SIMULATING CHLORIDE PENETRATION IN FLY ASH CONCRETE BY A FRACTAL DERIVATIVE MODEL

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In the real engineering field, the chloride ions behave abnormal diffusion phenomena in concrete caused by different compositions of the concrete which lead to the complex physical and chemical properties. This paper utilizes a fractal derivative model and a fractional derivative model to describe the diffusion phenomena. Furthermore, according to actual experimental data in the field, the fractional and fractal model can simulate the diffusion behavior of chloride ions in concrete. In comparison to the fractional derivative model, the fractal derivative model gives a simpler mathematical expression and lower calculation costs. In addition, the linear regression analysis method is used to establish an effective relationship between the internal composition of concrete and the parameters of fractal model such as fractal order α and diffusion coefficient D. As a result, the fractal model with the parameters estimated by above relationship can predict the diffusion behavior of chloride ions.

Key Words: fractal derivative, chloride diffusion, fly ash concrete, fractal order, diffusion coefficient

1. Introduction

For coastal roads, bridges, ports and other concrete structures, penetration of chloride ions is considered as a main cause of a variety of durability problems, such as steel corrosion, cracking of the concrete cover and even spalling. In order to solve problems in practice, many scholars have improved the traditional theory of chloride ion transport. In 1970, Collepardi et al. [1] were the first to advocate the use of Fick second diffusion law to characterize the apparent diffusion behavior of chloride ions in concrete, and put forward eight basic assumptions. Nilsson et al. [2] studied the nonlinear rule of chloride ion binding ability. Subsequently, Chalee et al. [3] put forward a chloride diffusion model with a time dependent diffusion coefficient and validated it by their experimental data. Furthermore, from many researches [4–6], it was concluded that the chloride diffusion coefficient is variable and a time-dependent parameter, which is generally considered as an anomalous diffusion phenomenon. Thus, in the studies of researchers [6–13], some anomalous diffusion models with a modified time-dependent diffusion coefficient have been developed to characterize anomalous diffusion processes. It is worth mentioning that Chen et al. [13] put forward a new fractional diffusion model with variable order to characterize the coupled chloride diffusion-binding processes in cement-based concrete, which can effectively present the memory attributes. However, these models ignored establishing further relationships between internal components and model parameters which caused the difficulty in application. In this paper, the linear regression method is used to construct the relationships between the components of concrete and the parameters of fractal model. Furthermore, the fractal model is extended to predict some actual physical engineering.

Recently, a fractal derivative diffusion model was developed as a powerful tool to characterize anomalous diffusion in complex systems [14–21]. As shown in Refs. [15, 16], the definition of fractal derivative was improved and employed to model water diffusion transport in unsaturated porous media. Previous research illustrated that the fractal derivative model is simple and gives a high precision [22]. It was also found that the fractal derivative model was suitable to describe the well-documented experimental data which exhibited stretched Gaussian distribution. In this work, the fractal derivative diffusion model is adopted to describe chloride ingress into concrete based on diffusion as the major transport mechanism.

With the deepening of the research on fly ash concrete, its application is becoming more and more widespread. From the material point of view, high performance concrete together with fly ash as the main mineral admixture has been widely used in coastal engineering, and its high resistance against chloride ion penetration has been proved [23,24]. Cheewaket et al. [25] reported that the application of fly ash can decrease the amount of free chloride ions which were found to affect the service performance of fly ash concrete. Chalee et al. [3] considered that the higher the fly ash content (< 50%), the higher the resistance of concrete to ingress of chloride salt becomes. Furthermore, the study of Yazici [26] shows that the penetration depth of chloride ions in 60% fly ash concrete is reduced by 50%. It has been well known that the water to binder (W/B) ratio is also an important factor affecting chloride diffusion. It was concluded by Chalee and Jaturapitakkul [3] that the reduction of W/B ratio leads to a decrease of the chloride diffusion coefficient and the application of fly ash with higher W/B ratio has shown to be more effective on reducing of chloride diffusion coefficient than that of Portland cement concrete with lower W/B ratio. In this paper an attempt will be made to establish the relationship between the parameters of the fractal diffusion model and fly ash concrete composition to avoid only describing or predicting the chloride ion penetration of existing concrete structures.

