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Developing a model for chloride transport through concrete considering the key factors

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ABSTRACT

The corrosion of chloride-induced on concrete is affected by many factors. In this research, an innovative empirical model was developed to predict the long-term chloride migration behavior in concrete, considering the effects of water-cement ratio, time, bonding effect, temperature, relative humidity, and concrete deterioration. The reliability and validity of the results evaluated by the empirical prediction model were verified by the chloride concentration data in concrete specimens exposed to the marine environment for 3, 5, and 10 years reported in the literature. Combined with the established empirical prediction model, the concrete model was regarded as a three-phase composite material composed of mortar, coarse aggregate, and interfacial transition zone. The effects of key factors on chloride migration were further analyzed by using the mesoscopic finite element numerical simulation method. It was observed that when temperature increases from 5 °C to 65 °C, chloride diffusion depth rises by 3.3 times. When relative humidity increases from 20% to 100%, chloride diffusion depth rises by 4.3 times. Also, the water-cement ratio, concrete deterioration, and chloride binding effect have a non-negligible impact on chloride migration.

1. Introduction

The steel bar corrosion caused by chloride plays a crucial role in reducing concrete durability [1–4]. In the marine and de-icing salt environment, chloride aggression causes non-uniform corrosion of rebar in concrete, which leads to cracking of concrete structures [5, 6]. The chloride permeability directly impacts the durability of concrete structures [7,8].

The chloride migration is impacted by many factors such as time, binding effect and water-cement ratio temperature, etc. Considering these factors, various chloride migration models have been proposed. A modified formula considering the variation of chloride diffusion coefficient with time in concrete was proposed [9]. Weyers et al. [10] revised the chloride ion diffusion model by

Abbreviations: C , chloride content; C_0 , initial chloride concentration; C_b , bounding chloride content; C_f , free chloride content; C_s , surface chloride concentration; C_t , total chloride content; D , chloride diffusivity; D_{28} , chloride ion diffusivity at a curing age of 28 days; f_K , effect parameter of concrete degradation on chloride diffusion; f_t , effect parameter of time on chloride migration; G , ideal gas constant; m , time decay factor; R , relation parameter between free and bound chloride; RH , relative humidity; RH_c , critical relative humidity; t , diffusion time; t_0 , reference time; U , activation energy in diffusion process; w/c , water-cement ratio; w_e , relation parameter between the volume of evaporable pore water and concrete volume; x , depth from the concrete surface; α , chloride ion linear binding coefficient.

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comprehensively considering the impact of erosion time on chloride migration. It was found that chloride diffusivity is significantly affected by temperature change [11]. Considering the multi-factor coupling conditions of concrete structure micro-defects, bonding effect, and chloride diffusivity varying with time, the chloride transport model of concrete was established [12]. Lu et al. [13] demonstrated the regression formula of crack on chloride diffusivity. Many studies have shown that the admixing of supplementary cementitious materials can significantly improve the working performance of concrete [14,15]. A chloride migration model in concrete mixed with fly ash was developed, considering the time dependence of diffusivity and chloride of external [16]. A model considering the impacts of stress level and erosion time on chloride migration under continuous compressive loading was developed [17]. Wang et al. [18] exploited the effect of material heterogeneity on chloride migration in concrete exposed to ocean tide environments for a long time.

The chloride diffusion model in concrete, which reflects the influence of multiple factors, has been extensively studied in the past. However, in the actual environment, chloride migration is affected by many environmental factors, such as relative temperature, humidity, exposure time. Therefore, a reasonable and accurate chloride transmission model considering key factors is significant to the lifetime prediction of concrete.

The present study aims to establish a chloride migration model considering the careful consideration of key factors such as erosion time, binding effect, temperature, relative humidity, and degradation. The analytical solution of chloride migration under the influence of key factors was given and validated by the long-term exposure experiment data of marine concrete. The critical factors affecting chloride migration in concrete were examined parametrically in a quantitative manner.

2. Chloride ion transmission model

The one-dimensional migration of chloride ions in saturated concrete in any direction can be expressed by Fick's second law [19]:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} \quad (1)$$

where C is the chloride content at depth x and time t (%); t is the diffusion time (s); D is the chloride diffusivity (m^2/s); x is the depth from the concrete surface (m).

The initial condition and boundary conditions are demonstrated as:

$$C(x, 0) = C_0 \quad (2)$$

$$C(0, t) = C_s \quad (3)$$

$$C(\infty, t) = C_0 \quad (4)$$

The analytical solution of chloride migration is presented [19]:

$$C(x, t) = C_0 + (C_s - C_0) \left(1 - \operatorname{erf} \frac{x}{2\sqrt{Dt}} \right) \quad (5)$$

where C_0 is the initial chloride ion concentration (%); C_s is the surface chloride concentration (%).

3. Influencing factors of chloride ion transmission

3.1. Water-cement ratio

A higher water-cement ratio will increase the porosity, which directly leads to the acceleration of chloride migration rate in concrete. The porosity and chloride diffusivity of concrete enhance with the enlarge of the water-cement ratio. Thomas and Bentz [20] derived the comparison expression between chloride diffusivity and the water-cement ratio of ordinary concrete at a curing age of 28 days.

$$D_{28} = 10^{-12.06+2.4w/c} \quad (6)$$

Where D_{28} is the chloride ion diffusivity (m^2/s); w/c is the water-cement ratio.

3.2. Exposure time

Chloride ion diffusion rate in cementitious material is related to cement hydration and time, as pore space in concrete is occupied by cement hydration products, which can break the chloride ion transport channels and affects the chloride diffusion property in concrete. Thomas and Bamforth [21] established the relationship between chloride diffusivity with time.

$$f_t = \left(\frac{t_0}{t} \right)^m \quad (7)$$

Table 1
Time decay factor m [22].

