

Five-phase modelling for effective diffusion coefficient of chlorides in recycled concrete

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With the utilisation of recycled aggregate concrete (RAC) in building construction, the durability of this material requires particular attention, especially in terms of chloride penetration. This paper presents a numerical study on the chloride diffusion mechanism within RAC. Considering the random distribution of recycled coarse aggregates (RCA), a five-phase RAC model – including new mortar, adherent old mortar, new interfacial transition zone (ITZ), old ITZ and original natural coarse aggregates – is proposed to predict the effective diffusion coefficient of chlorides in RAC. The parametric studies, based on a series of critical factors (i.e. volume fraction of RCA, adhesive ratio of old mortar, chloride diffusivity of adherent old mortar, thicknesses of old and new ITZs and chloride diffusivity of old and new ITZs), reveal the properties of each phase and their individual impact on the effective diffusion coefficient of chlorides in RAC. The results obtained indicate that, interestingly, the effective diffusion coefficient tends to vary in terms of its relationships with high-quality adherent old mortar, lower adhesive ratio of old mortar, smaller thickness of ITZs and relatively superior chloride penetration resistance of ITZs, which cannot be found from existing models and experiments.

Notation

C	concentration of chloride ions in cement paste matrix (mol/m^3)
D_{agg}	chloride diffusion coefficient of original natural coarse aggregate
D_{eff}	effective diffusion coefficient of chlorides
D_k	chloride diffusion coefficient of k th phase
$D_{\text{new,ITZ}}$	chloride diffusion coefficient of new interfacial transition zone
$D_{\text{new,mor}}$	chloride diffusion coefficient of new mortar
$D_{\text{old,ITZ}}$	chloride diffusion coefficient of old interfacial transition zone
$D_{\text{old,mor}}$	chloride diffusion coefficient of adherent old mortar
D_{RAC}	chloride diffusion coefficient of recycled aggregate concrete
h	height of the plain recycled aggregate concrete utilised in numerical model
J	flux of chloride ions ($\text{mol}/(\text{mm}^2/\text{s})$)
l	length of the plain recycled aggregate concrete utilised in numerical model

M_1	$M_1 = D_{\text{old,mor}}/D_{\text{new,mor}}$
M_2	$M_2 = D_{\text{old,ITZ}}/D_{\text{old,mor}}$ and $D_{\text{new,ITZ}}/D_{\text{new,mor}}$
R_{aom}	adhesive ratio of old mortar
T_{ITZ}	thickness of both new and old interfacial transition zones
$T_{\text{new,ITZ}}$	thickness of new interfacial transition zone
$T_{\text{old,ITZ}}$	thickness of old interfacial transition zone
t	time (s)
V_{RCA}	volume fraction of recycled coarse aggregates
x	coordinate in horizontal axis (mm)
y	coordinate in vertical axis (mm)
∇	Laplace differential operator

Introduction

To preserve natural resources and alleviate the burden on land-fill areas, one efficient way to utilise increasing amounts of construction and demolition waste is to form recycled coarse aggregates (RCA) for the production of recycled aggregate concrete (RAC) (Poon *et al.*, 2004; Su *et al.*, 2015a). However, because RCA derived from crushed concrete includes 65–70%

by volume of natural coarse and fine aggregates and 30–35% by volume of old cement pastes (Poon *et al.*, 2004; Wardeh *et al.*, 2014), compared with natural coarse aggregates (NCA), RCA have a higher water absorption capacity, lower density and Los Angeles abrasion coefficient and a more complex, inhomogeneous nature. Consequently, the properties of RAC, including mechanical properties and durability, are significantly different from those of natural aggregate concrete (NAC) (Olorunsogo and Padayachee, 2002; Su *et al.*, 2015b; Wardeh *et al.*, 2014; Yehia *et al.*, 2015; Zhang *et al.*, 2017a). To date, only some of the principal characteristics of RAC have been examined. For example, the experiments of Poon *et al.* (2004) indicate that the compressive strength of RAC is lower than that of NAC. Zhang and Zhao (2015) proposed an integrated interface parameter of RAC by an experimental study that included micro and macro properties, and concluded that the mechanical and durability properties of RAC are weakened as the proportion of RCA replacements increases. Bravo *et al.* (2015) analysed the durability performance of RAC using experimental methods and concluded that the use of recycled aggregate is highly detrimental, especially where fine recycled aggregates are used (Kumar *et al.*, 2017).