This article is structured as below. Section 2 provides introduction to the fractal and fractional derivative diffusion models. In Section 3, numerical results of the comparisons between the fractal and fractional derivative diffusion models are presented and the relationship between the parameters of the fractal diffusion model and fly ash concrete composition by regression analysis is investigated. Section 4 verifies the practicality of the fractal diffusion model. Finally, conclusions are made in Section 5.

2. Chloride diffusion models

2.1. fractal derivative model

Chen in Ref. [14] proposed a definition of the fractal derivative：

$$\begin{cases} \dfrac{du(t)}{dt^{\alpha}} = \lim\limits_{t_1 \to t} \dfrac{u(t_1) - u(t)}{t_1^{\alpha} - t^{\alpha}} \\ \dfrac{du(x)}{dx^{\beta}} = \lim\limits_{x_1 \to x} \dfrac{u(x_1) - u(x)}{x_1^{\beta} - x^{\beta}} \end{cases} \tag{1}$$

where α and β are orders of the fractal derivative in time and space, respectively. It should be noted that there is not convolution integral in the fractal derivative.

Ordinary derivative can be obtained from the fractal derivative based on the scaling transformation [14].

$$\begin{cases} \hat{t} = t^{\alpha} \\ \hat{x} = x^{\beta} \end{cases}. \tag{2}$$

The fractal derivative is an effective mathematic method in characterizing local and complex behavior. The space-time fractal derivative diffusion equation based on the above fractal derivative is written as [14]

$$\begin{cases} \dfrac{\partial u(x,\ t)}{\partial t^{\alpha}} = D_{\alpha,\beta} \dfrac{\partial}{\partial x^{\beta}} \left(\dfrac{\partial u(x,\ t)}{\partial x^{\beta}} \right) t > 0, \quad -\infty < x < +\infty, \\ u(x,0) = \delta(x), \end{cases} \tag{3}$$

where $0 < \alpha < 2$; $0 < \beta <= 1$, $D_{\alpha,\beta}$ is the generalized diffusion coefficient ($m^{2\beta}/s^{\alpha}$), $\delta(x)$ represents the delta function and $u(x,t)$ stands for the chloride concentration when the depth is equal to *x* and the time is equal to *t*.

When $\beta = 1$, the space-time fractal derivative diffusion equation can be reduced to a time-fractal diffusion model, which is a one-dimensional diffusion process and can be expressed as

$$\frac{\partial u(x,\ t)}{\partial t^{\alpha}} = D \frac{\partial}{\partial x} \left(\frac{\partial u(x,t)}{\partial x} \right) t > 0. \tag{4}$$

Based on the finite difference way, the discrete form of the time fractal derivative is written as

$$\frac{\partial u(x_i,\ t_{k+1})}{\partial t^{\alpha}} = \frac{u(x_i,\ t_{k+1}) - u(x_i,\ t_k)}{t_{k+1}^{\alpha} - t_k^{\alpha}}. \tag{5}$$

By applying the implicit format discrete method, the spatial fractal derivative section in Eq. (4) can be rewritten approximately by

$$\frac{\partial^2 u(x_{i+1}, t_{k+1})}{\partial x^2} = \frac{u(x_{i+1},\ t_{k+1}) - 2u(x_i,\ t_{k+1}) + u(x_{i-1},\ t_{k+1})}{h^2}, \tag{6}$$

where $h = L/N_x$, L is space length, N_x is the total space segment, x_i signifies the position of the *i*th point and t_k represents the time node of the *k*th point.

By substituting Eqs. (5) and (6) into Eq. (4), the discrete format of the time fractal diffusion equation can be obtained

$$\frac{u(x_i,\ t_{k+1}) - u(x_i,\ t_k)}{t_{k+1}^{\alpha} - t_k^{\alpha}} = D \frac{u(x_{i+1},\ t_{k+1}) - 2u(x_i,\ t_{k+1}) + u(x_{i-1},\ t_{k+1})}{h^2}, \tag{5a}$$

and hence,

$$\left(-D\frac{1}{h^2}\right) u_{i-1}^{k+1} + \left(\frac{1}{\Delta t_k} + D\frac{2}{h^2}\right) u_i^{k+1} - \left(D\frac{1}{h^2}\right) u_{i+1}^{k+1} = \left(\frac{1}{\Delta t_k}\right) u_i^k, \tag{6a}$$

where $\Delta t_k = t_{k+1}^{\alpha} - t_k^{\alpha}$ and $u(x_i,\ t_k) = u_i^k$. The numerical solution of Eq. (4) can be obtained by solving Eq. (6).