Marine environment	Type			
	Ordinary Portland cement	Fly ash	Slag	Silica fume
Underwater area	0.3	0.69	0.71	0.62
Splash area	0.37	0.93	0.60	0.39
Atmospheric area	0.65	0.66	0.85	0.79

Table 2
Molecular activation energy U [28].

OPC	$w/c = 0.4$	$w/c = 0.5$	$w/c = 0.6$
$U/(\text{kJ/mol})$	41.8 ± 4.0	44.6 ± 4.3	32.0 ± 2.4

Where f_t is the effect parameter of time on chloride migration; t_0 is the reference time, $t_0 = 28$ d; m is the time decay factor, and its values are shown in Table 1 [22].

3.3. Chloride ion binding effect

In concrete, there are two chloride forms free chloride and binding chloride. Binding chloride is produced by the physical binding and chemical binding of cement hydration. Sergi et al. [23] suggested that the volume of evaporable water affects the binding chloride content, and the total chloride content was shown as:

$$C_t = w_e C_f + C_b \quad (8)$$

where C_t is the total chloride content (%); C_f is the free chloride content (%); C_b is the content of bounding chloride (%); w_e is the ratio of the volume of evaporable pore water to the concrete volume, generally taking 8% [23].

Substituting Eq. (8) into Eq. (1) gives [23]:

$$\frac{\partial C_t}{\partial t} = \frac{\partial}{\partial x} \left(D w_e \frac{\partial C_f}{\partial x} \right) \quad (9)$$

$$D_{C_f} = \frac{D}{1 + \frac{\partial C_b}{w_e \partial C_f}} = \frac{D}{1 + R} \quad (10)$$

where R is the chloride ion binding coefficient.

At present, scholars have proposed the following chloride ion binding mechanisms: linear binding, Langmuir binding, and Freundlich isotherm binding [23–25]. To simplify the numerical calculation, a linear combination mechanism was used in this paper to carry out the numerical simulation.

$$C_b = \alpha C_f \quad (11)$$

$$f_{C_f} = \frac{1}{1 + \frac{\alpha}{w_e}} = \frac{1}{1 + R} \quad (12)$$

Where α is the chloride ion linear binding coefficient. Yu et al. [12] suggested that, for ordinary concrete, R equals 2–4. For high-performance concrete, R is 3–15.

3.4. Temperature

Temperature significantly affects chloride ion diffusion. First, the increasing temperature can accelerate the chloride diffusivity and reduce the chloride binding capacity. The increase of free chloride concentration will improve the chloride migration performance [26]. Second, high temperatures can accelerate cement hydration and reduce the concrete porosity. The concrete structure becomes denser, and the chloride ion permeability decreases. However, the increase in temperature leads to enhanced chloride migration capacity in the long run. Boddy et al. [27] presented a coefficient under the influence of temperature on chloride migration.

$$f_T = \exp \left[\frac{U}{G} \left(\frac{1}{T_0} - \frac{1}{T} \right) \right] \quad (13)$$

Where f_T is the effective temperature coefficient on chloride migration; T is the ambient temperature ($^{\circ}\text{C}$); T_0 is the initial temperature ($^{\circ}\text{C}$). Generally, the standard curing temperature is 293.15k (20 $^{\circ}\text{C}$). G the is the ideal gas constant, 8.314 J/(mol K^{-1}); U is the

Table 3
Mix proportions of concrete.

No.	W/C	Water (kg/m ³)	Cement (kg/m ³)	Fine aggregate (kg/m ³)	Coarse aggregate (kg/m ³)
A0	0.55	290	526	515	1020
B0	0.65	300	461	525	1105

activation energy in the diffusion process, and the values are shown in Table 2 [28].

3.5. Relative humidity in concrete

Chloride ion diffusion can only occur in a continuous water channel, and the number of water channels in concrete is affected by the relative humidity. Therefore, the chloride diffusivity reduces as relative humidity debases, and a critical value of relative humidity exists [29]. Bazant and Najjar [30] proposed the effect coefficient (f_{RH}) of relative humidity on chloride migration.

$$f_{RH} = \left[1 + \left(\frac{1 - RH}{1 - RH_c} \right)^4 \right]^{-1} \quad (14)$$

Where RH is the relative humidity (%); RH_c is the critical relative humidity (%), generally taking 75%.

3.6. Concrete deterioration

During service, the concrete structure will be deteriorated, thus accelerating the chloride diffusion. Wei et al. [31] summarized the influence coefficient of concrete degradation on chloride diffusion.

$$f_K = \begin{cases} \frac{1}{3} \left[1000(w/c)^2 - 1050(w/c) + 287 \right] & w/c \leq 0.5 \\ 4 & w/c > 0.5 \end{cases} \quad (15)$$

Where f_K is the influence coefficient of concrete degradation on chloride ion diffusion.

3.7. Chloride migration model under key factors

In summary, the chloride diffusivity under the effect of water-cement ratio, time, binding effect, temperature, relative humidity, and concrete deterioration is expressed as:

$$D = \frac{D_{28}}{1 + \frac{w}{w_c}} \left(\frac{t_0}{t} \right)^m \exp \left[\frac{U}{R} \left(\frac{1}{T_0} - \frac{1}{T} \right) \right] \left[1 + \left(\frac{1 - RH}{1 - RH_c} \right)^4 \right]^{-1} f_K \quad (16)$$

By introducing Eqs. (2)–(4) and Eq. (16) into Eq. (1), the chloride migration model under the influence of key factors can be obtained:

$$C(x, t) = C_0 + (C_s - C_0) \left[1 - \operatorname{erf} \left(\frac{x}{\sqrt{\frac{D_0 t_0^m}{(1-m)(1+R)} f_K f_H f_T t^{1-m}}} \right) \right] \quad (17)$$

4. Experiment and analysis

4.1. Experimental program

To verify the model of Eq. (17), this model was compared with the long-term natural exposure test of marine concrete [32]. The test site was located in Zhanjiang harbor, Guangdong, China. The annual average temperature of the test station was 23.3 °C, and the annual average relative humidity was 83.6%. The specimen used P-O 52.5 Ordinary Portland cement. Natural river sand with a fineness modulus of 2.51 and an apparent specific gravity of 2650 kg/m³ was utilized as fine aggregate. The Humen crushed gravel with an apparent specific gravity of 2640 kg/m³ was utilized as coarse aggregate. The mix proportions of concrete details are summarized in Table 3. The exposure ages of the specimen in the underwater area were 3, 5, and 10 years, respectively. The exposed specimens were drilled with coring powder to measure chloride content at different depths.

