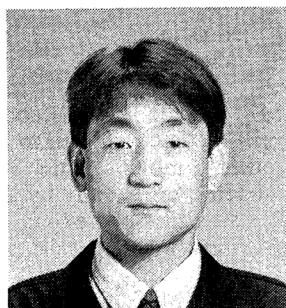
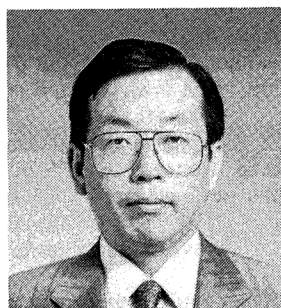


A New Evaluation System to Quantitatively Predict the Progress of Alkali-Silica Reaction

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Taketo UOMOTO

SYNOPSIS

A new test system to quantitatively evaluate alkali-silica reactivity of aggregate is proposed. In this test system, at first, change of soluble silica content of aggregate with the passage of time is measured by a leaching test at various temperatures. The results obtained from this test are analyzed on the basis of kinetics in order to obtain the diffusion coefficient of alkali in aggregate and alkali-silica ratio of reaction products, and also to evaluate the effect of temperature on the diffusion coefficient. Expansion behavior and ultimate expansion ratio of mortar bars can be calculated by using these values. Expansion curves of mortar bars observed experimentally almost coincide with the calculated ones. It was also found that the diffusion coefficient of alkali in sedimentary rocks change during ASR.

Keywords: alkali-silica reaction, model, kinetics, diffusion theory, temperature, leaching test, mortar-bar test

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1. INTRODUCTION

A mortar-bar test defined as ASTM C 227 and JIS A 5308 has been widely used to evaluate the alkali silica reactivity of aggregate. However, in the test, evaluation of the reactivity of aggregate is dependent on whether the expansion ratio of mortar bars made with the tested aggregate exceeds the criteria or not at each given period (0.05% at 3 months and 0.1% at 6 months). Thus, an increasing tendency of expansion cannot be considered in evaluating the reactivity in the mortar bar test. The aggregates can be mainly divided into two groups according to the differences in the expansion behavior observed during the mortar bar test. One aggregate group shows the expansion behavior in which the expansion rate reduces with time (aggregate group showing decreasing rate expansion), and the other shows the expansion behavior in which the expansion rate is constant or increasing with time (aggregate group showing constant rate expansion). Two typical expansion curves are schematically shown in figure 1. In this example, both aggregates are evaluated as inert because the expansion ratios of mortar bars cannot exceed 0.1% at 6 months. For the aggregate showing decreasing rate expansion, the judgment is considered as proper because the expansion ratio after 6 months is expected to be kept below 0.1%. However, for the aggregate showing constant rate expansion, the judgment is given as inert although the aggregate is considered to have the potential reactivity such as it exceeds 0.1% immediately after 6 months which may induce more expansion. Thus, such aggregates should be evaluated synthetically by using other values such as the ultimate expansion ratio and the period at which the expansion rate becomes sufficiently low. However, a problem is that it takes longer time to get these values.

On the other hand, a quick chemical test defined as ASTM C 289 and JIS A 5308 is also widely used to evaluate the reactivity of aggregate in a short time, and a good correlation between the judgments given by the mortar bar test and given by the quick chemical test has been experimentally recognized. This fact indicates that the expansion ratio of mortar bar at 6 months can be predicted by the result of the quick chemical test at least in a semi-quantitative manner. The quick chemical test is one of the leaching tests performed under relatively high temperature to accelerate reaction. Thus, it is possible to establish a more quantitative test method to evaluate the reactivity of aggregate in a short time by improving the leaching test.

The authors already proposed a kinetics-based model which quantitatively predicts the progress of alkali-silica reaction (ASR) and ensuing expansion due to ASR, and succeeded to explain the mechanism of the complicated expansion behavior such as pessimum effect of reactive aggregate content[1]. And, it was shown that the expansion curves of mortar bars made with the aggregates showing decreasing rate expansion could be simulated by analyzing the results from the quick chemical test[1].

In this paper, the authors propose a new test system which can evaluate the reactivity of aggregate quantitatively in a short time. In this system, the modified leaching test is used to detect the progress of ASR, then the results are analyzed on the basis of the previously proposed model.

2. A KINETICS-BASED MODEL AND UNKNOWN FACTORS

The mortar bar test can show the physical value observed as expansion, and it is expected to have close relationship with the actual damage in concrete structures due to ASR. Therefore, the authors urgently decided to establish the evaluation system to predict the progress of ASR under the mortar bar test condition, and the predicted results are verified by comparing to the experimentally observed expansion behaviors.

In order to analyze the progress of ASR, the previously proposed model is applied. This model is based on some assumptions. For example, the rate determining stage of ASR is assumed as the diffusion of alkali from surface into the reactive aggregate, and the rate of

reaction can be approximated by the following equation which is derived from Fick's first law.

$$dx/dt = Ck/x. \quad (1)$$

Here t , x , k and C represent time (hr), thickness of reacted layer (cm), diffusion coefficient of alkali ions in the reactive aggregate (cm^2/hr), and alkali concentration in pore water of cement matrix (mol/l), respectively. The k value, diffusion coefficient of alkali ions in reactive aggregate, is an important factor to evaluate the reactivity of aggregate and should be defined experimentally.

