

Research Article

A Mesoscopic Model for Predicting the Chloride Diffusivity' Representative Volume Element in Concrete as a Random Composite

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Chloride diffusivity is the most crucial factor for evaluating durability and predicting service life of concrete structures exposed to chloride environment. In the present paper, concrete is considered as a random heterogeneous composite of three phases: the aggregates, the matrix, and the interfacial transition zones (ITZ) between them. A mesoscopic model has been established based on the random aggregate model and a numerical model for calculating the chloride diffusivity has been proposed. The influences of aggregate size and the properties of the interfacial transition zones on chloride diffusivity of concrete are also discussed. The chloride diffusivity is size dependent because aggregates are randomly distributed in concrete. The chloride diffusivity under different sizes is analyzed by numerous numerical simulations, and the representative volume element (RVE) of chloride penetration into concrete is researched by the statistical method.

1. Introduction

Chloride-induced corrosion of reinforcement has been identified as one of the most predominant degradation mechanisms for reinforced concrete (RC) structures. Service life of RC structures mostly depended on the chloride diffusion coefficient of concrete. In view of its importance, considerable research has been undertaken, both experimentally and theoretically, on the determination of the chloride's diffusion coefficient of concrete. Concrete can be viewed as a composite material consisting of aggregate and mortar, where the aggregate is randomly distributed in the concrete.

On the mesoscopic scale $(10^{-6} \text{ m to } 10^{-3} \text{ m})$, the chloride diffusion coefficient in concrete that is viewed as a multiphase composite can be expressed as follows:

$$D_{\rm c}^{\rm eff} = f[C(i), D(i)], \qquad (1)$$

where C(i) is the volume fraction of the *i* phase and D(i) is the diffusion coefficient of chloride in the *i* phase. Some previous researchers have studied the diffusion coefficient of chloride in concrete by considering concrete as a two-phase composite. Zheng and Zhou [1] considered mortar as a twophase composite and developed an analytical model to predict the chloride's diffusion coefficient. Hobbs [2] and Xi and Bazant [3] considered concrete as a two-phase composite consisting of aggregate and mortar and proposed a model to predict the theoretical equations for the diffusion coefficient of chloride in concrete. During concrete placement, a thin layer of transition zone with large porosity appears at the interface between the aggregate and mortar due to the "wall effect" [4]. Mehta and Monteiro [5] showed that at mesoscopic scale, concrete can be viewed as a threephase composite consisting of aggregate, mortar, and transition zone. Bentz and Garboczi [6] stated that the thickness of the transition zone is generally about $20 \,\mu m$.

Netami and Gardoni [7] considered the thickness of the transition zone to be $30-100 \,\mu\text{m}$ from infiltration test results. Scrivener and Gartner [8] derived the thickness of the transition zone between 10 and $100\,\mu m$ by backscattered electron imaging. The presence of aggregate has diametrically opposite effects on the transport of chloride in concrete. On the one hand, the introduction of aggregate increases the curvature of the chloride's transport path and slows down the transport of chloride in concrete; on the other hand, the introduction of aggregate generates a transition zone between aggregate and mortar, which opens new channels for the transport of chloride in concrete and accelerates the transport of chloride in concrete. Yang and Cho [9], Care [10], and Kato and Uomoto [11] studied the transport of chloride in the transition zone by accelerated tests. Garboczi and Bentz [12] viewed concrete as a three-phase composite consisting of aggregate, mortar, and transition zone, and theoretically investigated the diffusion coefficient of chloride in concrete. Care and Herve [13] looked at concrete as an nphase composite and investigated the diffusion coefficient of chloride. At the mesoscopic scale, the use of numerical tools to predict the chloride diffusion coefficient is also a hot topic in durability research. Wang et al. [14] developed a model to predict the diffusion coefficient of chloride's diffusion in concrete using a lattice model. Zhou and Li [15] studied the permeability properties of concrete by looking at concrete as a three-phase composite and discussed specifically the effect of the transition zone on the permeability properties. Zheng et al. [16] modeled the aggregate, mortar, and transition zone separately and developed a numerical model to predict the chloride's diffusion in concrete.