4.2. Model establishment and parameter selection

According to the test conditions, the dilute material transfer module in the COMSOL Multiphysics 5.3a was selected for a solution. A

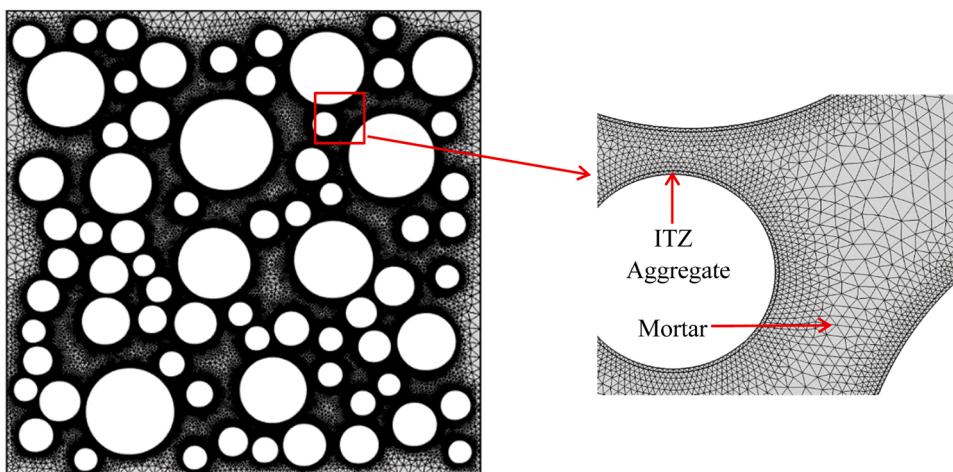


Fig. 1. Finite-element mesh of the two-dimensional concrete model.

Table 4
Model parameters and values.

Parameter	Value	Meaning
t_0	28d	Reference time
m	0.3	Time decay factor
f_K	4	Degradation coefficient
α	0.43	Chloride ion linear binding coefficient
RH	95%	Relative humidity in concrete
C_0	0	Initial chloride ion concentration
RH_c	75%	Critical relative humidity
U	41,650 J/mol	Diffusion activation energy
R	8.314	Ideal gas constant
D_{28}	$1.82 \times 10^{-11} \text{ m}^2/\text{s}$	Chloride ion diffusion coefficient for curing 28 d

Table 5
Surface chloride concentration C_s [32].

Water-cement ratio	Surface chloride concentration/%		
	$t = 3a$	$t = 5a$	$t = 10a$
0.55	0.3872	0.3891	0.4708
0.65	0.3815	0.4620	0.5731

two-dimensional concrete model of $100 \times 100 \text{ mm}$ was established. The finite-element mesh used in the simulation model of concrete with an aggregate volume fraction of 45% is displayed in Fig. 1. Table 4 lists the critical parameters adopted by the model. The left edge of the model was assumed to be the erosion surface, and the other three sides were supposed to be impermeable. The values of surface chloride content under various conditions are shown in Table 5 [32]. Transient analysis was utilized to solve the model. The solution time of the set model was consistent with the test time, and the solution step size was one day.

4.3. Experiment results and analysis

Comparison between predict outcomes and test data is presented in Fig. 2. The chloride content calculated by the chloride migration model matches the experimental data well. In general, the numerical method and the proposed chloride diffusion model are reasonable in this paper.

5. Parameter analysis

Using Eqs. (16) and (17) of the chloride migration model considering the influence of key factors proposed in this paper, the impact of temperature, relative humidity, water-cement ratio, concrete deterioration and chloride binding effect on the transmission of chloride in concrete can be analyzed from a mesoscopic perspective.

From the above discussion, a two-dimensional random aggregate model of $100 \times 100 \text{ mm}$ was established. The aggregate particle