2.2. fractional derivative model

At present, many scientists have proposed a variety of fractional derivative definitions [28] for different problems. These definitions are not only interconnected but also different. There is an advantage in solving practical project in the Caputo definition which contains the integral initial condition. It is expressed as

$$\frac{d^{\gamma} f(t)}{dt^{\gamma}} = \frac{1}{\Gamma(n-\gamma)} \int_0^t \frac{f^{(n)}(\tau)}{(t-\tau)^{\gamma+1-n}} d\tau, \qquad n-1 < \gamma < n, \tag{7}$$

where γ is the fractional derivative order, n is an integral number, and the definition of the function $\Gamma(\cdot)$ is $\Gamma(z) = \int e^{-t} t^{z-1} dt$ given by $Re(z) > 0$.

A fractional derivative model which is applied to study chloride penetration in concrete is given by [29]

$$\frac{\partial u^{\gamma}(x,t)}{\partial t^{\gamma}} = D_{\gamma}\frac{\partial u^{2}(x,t)}{\partial x^{2}}, \tag{8}$$

where the range of γ is from 0 to 1, D_{γ} is a generalized unchanging diffusivity (m^2/s^{γ}) and $u(x,t)$ stands for the chloride concentration when the depth is equal to *x* and the time is equal to *t*.

Nevertheless, finding analytical solutions of Equation (8) is not easy. Hence getting the numerical solution is necessary (reference is made to [30] for more details).

3. Results and discussion

3.1. Review of the experimental program

In 2008, Chalee et al. reported a series of experimental data on different concrete blocks over a period of 2 and 5 years under tidal exposure conditions [3,27]. In these experiments, the geometry of the cast concrete cube samples is 200×200×200 mm^3 whereas the W/ B ratios were different and were set at 0.45, 0.55 and 0.65. Moreover, concrete samples were prepared by employing class fly ash to substitute for Portland cement at 0%, 15%, 25%, 35% and 50% of weight of binder. All the casted samples were taken out from the molds after 24 hours and then were cured for 27 days in water. Thereupon they were carried to a tidal area and were exposed to wet-dry circles of seawater with temperatures ranging between 25℃and 35℃. Then the entire chloride content was determined according to ASTM C1152 [31].

3.2. Comparison of diffusion models

The validity of the present fractal derivative diffusion model was investigated by using Eqs. (4) and (8) to fit experimental data obtained from specimens in the marine environment [3,27]. By comparing experimental data with simulation results of the corresponding fractional diffusion model, the merits and distinct characteristics of the fractal derivative diffusion model are evaluated.

A series of experimental data and the corresponding simulation results in the fractal and fractional diffusion models are shown in Fig. 1. Based on these two diffusion models, the residual sum of squares (RSS) and the corresponding parameters are obtained (see Tables 1, 2 and 3) through the least square fitting of measurement data.

As shown in Fig. 1, the concentration of chloride ion in the test blocks decreases with the increase of depth and the diffusion tendency of both the fractal and fractional diffusion models is almost consistent when the depth is not so large. In this circumstance, these two models reveal almost the same diffusion rate. In addition, there is a key point between two models for the concrete blocks containing fly ash. Before the key point, the two models reflect almost the same rate of chloride diffusion. However, the rate of chloride diffusion in the fractal derivative model is slower than that of the fractional diffusion model over the key point in the terms for the fly ash concrete. Therefore, the fractal diffusion model which is used to describe chloride ion diffusion at a greater depth should be more reasonable compared to the fractional diffusion model.

