbonation is thought to be the most rapid. It should be noted that, in all studies, testing was done under constant $CO_2$ concentration, temperature, and humidity conditions.

Table 3 Conditions of carbonation tests

| Environmental conditions | | Materials | |
|---|---|---|---|
| $CO_2$ Concentration | Constant concentration (indoor～100%) | Cement | Ordinary portland cement |
| Temperature | Constant temperatue (10℃～40℃) | Aggregate | River sand, crushed stone |
| Relative humidity | Constant humidity (40%RH～60%RH) | Water-cement ratio | 0.30 ～ 0.80 |
| Initial curing period (standard curing) | 5 days ～ 91 days | Chemical admixture | Plain, air-entraining,<br>AE water-reducing agent,<br>superplasticizer |
| Initial dry-curing period | 0 Days ～ 28 Days (before carbonation test) | | |

All the experimental date shown in Fig.(7) was converted to represent the $CO_2$ concentration in an indoor natural environment (in this case, 0.07% by Eq.(6) proposed previously), in effect predicting values of carbonation rate under natural indoor exposure conditions. This is shown in Fig.(8). When the $CO_2$ concentration factor is eliminated in this way, the values fall in more or less the the same range. However, the carbonation coefficients plotted here are predicted values of the carbonation rate for the temperature and humidity conditions of the particular tests, so they include the influence on carbonation rate of differences in temperature, humidity, and initial curing conditions which are intrinsic properties of the materials and mix proportions.

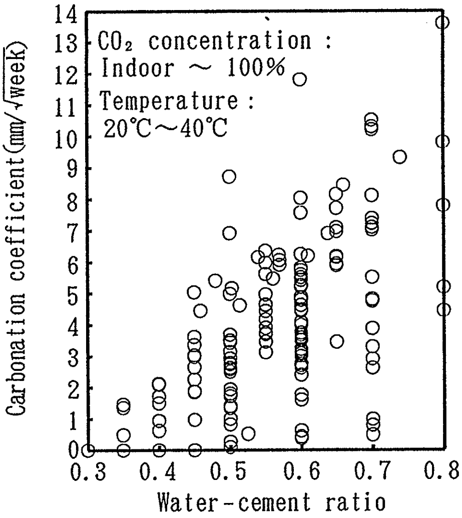

Fig.7 Results of accelerated carbonation tests conducted elsewhere

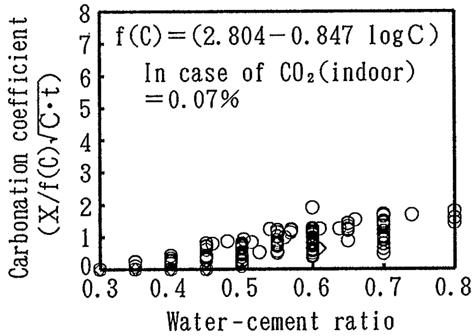

Fig.8 Calculated coefficient of carbonation in indoor exposure from results of accelerated carbonation tests (considering only $CO_2$)

### 4.3 Influence of temperature on carbonation rate

To investigate the effect of temperature on the carbonation rate in detail, the results are plotted in terms of temperature in Fig.(9). As this figure shows, the carbonation coefficient is greater when the temperature is high. It has been reported in a past study under constant $CO_2$ concentration (10%) and relatve humidity (80%) that the carbonation coefficient at a temperature of 40℃ is double that at a temperature of 20℃[5]. Further, according to a separate study, under constant $CO_2$ concentration of 5% and relative humidity of 60%, carbonation at a temperature of 30℃ was approximately 1.7 times faster than that at 10℃[6].

Since carbonation is a reaction between atmospheric carbon dioxide and calcium hydroxide, which is a hydration product contained in the concrete, it progresses through chemical reaction. The influence of temperature on chemical reaction rates is described by Arrehenius'theory, which states that at higher temperature a reaction will progress further. Hence, for the data shown in Fig.(9), an Arrhenius plot with the carbonation coefficient at 20℃ normalized to unity is shown in Fig.(10). If the inverse of absolute temperature is plotted on the abscissa and the logarithm of carbonation coefficient on the ordinate, Arrhenius'law has been followed if a straight-line relationship is obtained. Fig.(10) indicates a fair amount of variation depending on the data. However, if it is assumed that Arrhenius'law is holds, the influence of temperature on carbonation rate is as follows:

$$\ln k_T = (8.748 - 2563/T)$$

$$k_T = e^{(8.748-2563/T)} \quad \cdots\cdots\cdots\cdots (7)$$

where, T=absolute temperature(k).