Yu et al. [17] studied the chloride diffusion in concrete with different aggregate volume ratios (AVR) and interfacial transition zone (ITZ) by the random aggregate model. Zheng et al. [18] proposed a numerical algorithm for evaluating the chloride diffusion coefficient of concrete with crushed aggregates. Liu et al. [19, 20] developed a multiphase transport model to explains how the shape of aggregates affects the chloride diffusion in concrete and the effects of aggregates and corresponding ITZ on chloride diffusion in concrete and mortar.

The distribution of aggregates in concrete has two diametrically opposite effects on the transport of chloride in concrete. The random distribution of aggregates leads to values of the chloride's diffusion coefficient in concrete that are not constant, making it necessary to discuss the representative volume element (RVE) of the chloride's diffusion coefficient. Researchers have given different definitions of RVE depending on the purpose of the study. Hill [21] stated that RVE must contain enough information to reflect the microstructural inhomogeneity of the material (inclusions, particles, pores, fibers, etc.). Hashin [22] stated that RVE should be large enough to reflect enough information about the microstructure to satisfy its representativeness requirement. Drugan and Wills [23] stated that the RVE should be large enough relative to the microstructure to ensure that it reflects. Ostoja-Starzewski [24] considered RVE as the smallest volume containing enough statisticalmechanical properties that an increase in volume does not

cause a change in material properties. Kaint et al. [25], Gitman et al. [26], and Segurado and Llorca [27] argued that the properties of RVE must reflect the macroscopic properties of the material and be able to adequately reflect information about the microstructure of the material. Gitman et al. [26] gave the expression for the definition of RVE as follows:

$$\left|\frac{1}{V_{\rm L}+{\rm d}V}\int_{V_{\rm L}+{\rm d}V}g(V){\rm d}V-\frac{1}{V_{\rm L}}\int_{V_{\rm L}}g(V){\rm d}V\right|<\delta.$$
(2)

Equation (2) holds that when the specimen size reaches the RVE size, for any given positive number, the difference between the mean value of the field variable is less than the mean value.

According to the above scholars' definition of RVE, the RVE should be large enough relative to the microstructure to contain enough microscopic information and at the same time be much smaller than the macrostructure size. For composite materials, the size of RVE varies for different physical problems. The current studies on RVE of concrete mainly focus on elastic modulus, strength, etc. [28–31], while there are few studies on RVE of chloride's diffusion in concrete, so it is necessary to study the size dependence of chloride's diffusion in concrete and provide a theoretical basis for experimental testing of chloride diffusion coefficients and assessment of concrete structures in chloride environments.

In this study, a mesoscopic numerical model for predicting the chloride's diffusion coefficient is first developed, and the chloride's diffusion coefficient of concrete specimens of different sizes is analyzed to discuss the size dependence of the chloride diffusion coefficients, and finally, a RVE of chloride diffusion in concrete is discussed.

In this study, concrete was viewed as a three-phase composite material consisting of aggregate, mortar, and transition zone, and the mesoscopic scale structure of concrete is established using the aggregate stochastic model, and a mesoscopic numerical model for calculating the diffusion coefficient of chloride in concrete was developed. Considering that concrete being a nonhomogeneous material, the diffusion coefficient of chloride in concrete was size dependent. The diffusion coefficient of chloride at different sizes of specimens was investigated by 31,200 times numerical analysis, and the representative volume units of chloride diffusion in concrete were discussed by statistical analysis.