On the other hand, since the alkali concentration in pore water C reduces with the progress of ASR, value C must be expressed as a function of the total amount and alkali silica ratio of reaction products.

$$C = f(Tpr, RS). \quad (2)$$

Here, Tpr and RS represent the total amount of reaction products ($mol/l-mortar$) and alkali silica ratio of reaction products, respectively. RS is considered to be depending on type of aggregate and it also should be defined in experimental manner.

Therefore, in this model, the following factors are defined as unknown factors which should be obtained experimentally.

- i) Diffusion coefficient of alkali ions in the reactive aggregate.
- ii) Alkali silica ratio of the reaction products.

In the previous paper [1], these factors were obtained mathematically by comparing to the observed expansion behavior of mortar bars. However, in this paper, the establishment of a new evaluation system will be discussed and the way to define these factors experimentally will be shown.

3. METHOD TO DEFINE UNKNOWN FACTORS

The leaching test to define the unknown factors is performed basically in accordance with the quick chemical test, ASTM C 289 and JIS A 5308. However, the amount of soluble silica and consumed alkali are measured with the passage of time. Furthermore, in order to evaluate the effect of temperature on alkali diffusion coefficient, the test is performed at various temperatures.

Following equation can be obtained by integrating equation (1).

$$x = (2kCt)^{1/2}. \quad (3)$$

Since change of soluble silica contents of aggregate and the amount of consumed alkali with the passage of time are measured by the leaching test, alkali ion concentration C and alkali-silica ratio of reaction products (RS) are obtained directly.

On the other hand, as shown in equation (3), value $(2k)^{1/2}$ can be defined as the slope of the relationship between x and Ct as long as the linear relationship can be observed. Thus, if value x can be obtained, value k can also be calculated graphically from this relationship. Value x , thickness of reacted layer, can be calculated by the following procedure.

By assuming that the shape of aggregate is sufficiently approximated as sphere with equivalent radius R (cm), volume reaction ratio (α) of aggregate can be written as,

$$\alpha = 1 - (1 - x/R)^3. \quad (4)$$

α can be calculated by following equation at each given period under the quick chemical test condition .

$$\alpha = Sc * 60.08 / 10^6 \quad (5)$$

Where, Sc indicates the amount of dissolved silica at each given period ($mmol/l$), and 60.08 indicates the molecular weight of SiO_2 .

Thus, x value can be obtained by the following equation derived from equation (4).

$$x = R(1 - (1 - \alpha)^{1/3}) \quad (6)$$

4. EFFECT OF TEMPERATURE ON DIFFUSION COEFFICIENT OF ALKALI IONS

The leaching test is performed under rather high temperature compared to the mortar bar test. Thus, in order to know the rate of reaction at the temperature of mortar bar test condition, the effect of temperature should be evaluated in a quantitative manner.

It is widely known that a lot of chemical reactions are accelerated by increase in temperature. And it is also widely recognized that the effect of temperature on the reaction rate can be expressed by following Arrhenius's equation as the variation of logarithm of rate constant to temperature.

$$\log kr = (-Ea/RaT) + \log F. \quad (7)$$

Here kr, F, Ea, Ra and T represent rate constant, frequency factor, gaseous constant and absolute temperature, respectively.

In case of ASR, it is considered that the effect of temperature can be evaluated as the variation of diffusion coefficient of alkali ions in accordance with Arrhenius's equation.

5. TESTED AGGREGATES AND LEACHING TEST CONDITIONS

As mentioned previously, the reactive aggregates can be divided into two groups as the group showing constant rate expansion and the group showing decreasing rate expansion. The evaluation system proposed here should have sufficient accuracy in predicting both expansion behavior. Hence, the aggregate from both groups whose expansion behavior were already reported by some researchers are tested in this work. They are 4 sedimental rocks which are reported as showing constant rate expansion by Tamura et al [2] and one andesite from Setouchi area which is widely known as showing decreasing rate expansion. And quartz glass particle as a model of homogeneous aggregate is also tested. The properties of tested aggregates are shown in Table 1.

The leaching test is performed basically in accordance with the quick chemical test defined as ASTM C 289, but under variable temperatures as 60°C, 70°C and 80°C. In order to verify the effect of alkali concentration in equation (1), two aggregates are tested with different alkali concentration. The test conditions are shown in Table 2.

6. TEST RESULTS

6.1 Reaction Mechanisms of Two Aggregate Groups

All the test results obtained under 80°C are shown as the relationship between the thickness of reacted layer (value x) and value $(Ct)^{1/2}$. Figure 2 indicates the test result of quartz glass particle. And the result for andesite is shown in Figure 3. As can be seen from these figures, the relationships between values x and $(Ct)^{1/2}$ can be approximated as the straight lines which pass through the origin of coordinate axis. Only the value x measured at the latest period for the quartz glass particle is less than the approximated value. This is because of it cannot exceed the solubility of silica. Thus, the diffusion coefficient of alkali in the aggregates showing decreasing rate expansion can be easily calculated from the slopes of these lines.