Previously, the above-mentioned negative associations meant that RCA was disposed of as a base filler for road construction (Bordelon *et al.*, 2009; Tam and Tam, 2008; Yang *et al.*, 2011). However, with the improvement of RAC mixture design, the ready availability of RCA sources and the addition of mineral admixture (Kou and Poon, 2015; Sim and Park, 2011; Tam and Tam, 2007a; Vázquez *et al.*, 2014; Zhao *et al.*, 2017), it is believed that RAC could be successfully employed in building construction in a safe and sustainable way (Banjad Pečur *et al.*, 2015). The relevant studies demonstrate that the durability and mechanical properties of RAC can be improved by a two-stage mixing approach (Tam and Tam, 2007a) and alternative parent concretes (Kou and Poon, 2015). The experiments of Sim and Park (2011) showed that the compressive strength, chloride penetration resistance and carbonation resistance ability of RAC with 100% RCA were sufficient to justify its application in structural concrete. Kim *et al.* (2013) reported that the use of fly ash only reduces the compressive strength of RAC by a small amount, and specimens including fly ash exhibit much higher chloride resistance than those without fly ash. Kou and Poon (2015) demonstrated that the RCA derived from higher strength concrete (80 and 100 MPa) could be used to replace 100% NCA for the production of high-performance concrete with lower drying shrinkage and higher resistance to chloride ion penetration. Zhang and Zhao (2016) investigated reinforced concrete beams with RCA under the long-term coupled effects of sustained loads and chloride ion ingress; their study suggested that the mechanical properties of RAC beams are likely to be acceptable, but caution should be exercised with regard to inferior durability.

Nevertheless, with the utilisation of RAC in building construction (Li, 2009; Poon and Chan, 2007), the durability problem should be given careful attention, especially in coastal environments and where de-icing salt is used in conditions of severe cold and cold regions. It is well known that the deterioration of the performance in a construction is affected to a certain degree by the penetration of ions, especially chloride ions and other species interaction. Thus, it is appropriate to obtain more detailed and fundamental information about the characteristics of ionic transport in RAC considering the multiple and uncertain properties of RCA.

To characterise the behaviours of ionic transport in RAC, considerable efforts have been made to assess ionic transport using traditional approaches, including experimental or analytical methods. For example, some studies have shown by experiment that the chloride penetration resistance of RAC decreases with the incorporation of RCA (Andreu and Miren, 2014; Kou and Poon, 2006); at the same time, the chloride penetration rate and binding capacity increase with the incorporation of RCA (Villagrán-Zaccardi *et al.*, 2008). Debieb *et al.* (2010) utilised contaminated crushed concrete aggregates to investigate the durability of RAC and found that contaminated RCA are much more sensitive to chlorides than to sulfates (Geng *et al.*, 2016). Faella *et al.* (2016) reported the durability performance of structural concrete made with RCA and fly ash. Their experimental results concluded that the chloride ions' penetration is affected by the presence of RCA in concrete, and that fly ash enhances the resistance to chloride ion penetration. In the analytical research, most publications have employed a method to predict the chloride diffusivity of RAC more accurately as a multi-phase material. Damrongwiriyanupap *et al.* (2011) adopted a dissimilar multi-phase analytical model, including natural aggregate, existing cement paste, surface treatment layer, new cement paste and effective media, which was simplified from a specific mixture design of RAC used in practice, to characterise the chloride diffusion in RAC. Xiao *et al.* (2013) applied a unified analytical model which considered RAC as a five-phase material, namely, RCA or NCA, new mortar, old mortar, new interfacial transition zone (ITZ) and old ITZ, to predict the chloride diffusivity. A three-phase model comprising old attached mortar, natural aggregate and new mortar developed by Ying *et al.* (2016) was utilised to predict the distribution of the chloride diffusion coefficient of RAC through a probability density evolution method.