2. Numerical Modeling of Chloride Diffusivity

2.1. Random Aggregate Structural Model. In order to determine the diffusion coefficient of chloride's diffusion in concrete on the mesoscopic scale, it is first necessary to simulate the mesoscopic structure of concrete. Concrete at the mesoscopic scale can be viewed as a three-phase composite consisting of aggregate, mortar, and transition zone. At the mesoscopic scale, many scholars have established mesoscopic-structure models of concrete, such as the microplane model by Bazant and Gambarova [32, 33], the hard particle contact model by Zubelewicz and Bazant [34], and the random aggregate model proposed by Wang et al. [35]. These structural models provided reliable computational models for studying the macroscopic properties of concrete. Among them, the random aggregate model proposed by Wang et al. is very similar to the mesoscopic view structure of actual concrete in terms of shape, size, and coarse aggregate distribution. Therefore, in the paper, the random aggregate model proposed by Wang et al. would be used to study the chloride's diffusion in concrete. The geometric position of the aggregate in the concrete body obeys a certain distribution law and the geometric position of the aggregate can be determined spatially with the help of the Monte Carlo method. And then the random aggregate model of concrete is established according to the gradation of the aggregate and the geometric form of the aggregate. The method is to segment the coarse aggregate according to the grading curve, to calculate the quantity of aggregate in each segment by combining the volume content of the aggregate and the size of the concrete specimen, and to inspect samples according to its particle size and shape. These aggregates are considered to be polygonal in shape in the plane and are considered that the polygon have a geometric center form which the local coordinate system of the aggregate is established. In the process of generating these aggregates, the geometric location of these aggregates polar center, the number of sides of the aggregate polygon (random distribution with the number of sides of [3, 10]), and the polar diameter and polar angle of the polygon corner points are considered as random variables. The Monte-Carlo method is used in the take and place method that aggregates are buried in the mortar phase randomly and the internal aggregates are considered not to overlap, and the distance from the aggregates to the boundary is proportional to the particle size of the aggregates to simulate the sidewall effect between the coarse aggregates and the concrete boundary, and the aggregates gradation obeys Fuller curve:

$$Y = 100 \left(\frac{R - R_{\min}}{R_{\max} - R_{\min}} \right)^{0.5},$$
 (3)

where *Y* denotes the cumulative mass fraction of aggregates with particle size below *R*, R_{max} is maximum aggregate actual size, and R_{min} is minimum aggregate actual size.

Figure 1(a) shows the distribution of aggregates for a specimen size of $150 \text{ mm} \times 150 \text{ mm}$, where $R_{\text{max}} = 20 \text{ mm}$, $R_{\text{min}} = 5 \text{ mm}$, and aggregate volume percentage is 40%. In the finite element mesh of the random aggregate model, considering the particularly thin thickness of the transition zone, which is usually less than 100 um, the transition zone is dissected with 2-node linear mesh in the finite element model, as shown in Figure 1(d). For mortar and aggregate, 6-node triangular meshes are used for meshing of mortar and aggregates are shown in Figures 1(b) and 1(c).

2.2. Finite Element Analysis of Chloride's Diffusivity. Provided that concrete is in a saturated state, chloride ingress into concrete is by diffusion due to existing concentration gradient. The flux J_c of chloride in saturated concrete due to diffusion is governed by Fick's first law:

$$\mathbf{J}_{\rm c} = -D_{\rm c} \operatorname{grad}(C_{\rm fc}),\tag{4}$$

where J_c is the flux of chloride due to diffusion, D_c is the effective chloride diffusion coefficient, and C_{fc} is the concentration of chloride dissolved in the pore solution. For steady chloride diffusion process, establishing chloride mass conservation results,

$$\operatorname{div}[\operatorname{grad}(C_{\mathrm{fc}})] = 0. \tag{5}$$

In solving the numerical model for the diffusion coefficient of chloride ions in concrete, the boundary conditions are shown in Figure 2 and summarized as follows:

$$C_{fc} = C_2 \quad x = 0,$$

$$C_{fc} = C_1 \quad x = L_x,$$

$$\frac{\partial C_{fc}}{\partial \mathbf{n}} = 0 \quad y = 0, L_y.$$
(6)

According to the above boundary conditions, the chloride flux in the direction of the finite element model can be obtained from the finite element analysis; then, the average chloride's flux in the direction can be expressed as follows:

$$\overline{J}_x = \frac{1}{L_y} \int_{L_y} J_x \mathrm{d}y.$$
⁽⁷⁾

From Fick's first law, the diffusion coefficient of chloride in concrete can be expressed as follows:

$$D_{\rm c} = \frac{L_x}{C_1 - C_2} \overline{J}_x.$$
 (8)

2.3. Validation of the Numerical Model. Yang and Cho [9] investigated the diffusion coefficients of chloride in mortars with different aggregate volume contents by accelerated tests. In their experiments, Yang and Cho cast five groups of mortar specimens with different aggregates volume contents (0%, 10%, 20%, 30%, and 40%) and obtained the diffusion coefficients of chloride in mortar with different volume contents of aggregates by the tests. Figure 3 shows the diffusion coefficient of chloride in mortar predicted by the numerical model compared to the experimental values. In numerical tests, the random distribution of aggregates in the mortar is fully considered and 100 numerical samples are generated for each aggregate volume content. It can be seen that the numerical results agree well with the experimental results, which verifies the reliability of the numerical model in this study to some extent.