From Eq.(7), the carbonation coefficients at temperatures of 30 ℃ and 40 ℃ will be 1.33 times and 1.75 times that at a temperature of 20℃, respectively.

This gives approximate agreement with the past experimental results mentioned above. The activated energy, according to Arrhenius' theory, is then $2.13\times10^{-4}$ J $\mathrm{mol}^{-1}$[41].

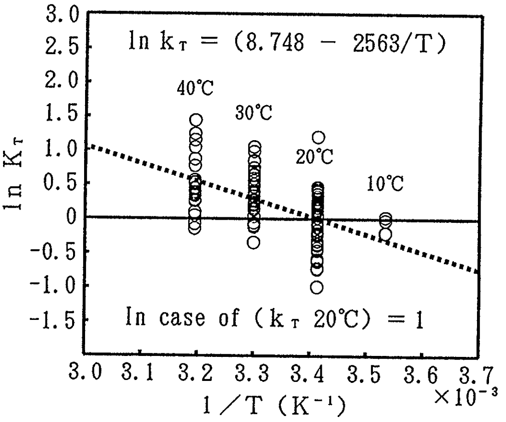

Fig.10 Arrhenius plot

The effects of temperature in Fig.(9) were converted to 20℃ and the carbonation coefficient for indoor exposure was calculated. The values obtained are shown in Fig.(11). By eliminating the influence of temperature, the range plotted in Fig.(9) has been reduced. Further, the carbonation rate coefficient plotted here is a predicted carbonation rate for a constant $CO_2$ concentration of 0.07% and temperature of 20℃, and the effect of humidity is included. However, since the test results are for humidity conditions of 40% to 60% RH, it may be assumed that the effect of temperature is reduced. That is to say, the gradient shown in Fig.(11) is mainly the effect of water-cement ratio on the carbonation rate.

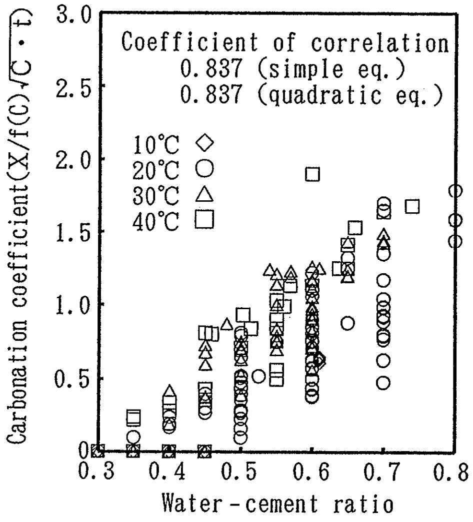

Fig.9 Effect of temperature on calculated carbonation coefficient in indoor exposure

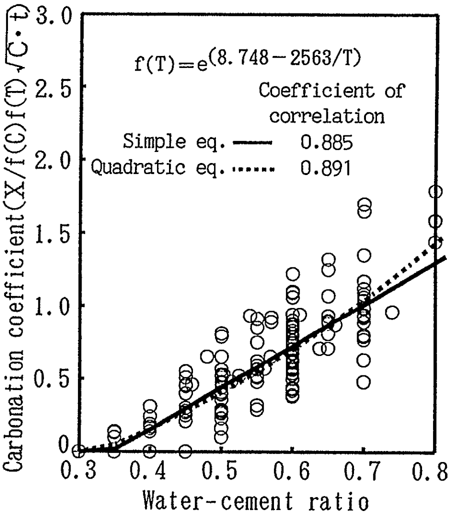

Fig.11 Calculated coefficient of carbonation in indoor exposure from results of accelerated carbonation tests (considering $CO_2$ and temperature)

## 4.4 Influence of water-cement ratio carbonation rate

The influence of water-cement ratio on carbonation rate has been studied in great depth. Here, the influence of water-cement ratio on Kishitani's, Shirayama's, and Yoda's equations and simple equation and quadratic equation for obtaining the effects of water-cement ratio from Fig.(11) are compared and the results are shown in Fig.(12). It is clear from the figure that both simple and quadratic equations agree well with the influence of water-cement ratio on carbonation rate. In other words, this suggests Eq.(6) and Eq.(7) which include $CO_2$ concentration and shows influence of temperature respectively. Carbonation rate formulas including $CO_2$ concentration, temperature, and water-cement ratio are given below. In these equations, the $CO_2$ concentration ranges from indoor exposure to 100%, the temperature from 10 ℃ to 40℃, and the water-cement ratio from 30% to 80%.