In Figures 4–7, the test results of the aggregates showing constant rate expansion are indicated. As can be seen from all these figures, the value of x increases slightly with the increase of $(Ct)^{1/2}$ at the early stage of ASR, and after that, x increases dramatically. This relationship can be approximated as a bilinear relationship. The test results indicate that the alkali diffusion coefficients in the aggregates showing constant rate expansion change during reaction. If we named the diffusion coefficient at an early stage of reaction as the initial diffusion coefficient (k_{ini}), and the changed coefficient as the post diffusion coefficient (k_{post}), the progress of the ASR of the aggregates showing constant rate expansion should be expressed by the following two equations.

Before the coefficient changes,

$$dt / dx = k_{ini} C/x \quad (8)$$

and, after it changes,

$$dt / dx = k_{post} C/x \quad (9).$$

All the diffusion coefficients obtained here are collected and shown in Table 3. It is obvious that the diffusion coefficients at the latter period are from fifteen to thirty times of that in the early stage. Furthermore, for the aggregates showing constant rate expansion, it is also found that all the values x at the point where the diffusion coefficient suddenly changes are almost same ($6-7 \times 10^{-6}$ cm). From these results and discussions, the mechanism of the quite long incubation period before expansion can be explained as the change of diffusion coefficient of alkali ion in the aggregates during reaction.

6.2 Verification for the Modeling of the Effect of Alkali Concentration

Figures 8 and 9 indicate the results of the leaching test performed under the different alkali concentration for andesite and sedimentary rock, respectively. In both cases, the relationships between the values x and $(Ct)^{1/2}$ show same tendency regardless of the difference in alkali concentration. Thus, it is confirmed that the effect of alkali concentration can be evaluated by equations (1), (8) and (9) in the model proposed here.

6.3 Effect of Temperature

The test results of andesite and sedimentary rock at various temperatures are shown in Figures 10 and 11, respectively. From these figures, it is found that slope of the lines which approximate the relationship between the values x and $(Ct)^{1/2}$ increase with an increase in temperature. And both the linearity observed in the test results of andesite and the bilinearity observed in the test results of sedimentary rock are kept with regardless of temperature changes. And it is also found that the value x at the point where the slope of line changes is same although the temperature varies. Thus, it is considered that the effect of temperature on the progress of ASR can be evaluated as the variation of the alkali diffusion coefficient to temperature without any change of mechanism. As described previously, the variation of diffusion coefficient to temperature is expressed by Arrhenius's equation. Here, the following

modified Arrhenius's equation is used to express the effect of temperature on the diffusion coefficient.

$$\log k = E_{app}(1/T) + k_{inf} \quad (10)$$

Where, E_{app} represents the activation energy and k_{inf} can be defined as a limiting value of the logarithm of diffusion coefficient. The results are plotted according to the equation (10) and shown in Figure 12. As shown in Figure 12, good linear relations corresponding to the Arrhenius's equation can be found. Because of this good linearity, the diffusion coefficient at various temperature can be calculated by using this equation.

6.4 Alkali-Silica Ratio of reaction Products

Figures 13 and 14 indicate the results of the leaching test for andesite and sedimentary rock (No.2) as the relationship between the amount of dissolved silica ($Sc; mmol/l$) and consumed alkali ($Rc; mmol/l$). As shown in these figures, the relationship is approximated as a straight line with intercept on vertical axis (R_c). The results suggest that ASR begins with large consumption of alkali without producing any significant amount of reaction products. After that, the alkali is consumed proportionally to the amount of dissolved silica.

7. CALCULATION UNDER THE MORTAR BAR TEST CONDITION

The amount of reaction products in the mortar bar can be calculated by the following equation with relation to the reactive aggregate size distribution[1].

$$T_{pr} = A \sum \alpha_i \beta_i / 60.08 \quad (11)$$

Where, T_{pr} : total amount of reaction products (mol/l -mortar)

A : amount of aggregate (g/l -mortar)

α_i : volume reaction ratio of aggregate particle with equivalent radius R_i

β_i : volume fraction of aggregate particle with equivalent radius R_i

60.08 : molecular weight of SiO_2 (g/mol)

7.1 Diffusion Coefficient of Alkali at 40°C

The mortar bar test is performed under 40°C. The diffusion coefficient of alkali in each tested aggregate at 40 °C can be calculated by equation (10) and shown in table 4.

7.2 Initial Alkali Concentration and Consumption of Alkali during Reaction

In the mortar bar test condition, the amount of cement in unit volume of mortar is defined as 600g/l, and the weight percent of alkali (Na_2O equivalent) in cement is defined as 1.2%. Thus, the total number of molecule in the unit volume of mortar is obtained as 0.232 mol/l ($600 \times 0.012 \times 2 / 62 : 62 = \text{molecular weight of } Na_2O$). On the other hand, the amount of water in unit volume of mortar is also defined as 300g. And the amount of free water (not chemically bounded water) can be calculated as 132g by subtracting the theoretically required water for hydration (28% of cement weight) from the total amount of water ($300g - (600g \times 0.28)$). From these, the alkali concentration in pore water may be obtained as 1.758 mol/l ($0.232 / 0.132$). However, of all the alkali only water soluble alkali can contribute to the increase of alkali concentration in pore water. Thus, it is important to identify the fraction of water soluble alkali to total alkali. In order to define this value, the equation presented by Diamond [3] is referred. Diamond measured the concentration of alkali ion in pore water extracted from the mortar made with cement containing different amount of alkali, and showed the relationship between the weight percent of alkali in cement and the concentration of alkali in pore water. Assuming that the fraction of water soluble alkali is 50% of the total

alkali, the alkali concentration calculated by the above mentioned procedure coincides with the value obtained by Diamond's equation. Thus, the fraction of water soluble alkali to total alkali is defined as 50% in this work. In case of mortar bar test, the amount of water soluble alkali in unit volume of mortar is obtained as $0.116 \text{ mol/l-mortar}$ (0.232×0.5).