3. Statistical Analysis of Chloride Diffusivity

Concrete is a heterostructured composite in which aggregates are randomly distributed. In most case, its composite behavior is size dependent. Many efforts have been given to



FIGURE 1: Random aggregate model and its mesh. (a) Random aggregate model. (b) Mortar mesh profile. (c) Aggregate mesh profile. (d) Transition zone mesh profile.



FIGURE 2: Schematic diagram of the model for solving the chloride diffusion coefficient.

research the size effect on concrete's elastic modulus and strength [26–29]. However, for the same composite, different physical processes can have different size effects. In the study, numerical samples of different sizes are generated randomly in this section and the statistical characteristics of chloride diffusivity are analyzed. Then, a concept closely related to the statistical characteristics of chloride diffusivity, representative volume element (RVE), is discussed in terms of numerical sample size and expected simulation error.



FIGURE 3: Numerical solution versus Yang test value.

FIGURE 4: Random aggregate model with different sizes of specimens. (a) 100 × 100 mm. (b) 150 × 150 mm. (c) 200 × 200 mm. (d) 250 × 250 mm.



FIGURE 5: Relative chloride's diffusion coefficients in concrete at different sizes (k = 0).

3.1. Chloride Diffusivity Analysis of Random Samples. Chloride's diffusivity of concrete under different sizes is analyzed in this section. Figure 4 shows samples of different size generated by the RAS model. Numerical sample sizes are scaled for 100 mm to 250 mm with 50 mm as increase grade. For each sample size, 100 random aggregate structures are generated with $D_{\min} = 5$ mm, $D_{\max} = 20$ mm, the aggregates' volume fraction $f_{agg} = 0.4$, and the aggregates' distribution obeys Fuller's curve. In all numerical samples, the ITZ width is retained as $t_{\text{TTZ}} = 30 \,\mu$ m, four chloride diffusivity contrast rations are taken: $\gamma = D_{\text{ITZ}}/D_{\text{m}} = 1, 5, 10$, and 25, and three linear bind factors are considered k = 0, 0.08, and 0.16. Thus, for some k and γ values, 2600 simulations are taken, and 31200 simulations are performed for all chloride diffusivity analysis. Figures 5–7 show the numerical simulation results and the mean values for different size. It can be observed that the relative chloride diffusivity is dependent on k and γ , the chloride diffusivity increases with the increase of γ and with the decrease of k. The dispersion of chloride decreases substantially with the increase of sample size. This size-



FIGURE 6: Relative chloride's diffusion coefficients in concrete at different sizes (k = 0.08).



FIGURE 7: Relative chloride's diffusion coefficients in concrete at different sizes (k = 0.16).

		U	•	
k	γ	$D_{\rm c}^2$	A ₂	β
	1	0.2352	5.3555	1.0351
0	5	0.3307	4.8673	1.0527
0	10	0.6739	3.6186	1.0894
	25	3.1959	1.3432	1.1120
	1	0.0598	5.0678	1.0370
0.00	5	0.1672	2.5561	1.0561
0.08	10	0.5252	1.4607	1.0941
	25	3.0919	0.5805	1.1212
	1	0.0282	4.5781	1.0384
0.16	5	0.1395	1.4822	1.0594
0.16	10	0.5025	0.8720	1.1020
	25	3.0840	0.4909	1.1542

TABLE 1: Parameter fitting results for different k values and γ values.



FIGURE 8: Parameter fitting results for different γ values (k = 0).

dependent dispersion is characteristic for properties of random heterogeneous composites. Theoretically this dispersion can vanish as the sample's size attains the representative volume element (RVE)'s size. The REV's size for chloride diffusion in concrete is evaluated by using the statistical method.