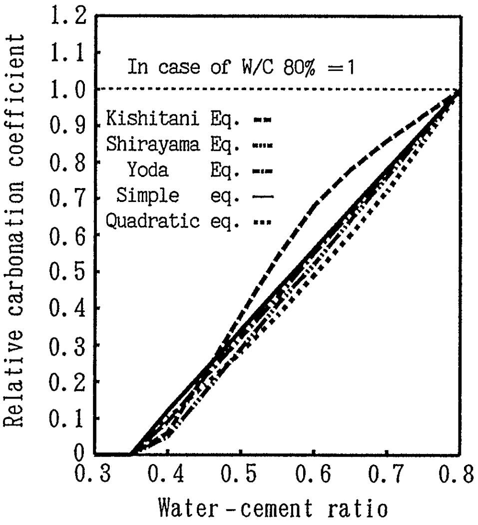

Fig.12 Effect of water-cement ratio on carbonation rate in concrete

- Simple equation of water-cement ratio

$$X=(2.804-0.847\log C)\cdot e^{(8.748-2563/T)}$$

$$\times(2.94\,WC-101.2)\times10^{-2}\times\sqrt{C\cdot t} \quad \cdots\cdots\cdots\cdots (8)$$

- Quadratic equation of water-cement ratio

$$X=(2.804-0.847\log C)\cdot e^{(8.748-2563/T)}$$

$$\times(2.39\,WC^2+44.6\,WC-3980)\times10^{-4}\times\sqrt{C\cdot t} \quad \cdots\cdots\cdots\cdots (9)$$

where, $X$=carbonation depth(mm), $C$=$CO_2$ concentration (%), $T$=absolute temperature (k), $WC$=water-cement ratio (%),and $t$=elapsed time (weeks).

## 4.5 Comparison of accelerated carbonation tests and calculated values

A comparison of experimental values of carbonation coefficient obtained in accelerated carbonation tests at various research institutions and values calculated from Eq.(9) is shown in Fig.(13). For past equations, where $CO_2$ concentration and temperature are not considered, the graph is not uniform. Whereas, using the equations proposed here, experimental and calculated values are within the range ±40%. Thus, by using Eq.(9), the correlation between accelerated carbonation and actual carbonation during indoor exposure can be described to a certain extent.

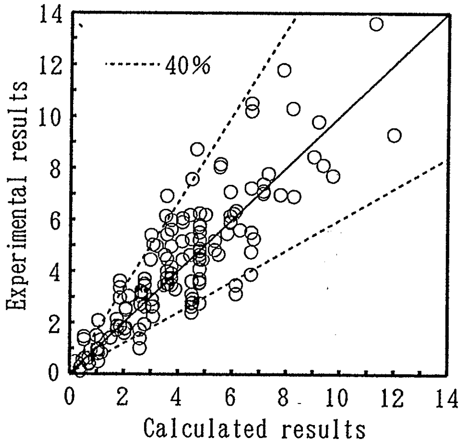

Fig.13 Comparison of experimental and calculated carbonation coefficients

## 5. Conclusions

The study reported here concerns carbonation tests carried out under different $CO_2$ concentrations. The aim was to explain the effects of $CO_2$ concentration on carbonation rate, verifying the results by contrasting them with past accelerated carbonation test results. The results attained in the study are as follows.

(1) As a result of carbonation tests in environments with different $CO_2$ concentrations, the multiplying factor for carbonation due to $CO_2$ concentration was determined. The effect of $CO_2$ concentration on carbonation rate is expressed as shown in Eq.(6), as a carbonation rate formula which includes a term for $CO_2$ concentration.

(2) By eliminating the effects of $CO_2$ concentration indicated in Eq.(6), it was possible to determine the effects of temperature on carbonation rate using Arrhenius' law.

(3) By clarifying the effects of $CO_2$ concentration and temperature on carbonation rate where effects of other factors on carbonation rate were expressed as functions of water-cement ratio.

(4) By making comparing the results of accelerated carbonation tests with the carbonation rate formula (including the effects of $CO_2$ concentration) proposed in this paper, the carbonation rate formulas given in Eq.(8) or (9) were proposed. This take account of factors other than $CO_2$ concentration, such as temperature and water-cement ratio.

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