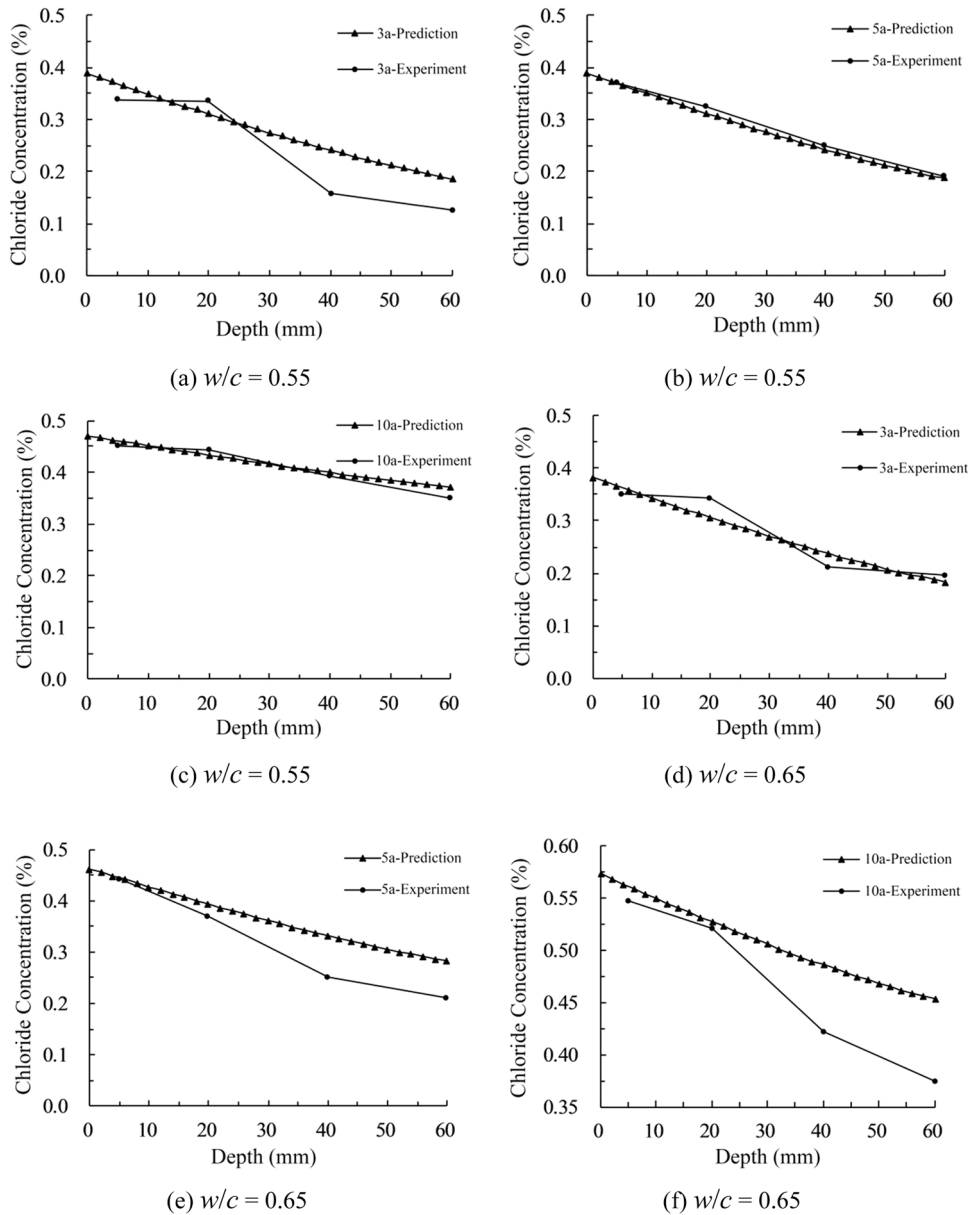


Fig. 2. Comparison between numerical simulation and experimental data.

size was between 5 and 20 mm, the ITZ thickness was 0.1 mm, the ITZ diffusivity was 10 times that of mortar, and the volume content of aggregate was 30%. The volume content of the ITZ was 1.175%. To appraise the chloride transport capacity, the initial parameter is set as $D_{cp} = 7.476 \times 10^{-12} \text{ m}^2/\text{s}$, $D_{itz} = 7.476 \times 10^{-11} \text{ m}^2/\text{s}$ [33], and $w/c = 0.4$. On the diffusion boundary, assuming that the chloride concentration on the upper, lower and right surfaces are zero, set the chloride concentration on the left surface to 0.5%.

5.1. Temperature

To lucubrate the impact of temperature on chloride migration, five temperature values ($T = 5 \text{ }^\circ\text{C}$, $20 \text{ }^\circ\text{C}$, $35 \text{ }^\circ\text{C}$, $50 \text{ }^\circ\text{C}$ and $65 \text{ }^\circ\text{C}$) and exposure time of 90 d were selected for numerical analysis. Fig. 3 shows the chloride ion concentration distribution at different temperatures. The results show that with the increase of temperature, the chloride transmission is accelerating, and the chloride diffusion depth is also increasing. The temperature increases from $5 \text{ }^\circ\text{C}$ to $65 \text{ }^\circ\text{C}$, and the chloride diffusion depth rises by 3.3 times.

Fig. 4 presents the variation of chloride migration depth under different temperatures. As can be noted from Fig. 4, the chloride content fluctuates at a specific temperature, which is caused by the blocking of aggregates and the enhancement of the ITZ. The general law shows that when the temperature increases, chloride transmission is significantly accelerated, and chloride concentration