Thus, it can be concluded from Table 1 that the trend of order α in the specified W/B with the change of a volume of fly ash replacement (FAR) is not obvious. Tables 2 and 3 show that every RSS of the fractal model is smaller than the corresponding one of the fractional model with the same number of parameters, especially when the FAR is really large, which demonstrates that the fractal diffusion model enjoys a particular advantage of higher accuracy. Moreover, from Table 4, it can be found that the fractal diffusion model requires less calculation time compared to the fractional diffusion model, especially when the number of nodes is really large. Therefore, it is testified that the proposed fractal model is a more dependable and effective model to describe chloride ions diffusion and requires less calculation time.

3.3. Parameters of the fractal diffusion model

As mentioned above, the parameters *D* and α in the fractal diffusion model can be obtained by using the fitting method of experimental data. However, such a fitting method requires a significant amount of experimental data of concrete during a very long exposed period, which means that the fractal diffusion model can only be applied to describe or forecast the penetration of chloride ions for existing concrete structures. The durability of concrete structures also cannot be predicted based on concrete composition and environmental factors, which cannot provide the reference and guidance for durability design of new concrete structures. In order to overcome this shortcoming, it is necessary to establish a relationship between the parameters of the fractal diffusion model and the parameters characterizing fly ash concrete composition or environmental factors. In the following section, an attempt will be made to achieve this.

In the literature [3, 27], a large number of comparative experiments were conducted to explore the effect of two relevant factors, i.e. the W/B ratio and the FAR, on the chloride ions diffusion in fly ash concrete. Information on the specific raw material selection, concrete mix ratio, experimental test method and results can be obtained from the original literature [3, 27]. Next, the experimental results obtained in the literature are used to establish the relationship between the two material factors and the parameters of the fractal diffusion model.

Using the least squares fitting calculation procedure, the parameters *D* and fractal order α can be obtained from the simulation of the fractal diffusion model for chloride ion penetration in different concrete test blocks. The fitting results providing *D* and α are shown in Table 3. From the RSS it is observed that the correlation of the relationship of the parameters can be evaluated.

3.3.1 The fractal derivative order α

Through observation and analysis, it is found that the order α does not change much with the increase of the FAR for the specified W/B ratio, but fluctuates around an intermediate value. Hence, the average value of α is used in this section as the fractal derivative order under the specified W/B ratio (see Table 5). As regression equation is used to fit the mean value of α by a linear equation. The results are revealed in Figure 2.

$$\alpha = 0.4560\mathrm{W/B} + 0.6484 \tag{9}$$

As can be seen from Figure 2, the fractal order tends to increase linearly with the addition of W/B ratio in fly ash concrete. In order to better reflect the influence of W/B ratio and FAR on chloride diffusion in concrete and reduce the divergence, here we first determine the fractal order α according to the fitting equation (9). Then the diffusion coefficient D (see Table 6) was obtained by refitting the experimental data according to the least square method. Through the analysis of the RSS, it is noted that the order of formula calculation does not affect the fitting effect.

3.3.2 Chloride diffusion coefficient D

Assuming that two factors W/B and the FAR influencing D are independent of each other, by using the regression analysis method, the optimal regression equation of the corresponding chloride diffusion coefficient D can be obtained

$$\mathrm{D} = 42.6156 \times (\mathrm{W/B})^{0.7455} F^{-0.3737}. \tag{10}$$

As shown in Figure 3, the diffusion coefficient in fly ash concrete is positively correlated to the W/B ratio, and is negatively correlated to the FAR. That means that the D increases with an increase of the W/B ratio, and decreases with an increase of the FAR. These variations match reasonably with the real behavior of chloride diffusion because concrete including fly ash will demonstrate good chloride resistance and the pore size of it is smaller than concrete without fly ash [32-34].

So far, an empirical regression model of the parameters, which has two influencing factors: W/B ratio and FAR, is established. After the model is determined, it is more convenient for us to predict penetration of chloride ions in fly ash concrete under similar conditions. This provides a useful reference for the durability design of concrete structures exposed to a chloride-laden environment and the evaluation and maintenance of existing concrete structures.