A large alkali consumption at the beginning stage of ASR was described in the previous chapter. Therefore, in order to calculate the initial alkali concentration in pore water, this consumption also should be considered. In this work, it is assumed that the amount of initially consumed alkali is proportional to the total surface area of reactive aggregate. Thus, the amount of alkali initially consumed by unit surface of aggregate can be calculated from the intercepts of the approximated lines in Figures 13 and 14, and from the total surface area of aggregate whose weight and equivalent radius are 1000g and 0.01125cm , respectively. Since the total surface area of aggregates tested in the quick chemical test and the mortar bar test are $1046 \times 10 \text{ cm}^2$ and 519.5 cm^2 respectively, the amount of initially consumed alkali in unit volume of mortar can be calculated by the following equation.

$$C_{cons} = (519.5/1046) * R_{c_{int}} / 1000 \quad (12)$$

Where, C_{cons} indicates the amount of initially consumed alkali in unit volume of mortar (mol/l-mortar), and $R_{c_{int}}$ indicates the intercept observed in the relationship between the values R_c and S_c . From these discussions, the alkali concentration in pore water during reaction can be calculated by the following equation.

$$C = (0.116 - C_{cons} - Tpr * RS) / 0.132 \quad (13)$$

Where, RS indicates the alkali-silica ratio of reaction products observed as the slope of the approximated line for the relationship between the values R_c and S_c .

Equations (1) to (13) are applied to calculate the amount of reaction products in unit volume of mortar. As can be seen from the interrelationship between equations (1) and (13), this problem should be defined as a strong nonlinear problem. Thus, the thickness of reacted layer x is calculated using "step-by-step" integration of equation (1) with sufficiently short calculation step, referring the latest calculated value of alkali concentration.

The calculation process in predicting the amount of reaction products is summarized and shown in Figure 15.

8. COMPARISON BETWEEN CALCULATED AND OBSERVED EXPANSION BEHAVIORS

The expansion behaviors of mortar bars made with the aggregate showing constant rate expansion are collected and shown in Figure 16 (by Tamura et al [2]). The method presented by the authors [1] is applied to predict the expansion ratio of mortar bars from the calculated amount of reaction products. In this method, the presence of the absorbing zone in mortar bar is assumed. Then, by comparing the calculated amount of reaction products to the actual expansion ratios, the amount of total absorbing zone (Ter) and the experimental constant E can be obtained from the slope and the intercept of the approximated line shown in Figure 17. Finally, the expansion ratio of mortar bar is predicted by the following equation.

$$\epsilon = E * (Tpr - Ter) \quad (14)$$

Where, ϵ indicates the predicted expansion ratio (%), and E and Ter indicate the experimental constant to calculate expansion ratio from the amount of reaction products which contributes to induce expansion ($\% \text{xl-mortar/mol}$), and the amount of total absorbing zone (mol/l-mortar), respectively.

Figure 18 shows the predicted expansion curves of the mortar bars made with the aggregates showing constant rate expansion. As can be seen from Figures 16 and 18, a good similarity between calculated and observed expansion curves can be seen. In particular, the periods when the calculated expansion ratio suddenly increase almost coincide with the periods

observed in actual expansion curves.

Figures 19 and 20 show the calculated and observed expansion curves of mortar bar made with andesite. It is found that the calculated curve also shows the decreasing rate expansion behavior which is similar to the observed one.

From these comparisons, it is felt that the procedure presented here is useful to predict the expansion curves of both mortar bars showing constant rate expansion and decreasing rate expansion.

9. A NEW SYSTEM TO EVALUATE ALKALI SILICA REACTIVITY OF AGGREGATE

From the calculated and experimentally obtained results in this work, it is confirmed that the amount of reaction products in mortar bars can be predicted by using the result of the modified leaching test. This method can also show the ultimate amount of reaction products which is considered to have close relationship with the potential reactivity of aggregate. Therefore, the proposed method is useful to evaluate the alkali-silica reactivity of aggregate in a more quantitative manner than the conventional method. From now on, the authors refer to this method as "Uomoto-Furusawa-Ohga method", a new system to evaluate alkali-silica reactivity of aggregate. The procedures of this "U.F.O. method" are as follows.

I) The leaching test is performed on the aggregate basically in accordance with the quick chemical test defined as ASTM C 289 or JIS A 5308 at least under two different temperature levels. The results of the leaching test (amount of dissolved silica $Sc(mm\text{ol/l})$ and consumed alkali $Rc(mm\text{ol/l})$) should be detected with the passage of time.

II) By the equation (6), thickness of the reacted layer is calculated.

III) By linear or bilinear relationship which can approximate the relationship between the values x and $(Ct)^{1/2}$, the diffusion coefficient of alkali in the aggregate at each temperature is obtained.