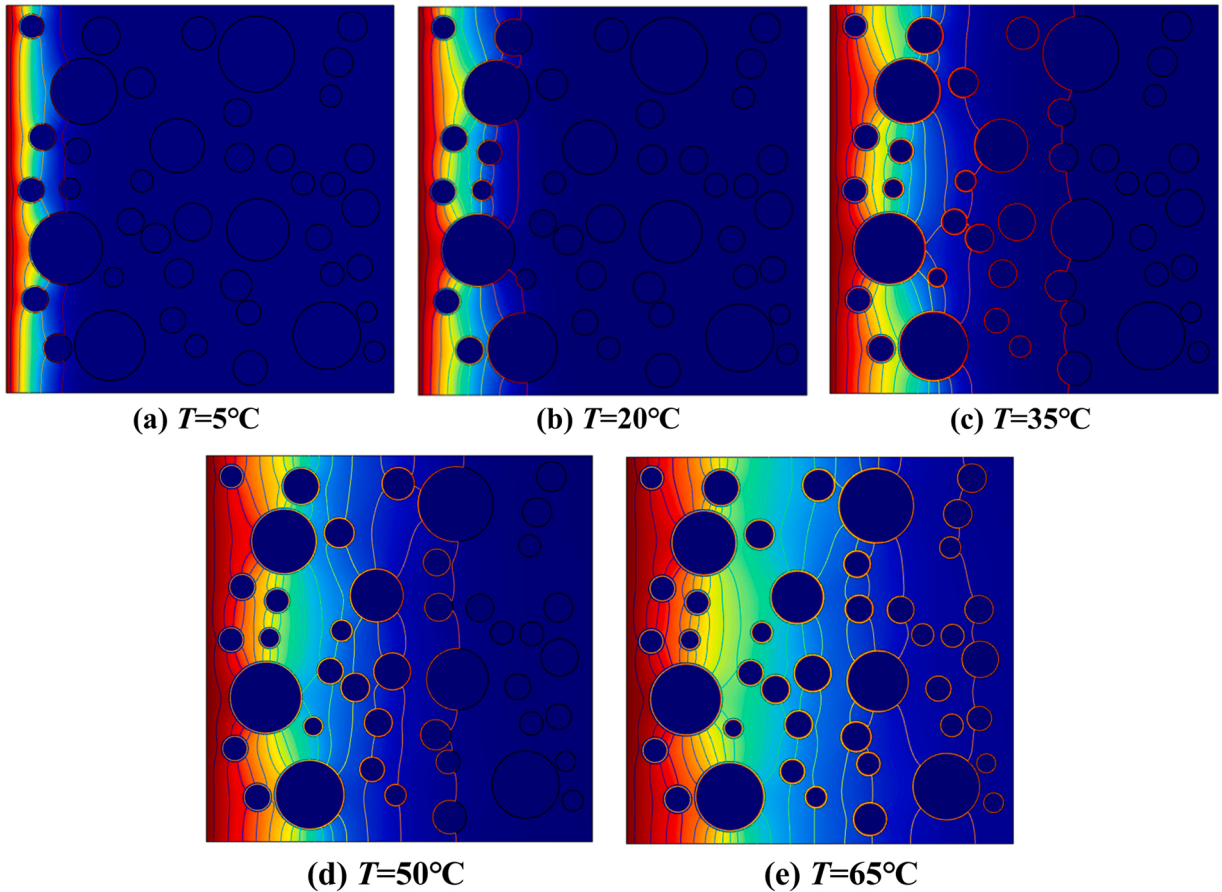


Fig. 3. Chloride concentration distributions under different temperature.

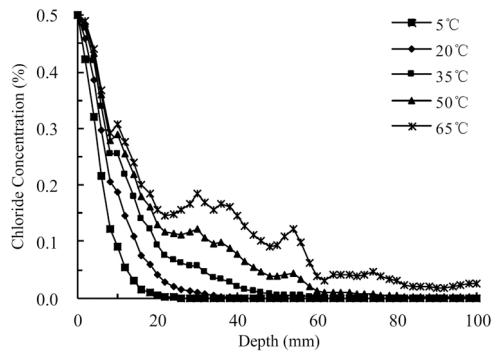


Fig. 4. The influence of temperature on the depth of chloride diffusion.

increases at the same depth. The penetration depth also rises with temperature.

5.2. Relative humidity in concrete

Similarly, the impact of relative humidity on chloride transmission, five groups of relative humidity ($RH=20\%$, 40% , 60% , 90% , 100%) were selected for numerical analysis. The distribution diagram of chloride concentration under different humidity is presented in Fig. 5. This figure indicates that the greater the relative humidity, the greater the average chloride concentration at a certain depth. Relative humidity increases from 20% to 100% , and the chloride ion diffusion depth expand by 4.3 times.

The relationship between chloride ion diffusion depth and concentration under different relative humidity is described in Fig. 6. This figure manifests that the greater the relative humidity, the greater the average chloride concentration at the same depth. Between