4. Practical applications

According to the corresponding formula mentioned above, it is easy to predict chloride ion penetration in concretes having different W/B ratio and FAR under similar conditions. In the literature [6, 35], the distribution of chloride ion after a period of exposure with other W/B ratios and FARs can be used to verify our model. The model parameters of the fractal diffusion model obtained by the fitting formula are shown in Table 7. The parameters of the fractal diffusion model obtained by a direct fitting calculation are indicated in Table 8.

Comparing the results of Tables 7 and 8, it is seen that the RSS between the results obtained from the fitting formula of model parameters and the experimental data is slightly larger than that obtained from the direct fitting results, which is considered acceptable. In addition, the results shown in Figs. 4 and 5 also confirm that the predicted values by the fitting formula of model parameters correlate reasonably well with test data for chloride penetration. In other words, what is shown above indicates that the relationship between the model parameters and the parameters of concrete materials is reasonable and effective.

5. Conclusions

The fractal derivative model is suitable to describe chloride penetration in fly ash concrete. The simulation results show that the fractal derivative model could describe the abnormal diffusive process with lower calculation costs and higher accuracy compared with the fractional derivative model. It is worth noting that the values of fitting parameters α and D of the fractal and fractional diffusion models are very close. According to the regression analysis of existing experimental data, the quantitative relationship between the parameters of fractal diffusion model and the material factors (such as W/B ratio and the FAR) is obtained and verified. The results obtained clearly indicate that the fractal order α increases linearly with the increase of W/B ratio in fly ash concrete while the chloride diffusion coefficient D is positively correlated to the W/B ratio and is negatively correlated with the FAR. Furthermore, it is believed that the fractal diffusion model can be extended to predict the diffusion process of chloride ions in other untested fly ash concrete through the analysis of relationship between model parameters and specific components of the concrete.

In addition, the durability of concrete structures under loading conditions and the spatial correlation modeling of chloride ion transport in concrete needs further research.

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Table 1. Model parameters determined at 2-year exposure in fractal and fractional diffusion models, respectively

Cement Type	Fractal Model		Fractional Model	
	α	$D(\times 10^{-12} m^2/s^{\alpha})$	α	$D(\times 10^{-12} m^2/s^{\alpha})$
I45	0.8189	26.6976	0.7988	21.2948
I45FA15	0.8799	8.6550	0.8589	9.3809
I45FA25	0.8636	7.5892	0.8415	8.4163
I45FA35	0.8515	6.4956	0.8316	7.6205
I45FA50	0.8598	3.3656	0.8378	3.7595

I45, FA15, FA25, FA35, FA50: W/B ratio 0.45, FAR 15%, FAR 25%, FAR 35%, FAR 50%, respectively.

Table 2. Mean square errors of two different models at 2-year exposure

Cement Type	Fractal Model	Fractional Model
I45	0.0985	0.1256
I45FA15	0.0330	0.0381
I45FA25	0.0301	0.0356
I45FA35	0.0024	0.0033
I45FA45	0.0015	0.0051

Table 3. Fitting parameters of fractal diffusion model at 5-year exposure

Cement Type	α	$D(\times 10^{-12} m^2/s^{\alpha})$	RSS
I45	0.8191	26.7689	0.0985
I45FA15	0.8815	8.6222	0.0330
I45FA25	0.8615	7.5702	0.0301
I45FA35	0.8511	6.4679	0.0024
I45FA45	0.8613	3.3958	0.0525
I55	0.8513	33.5319	0.2385
I55FA25	0.9153	9.5978	0.0974
I55FA50	0.9310	7.3451	0.3643
I65	0.8790	39.6788	0.3482
I65FA15	0.9980	9.9603	0.4165
I65FA25	0.9987	8.7899	0.0400
I65FA35	0.8990	8.2845	0.0252
I65FA50	0.9560	5.6082	0.1227

I45, I55, I65: W/B ratio 0.45, W/B ratio 0.55, W/B ratio 0.65, respectively.