IV) The effect of temperature on the diffusion coefficient can be quantitatively evaluated by Arrhenius's equation, and the diffusion coefficient at 40°C is obtained by this equation.

V) From the relationship between Sc and Rc , the amount of initially consumed alkali and alkali silica ratio of reaction products are obtained.

VI) By using equations (1),(4),(11) and (13), the amount of reaction products in the mortar bar is calculated.

VII) By equation (14), the expansion ratio of mortar bar can be predicted.

VIII) By continuing the calculation until all the alkali is used up, the ultimate amount of reaction products is calculated.

10. CONCLUSIONS

The following topics have been made clear by this work.

- i) The diffusion coefficient of alkali in the aggregate showing constant rate expansion changes during reaction.
- ii) Much amount of alkali is consumed at the beginning stage of reaction without producing any significant amount of reaction products.
- iii) The effect of temperature on the diffusion coefficient can be evaluated quantitatively by Arrhenius's equation.

ACKNOWLEDGEMENTS

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REFERENCES

- [1] T.Uomoto and Y.Furusawa, Concrete Library of JSCE, No.20, pp.177-186, December 1992
- [2] H.Tamura, T.Takahashi and M.Ohashi, Proc. of JCI Sympo. on Test Method for Determination of AAR in Concrete, pp.67-72, 1991, (in Japanese)
- [3] S.Diamond, Proc. of 8th Inter. Conf. on AAR, Kyoto, pp.101-105, 1989

Table 1 Tested Aggregate

No.	Type of Aggr.	Type of Expansion
1	Sandstone	Constant Rate Expansion
2		
3		
4		
5	Quartz Glass	Dec. Rate Exp.
6	Andesite	(Dec. Rate Exp.)

Table 2 Test Conditions

Temperature	60,70,80 degrees
NaOH Conc.	1 mol/l
Aggregate Size	150-300 micrometers
Solid/Liquid Ratio	25g/25ml

Table 3 Obtained Diffusion Coefficients

No.	<i>kini</i> (cm2/hr)	<i>kpost</i> (cm2/hr)	x when diffu. coeff. changes
1	2.158E-12	6.168E-11	7.076E-6
2	6.069E-13	3.017E-11	6.332E-6
3	1.954E-12	4.125E-11	6.737E-6
4	8.115E-13	1.214E-11	6.139E-6
5	1.087E-9		
6	2.463E-10		

Table 4 Calculated Diffusion Coefficients
at 40 Degrees

No.	$k_{ini}(cm^2/hr)$	$k_{post}(cm^2/hr)$
1	3.427E-13	6.152E-12
2	1.233E-14	2.696E-12
3	8.510E-14	9.687E-13
4	7.249E-14	1.921E-12
5	2.840E-11	
6	1.079E-11	