To explore the statistical characteristics of chloride's diffusivity in terms of samples size, the binding effect, and ITZ properties, the chloride diffusivity are considered to be an ergodic stationary random function. Then, a power law is chosen for the variance of overall chloride diffusivity:

$$D_{\rm c}^2(A) = D_{\rm c}^2 \times \left(\frac{A_2}{A}\right)^{\beta},\tag{9}$$

where $D_c^2(A)$ represents the variance of chloride diffusivity of a realized numerical sample of size A, A_2 is a pure geometrical factor, and β is the power index. The above relation can be converted to linear relationship in the logarithm scale:

$$\log\left[D_{\rm c}^2(A)\right] = -\beta\log(A) + \left[\log(D_{\rm c}^2) + \beta\log(A_2)\right].$$
(10)



FIGURE 9: Parameter fitting results for different γ values (k = 0.08).



FIGURE 10: Parameter fitting results for different γ values (k = 0.16).

k	γ	RVE size (mm)		RVE size/D _{max}			
		$\varepsilon = 1\%$	$\varepsilon = 2\%$	$\varepsilon = 5\%$	$\varepsilon = 1\%$	$\varepsilon = 2\%$	$\varepsilon = 5\%$
0	1	458.08	234.49	96.76	22.90	11.72	4.84
	5	451.06	233.49	97.78	22.55	11.67	4.89
	10	432.10	228.70	98.62	21.61	11.44	4.93
	25	423.70	227.17	99.65	21.19	11.36	4.98
0.08	1	429.89	220.33	91.60	21.49	11.02	4.58
	5	420.24	218.00	91.55	21.01	10.90	4.58
	10	409.30	217.22	94.01	20.47	10.86	4.70
	25	401.76	216.51	95.62	20.09	10.83	4.78
0.16	1	404.42	207.46	85.84	20.22	10.37	4.29
	5	398.90	207.36	87.31	19.95	10.37	4.37
	10	394.53	210.34	91.58	19.73	10.52	4.58
	25	401.04	219.99	99.45	20.05	11.00	4.97

TABLE 2: RVE size estimation under different expectation errors.

TABLE 3: RVE size estimation for different number of numerical tests.

k	γ	RVE size (mm)		RVE size/ D_{MAX}			
		$N_{0} = 1$	$N_0 = 5$	$N_{0} = 10$	$N_{0} = 1$	$N_0 = 5$	$N_{0} = 10$
0	1	458.08	210.53	150.63	22.90	10.53	7.53
	5	451.06	210.01	151.10	22.55	10.50	7.56
	10	432.10	206.43	150.18	21.61	10.32	7.51
	25	423.70	205.48	150.46	21.19	10.27	7.52
0.08	1	429.89	197.82	141.62	21.49	9.89	7.08
	5	420.24	196.14	141.27	21.01	9.81	7.06
	10	409.30	196.16	142.90	20.47	9.81	7.15
	25	401.76	196.00	143.89	20.09	9.80	7.19
0.16	1	404.42	186.32	133.45	20.22	9.32	6.67
	5	398.90	186.63	134.56	19.95	9.33	6.73
	10	394.53	190.09	138.80	19.73	9.50	6.94
	25	401.04	199.71	147.91	20.05	9.99	7.40

The variance of chloride diffusivity D_c^2 can be expressed as follows:

$$D_{\rm c}^2 = \sum_{i=1}^3 \left(D_i^* \right)^2 f_i - \left(\sum_{i=1}^3 D_i^* f_i \right)^2, \tag{11}$$

where f_i represent the *i*th phase volume fraction and D_i^* represents the effective chloride diffusivity in the *i*th phase.

Submitting the numerical results in Figures 5-7 into equation (10), the variance of overall chloride diffusivity is fitted well for different contrast rations y and binding coefficient k. Table 1 and Figures 8–10 represent the relevant fitting results. It can be observed that the correlation coefficient is larger than 0.985 for all cases, which indicates that the power law is well suited. A_2 decreases sharply as the contrast ratio γ increase. This can be attributed to the small fraction but high local chloride diffusivity of ITZ. The power index β is larger than 1 and also very close to 1, which means that the variance of chloride diffusivity decreases slowly with increase in the sample size. It can also be observed that the power index β increases with the increase in k and y values. This can be attributed to specific structures of three-phase composite behavior: impermeable aggregates coated by ITZ in which chloride diffuses faster than that in mortar. During the chloride diffusion process, an ITZ layer with high local chloride diffusivity renders the coated aggregates to "diffusion-able" to some extent. The value k and γ have the same effect on the dispersion of the equivalent chloride's diffusion coefficient in concrete. The larger the values of k and γ are, the smaller the difference between the diffusion properties of chloride in mortar and aggregate, thus making the dispersion of the effective diffusion coefficient of chloride in concrete smaller.