Table 4. The computational time for the fractal and fractional diffusion models under the condition of D=1 and C_0=0.5 (Intel Core i7, RAM 8 GB, 64 bit windows 10 and MATLAB 2016b).

model	order α	Time (s)			
Number of nodes		10	100	200	500
Fractal	1	0.004162	0.099568	0.491546	9.094458
Fractional		0.006367	0.411421	1.166816	15.385838
Fractal	0.9	0.004371	0.131031	0.508139	10.571811
Fractional		0.007920	0.507326	2.812026	39.928678

Table 5. The change of order α of the fractal diffusion model with W/B ratio

W/B	0.45	0.55	0.65
α	0.8549	0.8992	0.9461

Table 6. Fitting parameters of fractal diffusion model

Cement Type	α	D($\times 10^{-12}$m^2/s$^\alpha$)	RSS
I45	0.8549	23.3564	0.0985
I45FA15		9.5360	0.0330
I45FA25		7.7622	0.0301
I45FA35		6.3753	0.0024
I45FA45		3.4766	0.0525
I55	0.8992	27.1373	0.2385
I55FA25		8.2905	0.0974
I55FA50		6.3995	0.3643
I65	0.9461	30.8374	0.3482
I65FA15		12.0866	0.4166
I65FA25		10.6945	0.0400
I65FA35		6.9421	0.0252
I65FA50		5.8197	0.1227

Table 7. The model parameters of fractal diffusion model obtained by fitting formula

Cement Type	α	D($\times 10^{-12}$m^2/s$^\alpha$)	RSS
I38FA20	0.8217	6.7539	0.1701
I52FA30	0.8855	7.3363	0.1577

I38, I52, FA20, FA30: W/B ratio 0.38, W/B ratio 0.52, FAR 20%, FAR 30%, respectively.

Table 8. The model parameters of fractal diffusion model obtained by direct fitting calculation formula

Cement Type	α	D($\times 10^{-12}$m^2/s$^\alpha$)	RSS
I38FA20	0.8198	6.7316	0.1677
I52FA30	0.8913	7.3506	0.1209