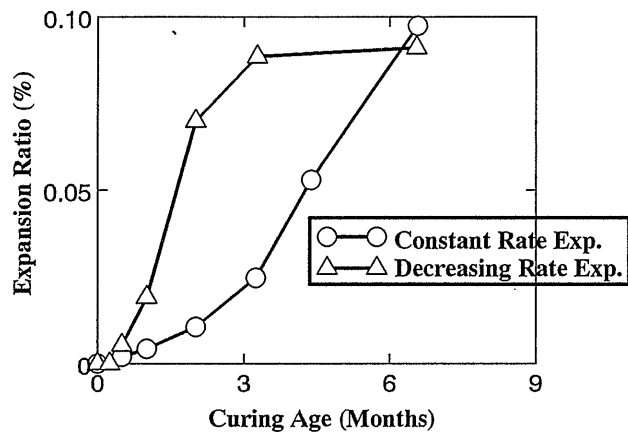


Fig.1 Two Typical Expansion Behaviors

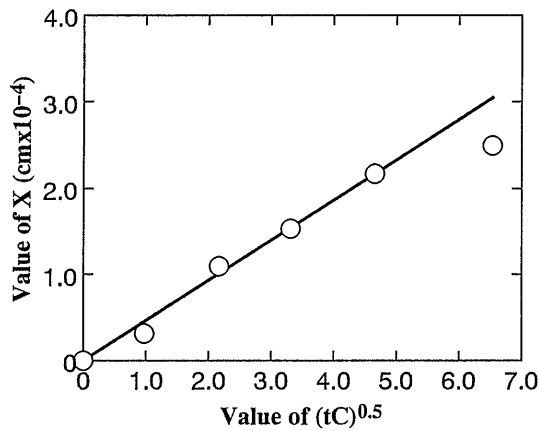


Fig.2 Test Results for Aggregate No.5

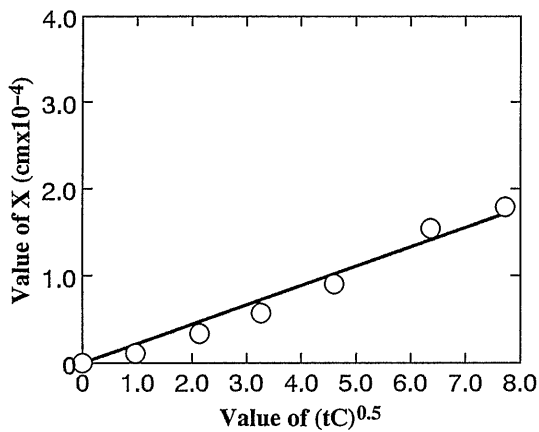


Fig.3 Test Results for Aggregate No.6

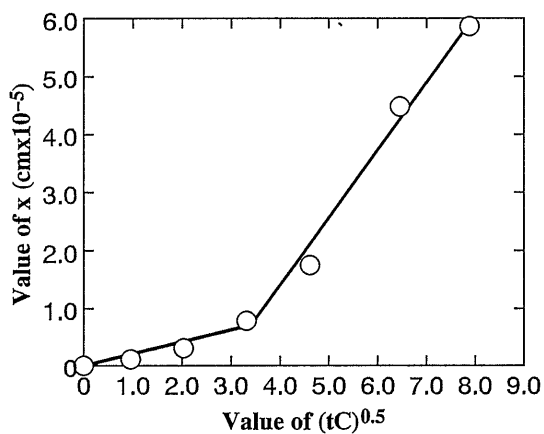


Fig.4 Test Results for Aggregate No.1

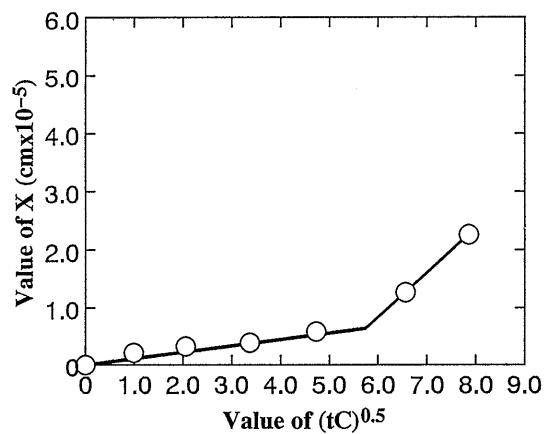


Fig.5 Test Results for Aggregate No.2

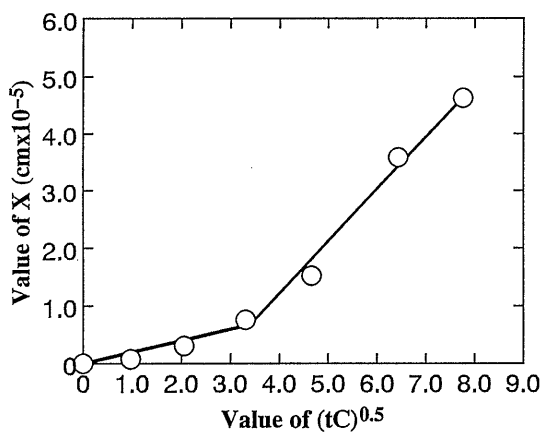


Fig.6 Test Results for Aggregate No.3

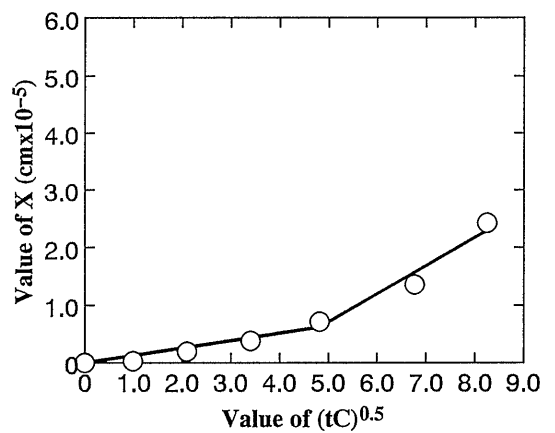


Fig.7 Test Results for Aggregate No.4