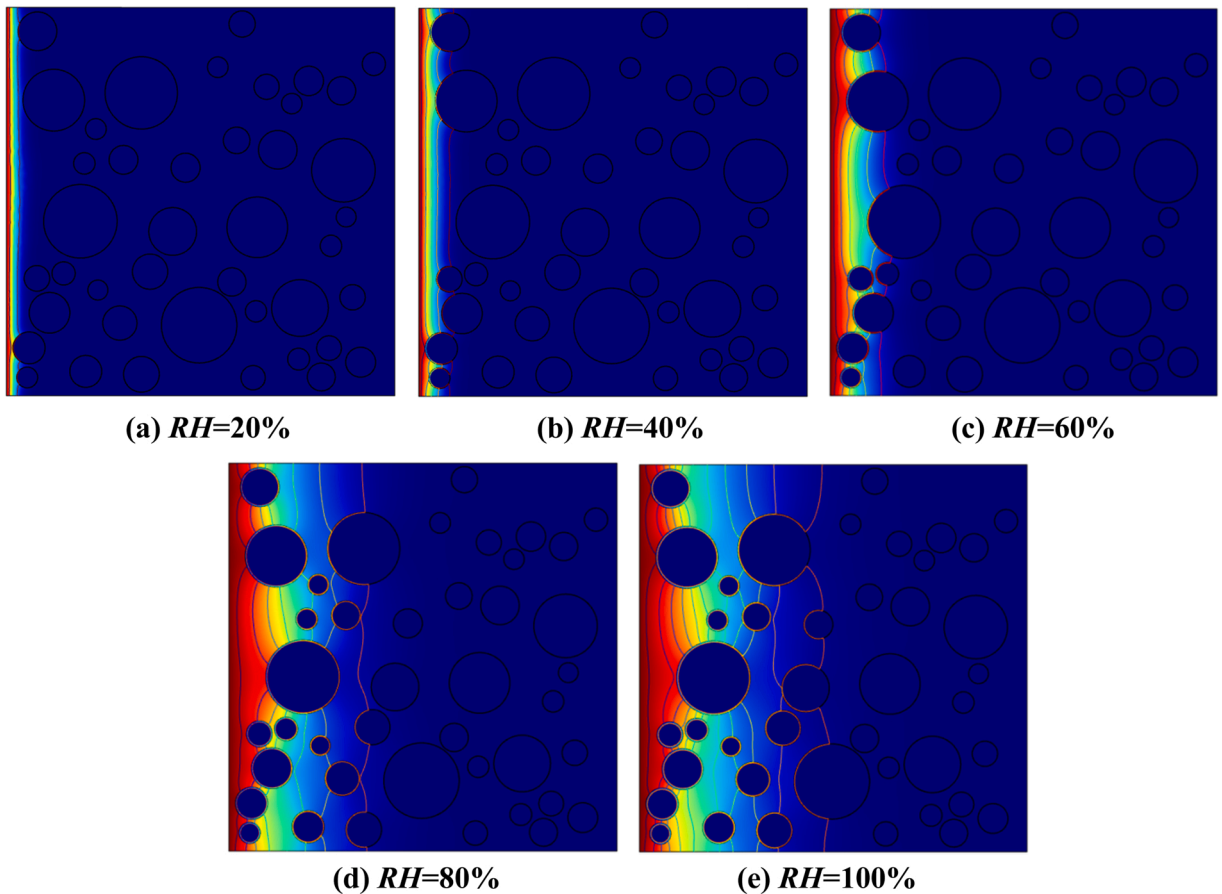


Fig. 5. Chloride concentration distributions under different humidity.

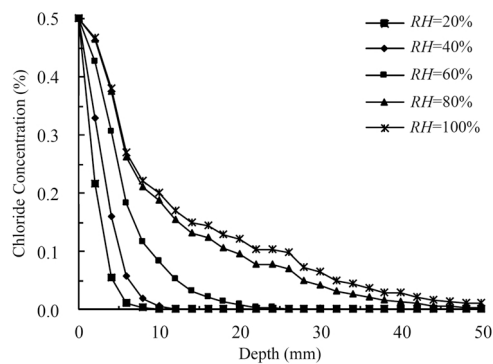


Fig. 6. The impact of relative humidity on the depth of chloride migration.

40% and 80%, relative humidity has a more significant effect on chloride transmission in concrete.

5.3. Water-cement ratio

The influence of five groups of water-cement ratios ($w/c = 0.2, 0.3, 0.4, 0.5, 0.6$) on the chlorine transport concentration distribution is shown in Fig. 7. With the w/c increase, the transmission of chloride in concrete accelerates, and the diffusion depth of chloride increases accordingly. The w/c augmented from 0.2 to 0.6, the diffusion depth of chloride increased by 1.3 times.

The relationship between chloride ion diffusion depth and concentration under different w/c is represented in Fig. 8. When the w/c rises, although chloride concentration fluctuates at a certain depth, the overall performance is that the average chloride content

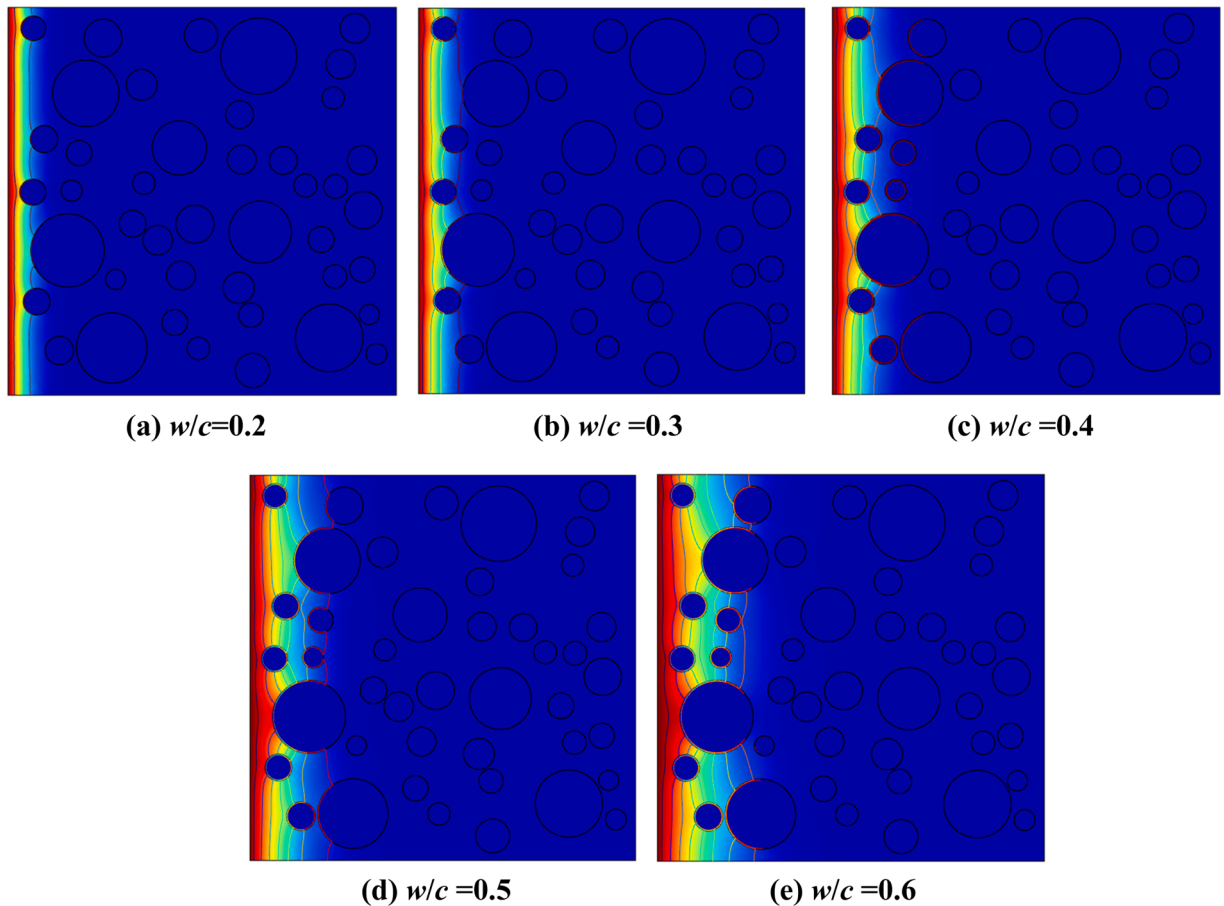


Fig. 7. Chloride concentration distributions under different water-cement ratios.