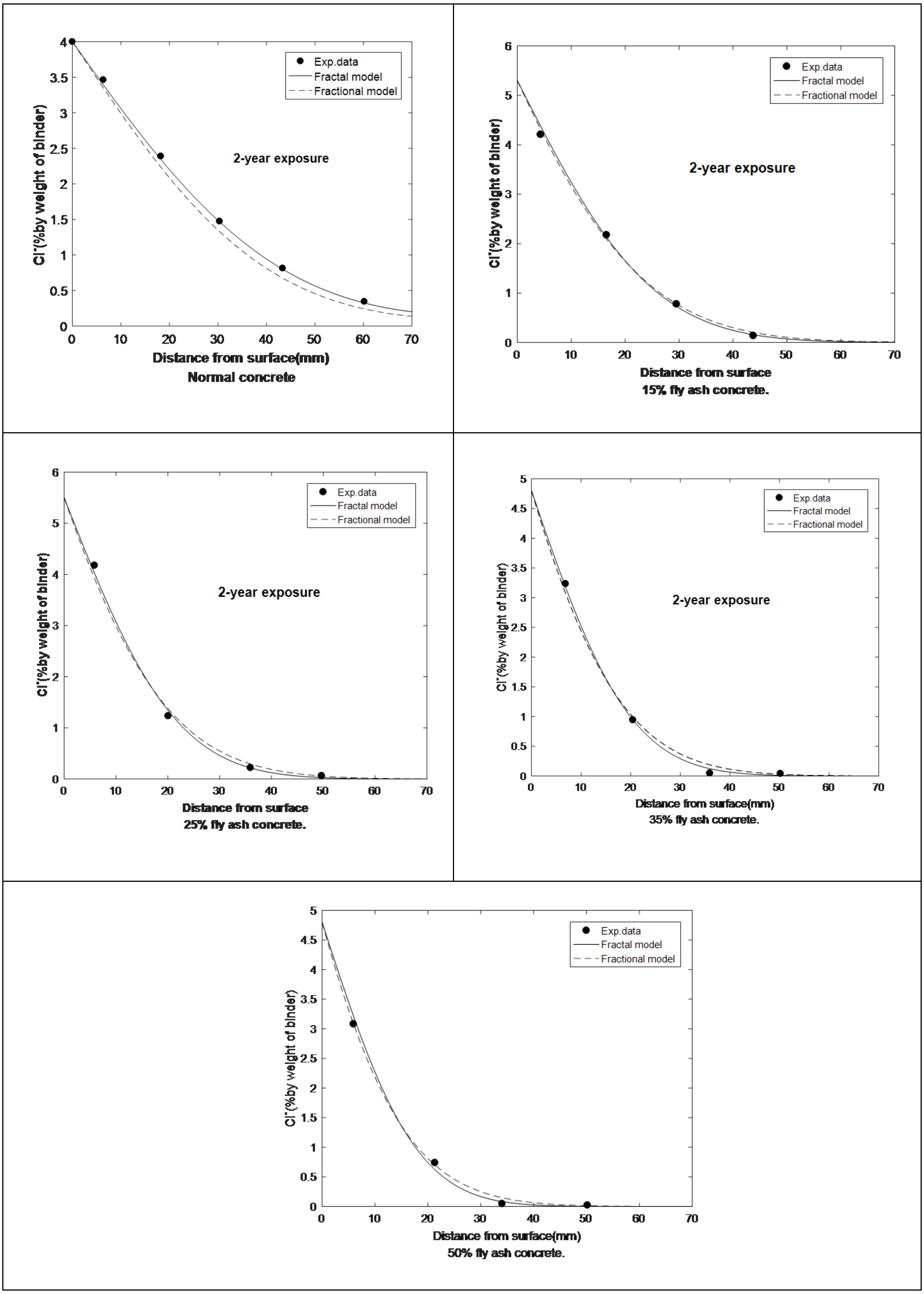

Fig. 1. Chloride profiles based on the experimental data of Chalee et al. [3], the prediction by fractal model and the fractional model for concrete with W/B ratio of 0.65 at 2-year exposure.

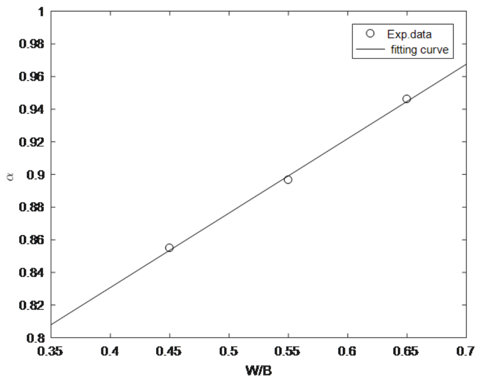

Fig. 2. Change diagram of the fractal order α with W/B ratio.

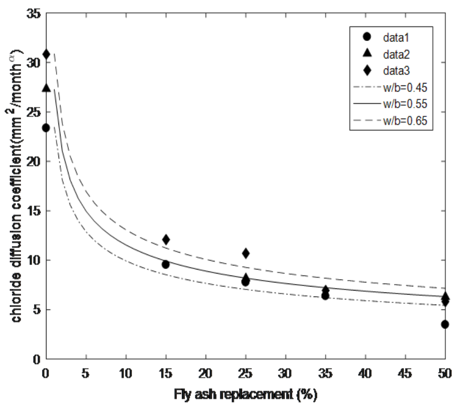

Fig. 3. The relationship between chloride diffusion coefficient and the FAR at different W/B ratios.

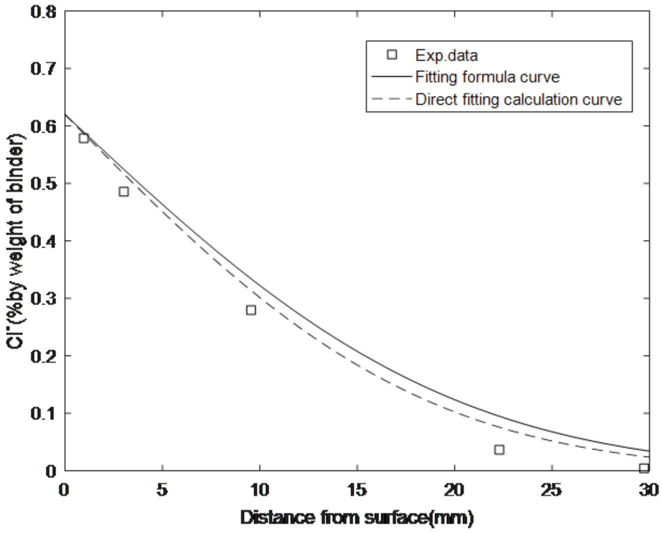

Fig. 4. Chloride profiles based on the experimental data of Oh et al. [35], the prediction by fractal model with parameters obtained by fitting formula and the prediction by fractal model with parameters obtained by direct fitting calculation formula at 10-year exposure.