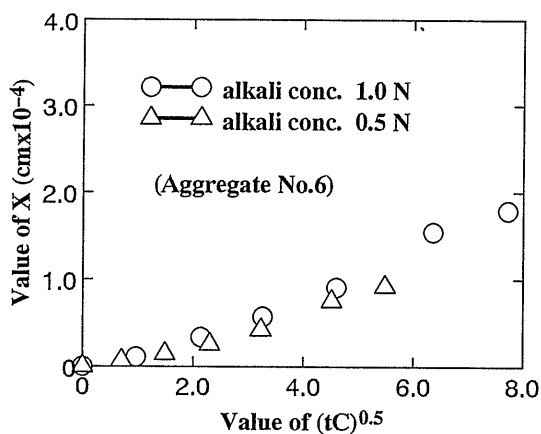


Fig.8 Results with Different Alkali Concentration

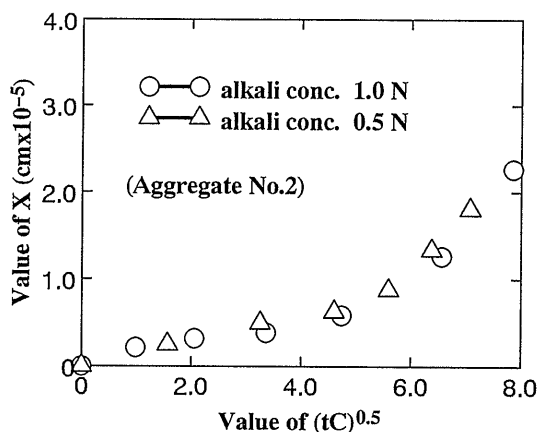


Fig.9 Results with Different Alkali Concentration

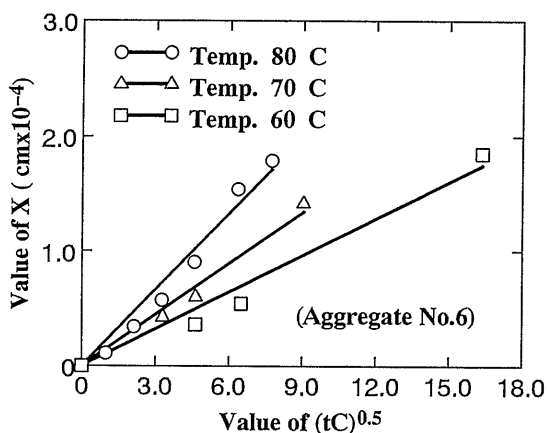


Fig.10 Results under Various Temperatures

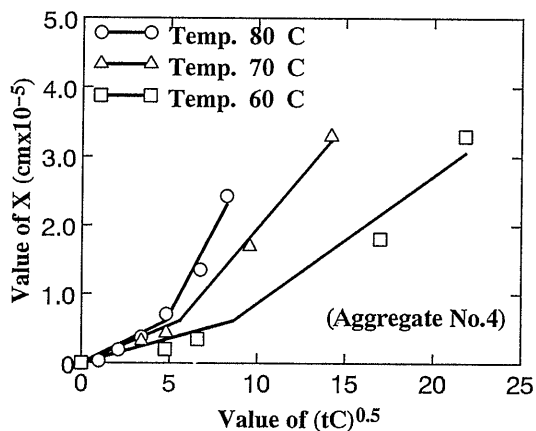


Fig.11 Results under Various Temperatures

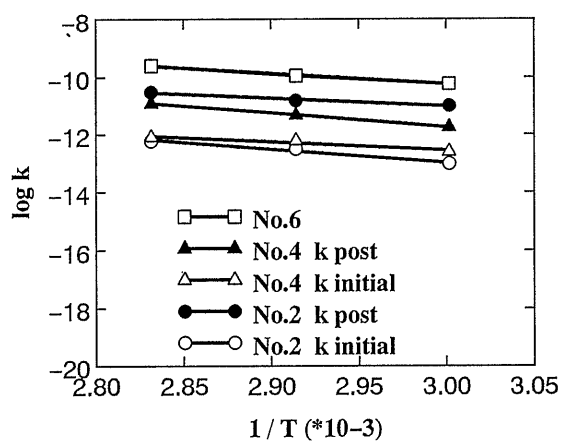


Fig.12 Effect of Temperature

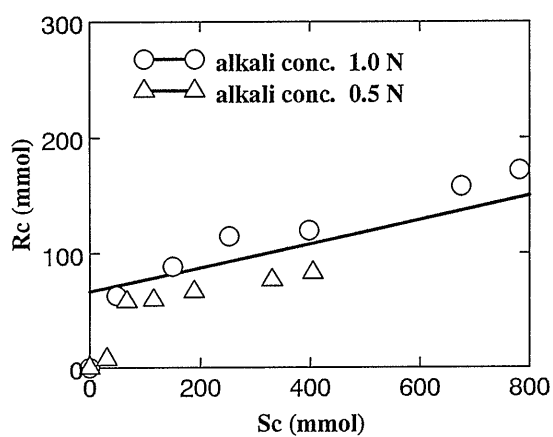


Fig.13 Relationship between Sc and Rc (No.6)

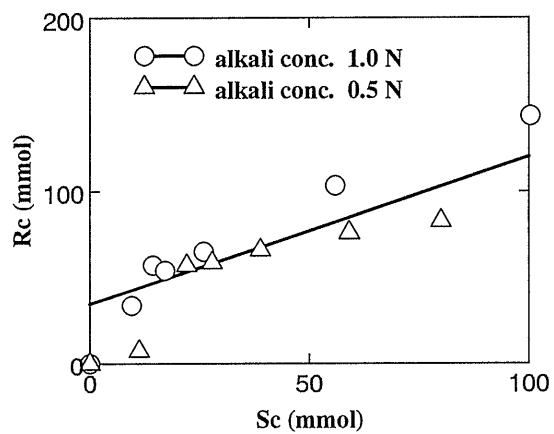


Fig.14 Relationship between Sc and Rc (No.2)