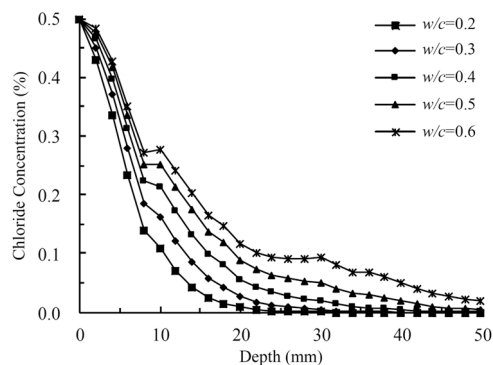


Fig. 8. The influence of water-cement ratios on the depth of chloride diffusion.

increases at the same depth. This is due to the expansion of the w/c resulting in the enhancement of porosity and permeability of concrete.

5.4. Concrete deterioration

The effects of four groups of concrete degradation influencing coefficients ($K = 4, 8, 12, 16$) on chloride ion transfer were studied. The chloride concentration profiles for different concrete degradation affecting coefficients are shown in Fig. 9. This figure manifests that the chloride diffusion rate accelerates with the increase of the deterioration influence coefficients. The degradation effect coefficient increases from 4 to 16, and the diffusion depth increases by 0.7 times.

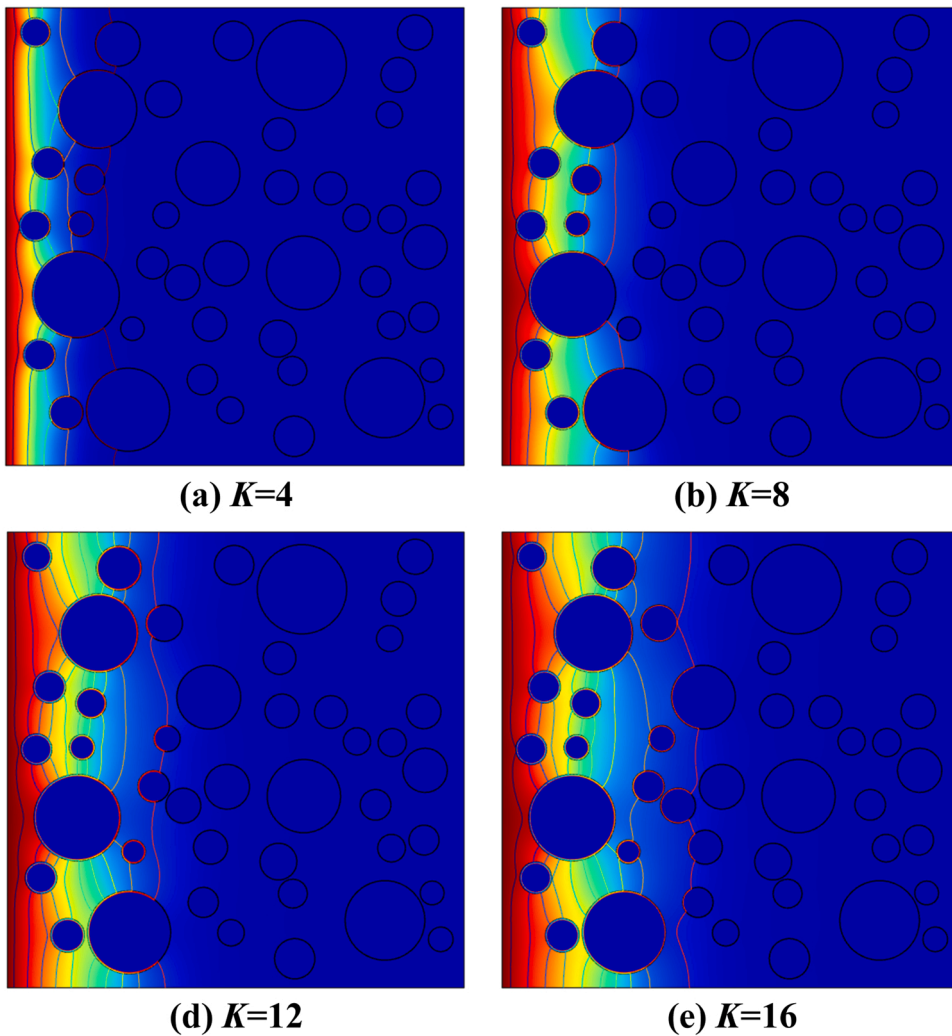


Fig. 9. Chloride concentration profiles for under different concrete degradation coefficients.

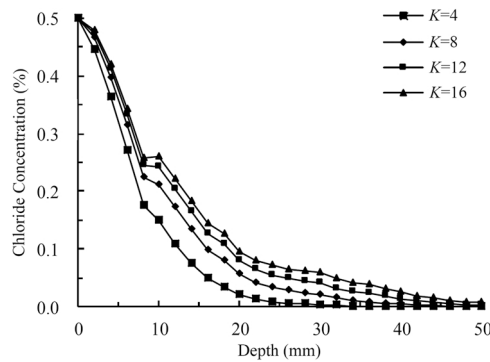


Fig. 10. The influence of degradation degrees on the depth of chloride diffusion.

The relationship between chloride ion diffusion depth and concentration under diverse concrete degradation coefficients is presented in Fig. 10. From Fig. 10, with the raising of depth, the chloride content reduces gradually. With the raising of influence coefficient of degradation, the chloride concentration at the same depth raises.