3.2. Estimation of RVE for Chloride Diffusion in Concrete. As a nonhomogeneous material, concrete needs to be studied at the mesoscopic scale if its macroscopic properties are to be clearly understood, and the RVE is the bridge between the macroscopic model and the mesoscopic model of concrete. Theoretically, the macroscopic properties are stable after the specimen size reaches the RVE size. The dispersion of the chloride's diffusion coefficient becomes smaller with increasing specimen size but never disappears as can be seen in the numerical results in Figure 5. The statistical numerical model calculates the RVE size with the expected accuracy according to the mathematical statistical theory [36]:

$$\varepsilon = \frac{2D_{\rm c}(A)}{M_0\sqrt{N_0}},\tag{12}$$

where M_0 is the mean value, N_0 is the number of independent samples, and ε is the expected error. Substituting equation (9) into equation (12), the size of the RVE at the expected error value can be expressed as follows:

$$A = A_2 \left(\frac{4D_c^2}{\varepsilon^2 M_0^2 N_0}\right)^{1/\beta}.$$
 (13)

By substituting the fitted data from Table 1 into (13), the size of the RVE for chloride's diffusion in concrete is obtained for different values k and γ and different expected errors ε . Table 2 shows the estimated RVE size of chloride's diffusion in concrete for different expectation error cases. It can be seen that when the expected error is 1%, the RVE size is about 390-460 mm, which is 19-23 times of the maximum aggregate size, while when the expected error is 5%, the RVE size is about 85-100 mm, which is 4-5 times of the maximum aggregate size. Another important application of equation (13) is to calculate the RVE size for different number of numerical tests. Table 3 shows the estimated RVE size for different number of numerical tests with an expected error of 1%. It can be seen that, for = 1, the RVE size is approximately 390-460 mm and 19-23 times the maximum aggregate size, while for = 10, the RVE size is approximately 130–151 mm and 6-8 times the maximum aggregate size.

4. Conclusion

The chloride's diffusion coefficient is the most critical factor for durability design, assessment, and life prediction of concrete structures in chloride salt environments. In the study, concrete is viewed as a three-phase composite material consisting of aggregate, mortar, and transition zone, and the mesoscopic structure of concrete is established using the aggregate random model to build a mesoscopic-scale numerical model for calculating the chloride's diffusion coefficient in concrete. Considering that concrete is a nonhomogeneous material and the diffusion coefficient of chloride in concrete is size dependence, the chloride's diffusion coefficients in specimens with different sizes were investigated by 31,200 times value analysis, and the RVE of chloride's diffusion coefficients in concrete were discussed by statistical analysis. The following conclusions can be drawn in this study.

- The chloride's diffusion coefficient in concrete is closely related to the aggregate volume content, the size of the aggregate particle size, and the properties of the transition zone.
- (2) The chloride's diffusion coefficient in concrete is related to the specimen size, the smaller the size, the greater the dispersion of chloride's diffusion coefficient, and the dispersion of chloride's diffusion coefficient decreases as the specimen size increases. The value k and the value γ have opposite effects on the mean value of chloride's diffusion coefficient,

the larger the value of k, the smaller the mean value of chloride's diffusion coefficient; the larger the value of γ , the larger the mean value of chloride's diffusion coefficient. The value of k and the value of γ have the same effect on the dispersion of chloride's diffusion coefficient, and the larger the value of k and the value of γ , the smaller the dispersion of chloride's diffusion coefficient. The dimensions of the RVE of chloride's diffusion in concrete were estimated by statistical analysis, which were related to the expectation error and the number of numerical samples.

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that they have no conflicts of interest regarding the publication of this paper.

Authors' Contributions

JianXiang Xie and Gang Lin contributed equally to this work.

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