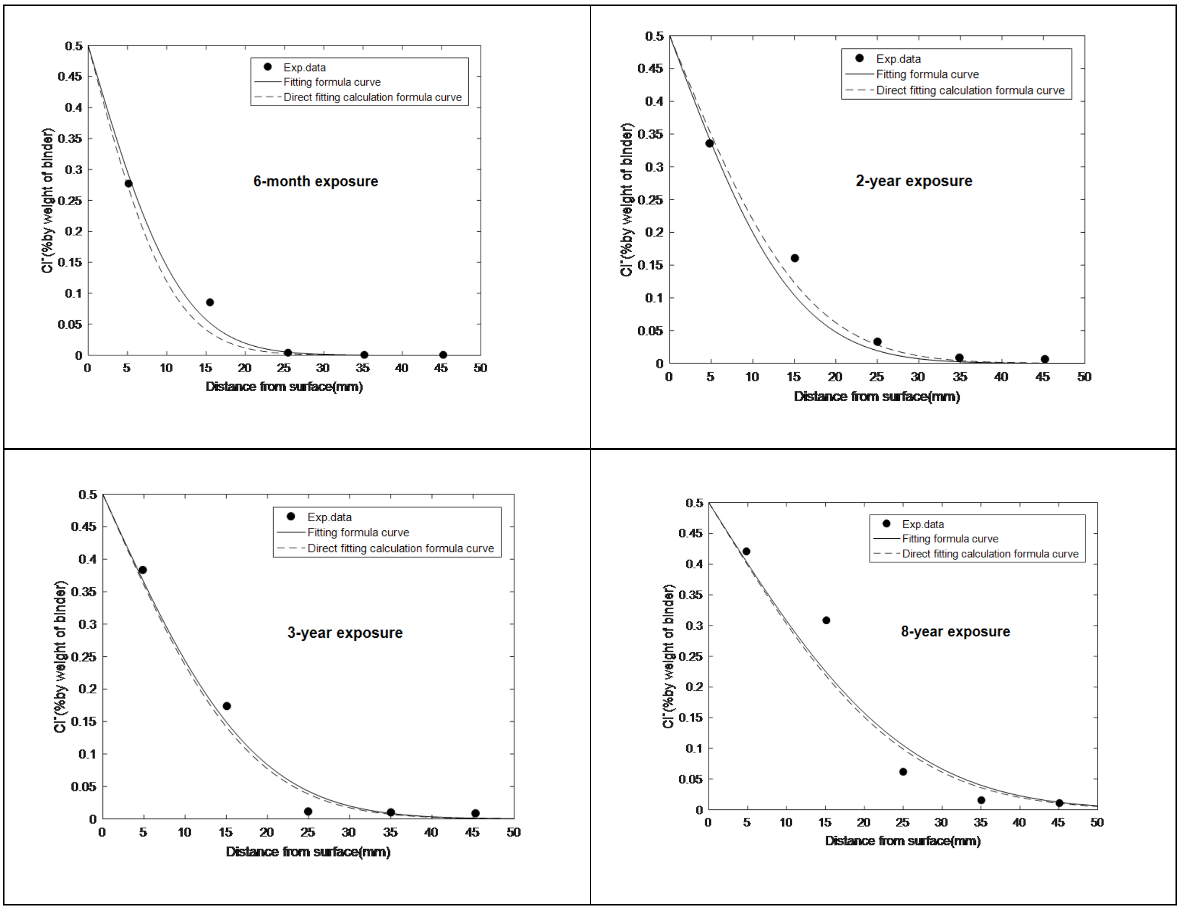

Fig. 5. Chloride profiles based on the experimental data of Thomas et al. [6], the prediction by fractal model with parameters obtained by fitting formula and the prediction by fractal model with parameters obtained by direct fitting calculation formula.