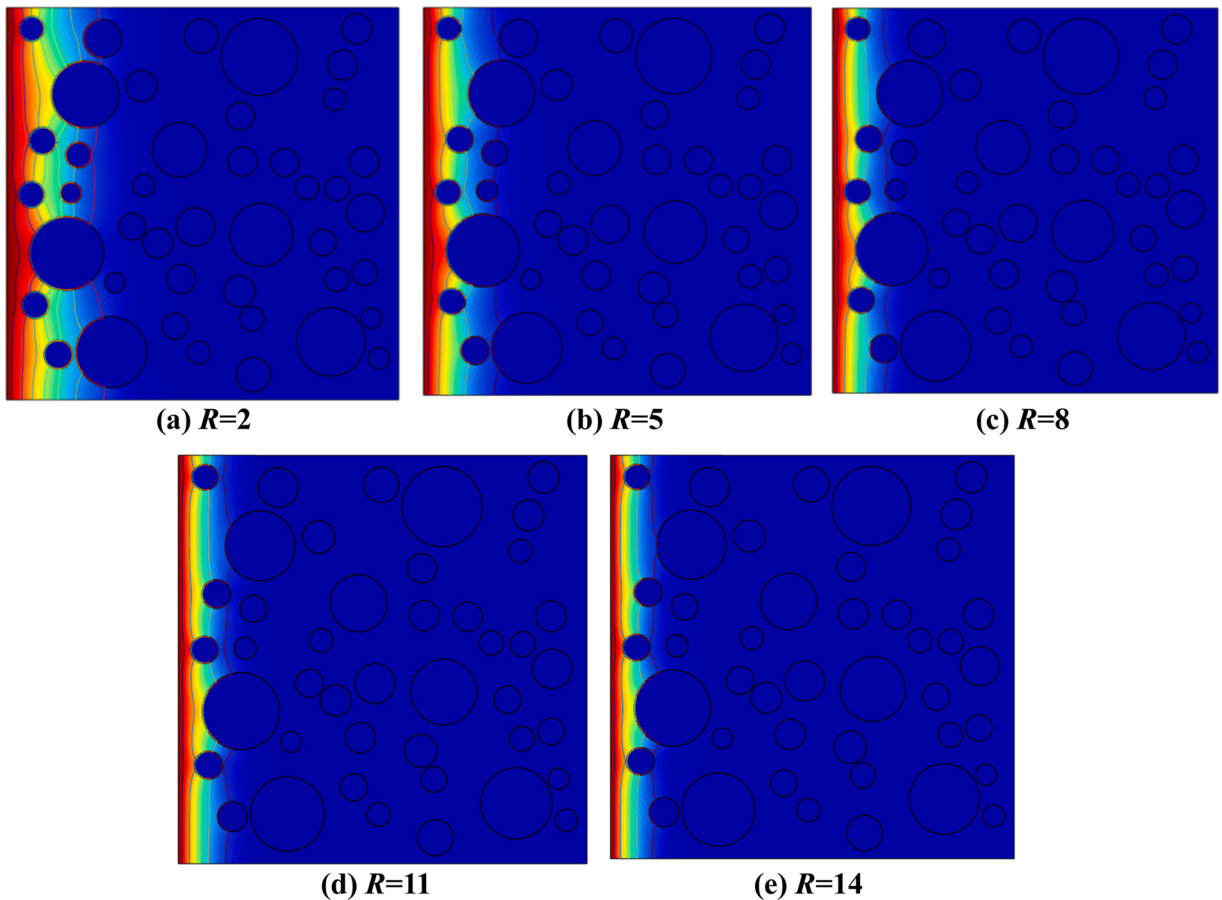


Fig. 11. Chloride concentration profiles for under different chloride binding degrees.

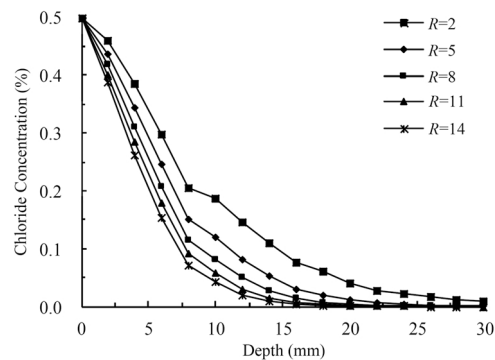


Fig. 12. The influence of different degrees of chloride ion binding on the depth of chloride migration.

5.5. Chloride ion binding

The effects of five different degrees of chloride ion binding ($R = 2, 5, 8, 11,$ and 14) on chloride migration in concrete were analyzed. Fig. 11 explains the chloride distribution under diverse chloride binding degrees. Obviously, the stronger the chloride binding, the lower the free chloride content. When the chloride binding coefficient rises from 2 to 14, the depth of chloride diffusion decreases by 0.9 times.

Fig. 12 shows the relationship between the average chloride concentration and depth under diverse binding effects. It can be found that the stronger the chloride binding, the lower the chloride concentration at a certain depth.

6. Conclusions

In this paper, through an in-depth analysis of influencing factors of chloride transmission, a chloride migration model considering key factors was developed. The proposed model was validated by the test data of marine concrete. On a mesoscale, the impact of key factors on chloride transmission was analyzed by numerical simulation. Combined with the above results, the following observations can be reached:

- (1) For optimum analysis of the influencing factors of chloride migration, a novel and reliable chloride migration model is established to generally consider the influences of water-cement ratio, time, chloride ion binding effect, temperature, relative humidity, and concrete deterioration in concrete.
- (2) Temperature increase will accelerate chloride transmission. When the temperature rises from 5 °C to 65 °C, the chloride diffusion depth increases by 3.3 times.
- (3) Relative humidity augment can significantly promote chloride migration behavior. When relative humidity increases from 20% to 100%, the chloride diffusion depth increases 4.3 times.
- (4) The chloride diffusion performance in concrete enhance with the rise of water-cement ratio and concrete deterioration.
- (5) The greater the binding effect, the lower the content of free chloride in concrete.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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