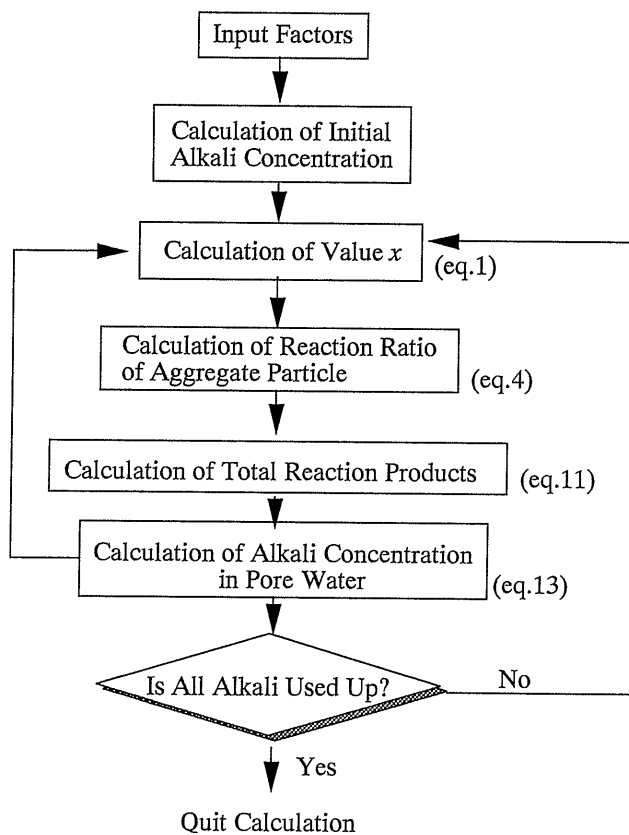


Fig.15 Calculation Procedure

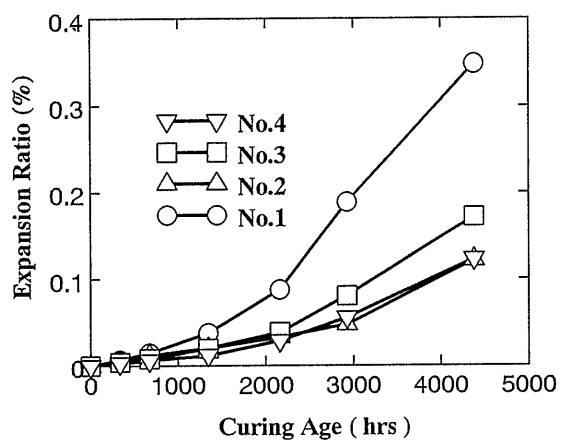


Fig.16 Observed Expansion Curves

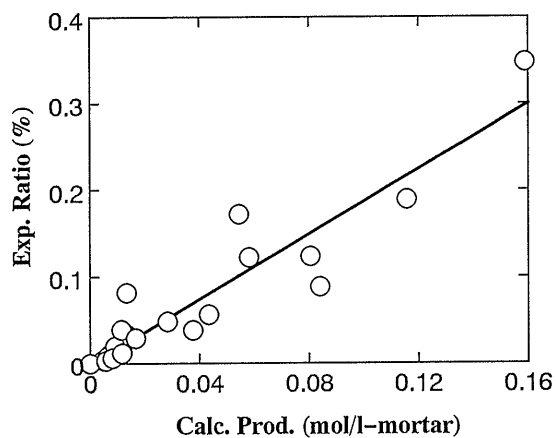


Fig.17 Expansion Ratio and Calculated Products

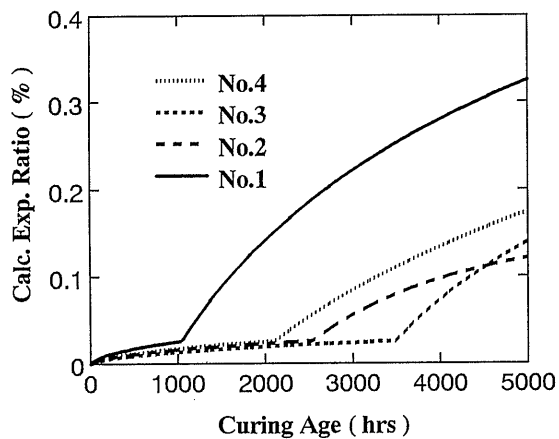


Fig.18 Calculated Expansion Curves

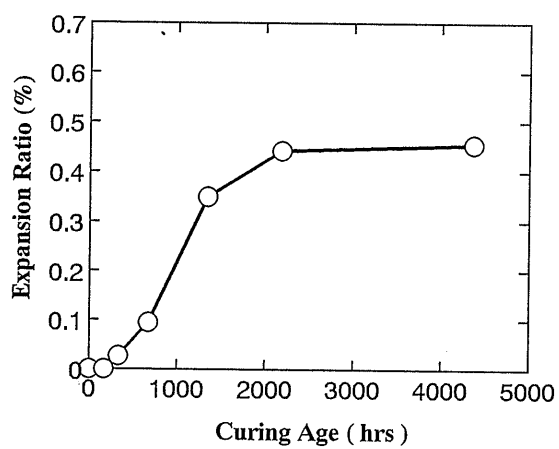


Fig.19 Observed Expansion Curve (No.6)

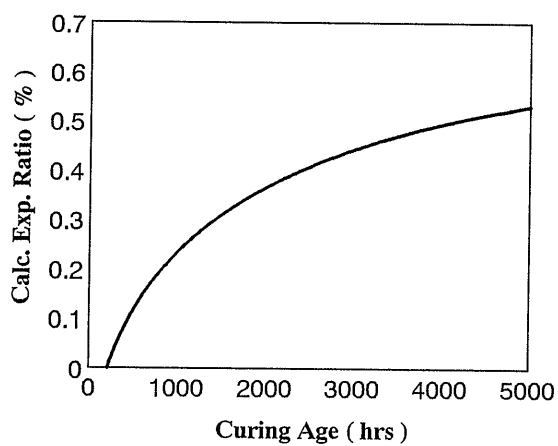


Fig.20 Calculated Expansion Curve (No.6)