Although the aforementioned experiments have shown the phenomenon of ion penetration in RAC, few of them have described the details of the chloride diffusion mechanism combined with hetero-structured RAC. These earlier reported experiments were also inappropriate to tackle the influences of individual components of RAC on chloride diffusion in concrete. The analytical studies mentioned above could only focus

on the transport of chlorides in a one-dimensional medium. Just one of them makes use of a multi-phase model, which is limited to a certain method of mixture design. The ability to model the influence of RCA as a constituent material is appealing. This would support efforts to obtain a more comprehensive understanding of chloride diffusion and assist in the development of more accurate predictions of degradation and service life of RAC.

As regards NAC, the present numerical simulation models generally regard NAC as a two-phase (Feng *et al.*, 2016; Liu *et al.*, 2012; Zeng, 2007), three-phase (Liu *et al.*, 2015a; Zheng and Zhou, 2008) or four-phase (Sun *et al.*, 2011) composite material. Note that, similarly to NAC, RAC is also a multi-phase material with complicated microstructural organisation as new mortar, RCA, new ITZ between the RCA and the new mortar, and old ITZ between the original NCAs and adherent old mortar. A three-phase model as natural or recycled coarse aggregate, a porous matrix and ITZ between them developed by Zhou and Ke-Fei (2012) was used to solve the steady permeation problem on a two-dimensional numerical sample with the finite-element method. A five-phase RAC model, with only one recycled aggregate, with old and new ITZ as interphases, new mortar, old attached mortar and original aggregate as continuous phases, was further developed and applied to describe the effect of individual recycled aggregate on chloride penetration in a localised part of the concrete (Xiao *et al.*, 2012). Ying *et al.* (2013) proposed four kinds of modelled RAC with different RCA replacement ratios to describe the effects of RCA distribution on chloride diffusion. An analogous five-phase and one-dimensional RAC model was also applied to develop and implement a stochastic service-life model for chloride-induced corrosion in reinforced RAC (Iii, 2014).

When using a numerical simulation method to investigate the ion transport properties of RAC, it is essential to take two important variables of RCA into account: the adherent old mortar remaining on the RCA, and the old ITZ between the original NCAs and the adherent old mortar. Consequently, owing to the RCA component, the mechanism of ion transport in RAC is more complicated than that in NAC. It has long been suspected that RCA can serve as weak or preferential parts for the ingress of aggressive species, yet the significance of their influence on the durability of RAC is not well understood. In particular, the adherent old mortar and the old ITZ between the RCA and the adherent old mortar are different from the new mortar and the new ITZ (Abbas *et al.*, 2009; de Juan and Gutiérrez, 2009; Hou *et al.*, 2015; Poon *et al.*, 2004; Tam and Tam, 2007b; Zhang *et al.*, 2017b). An individual ITZ model between new and old cement paste in RAC has even been developed by Li *et al.* (2016) based on the software CHMHYD3D. The results indicate that this kind of ITZ could strengthen the bonding of new and old cement paste.

In order to reveal the details of the chloride diffusion mechanism combined with the random distribution of RCA in RAC, on the one hand, the way in which chloride diffusion takes place in the inner RAC should be understood, and, on the other hand, how the individual constituents of RAC influence that chloride diffusion should be identified. Accordingly, in this paper, a numerical five-phase model at the mesoscopic scale of RAC is proposed to simulate chloride diffusion in RAC. It is very important to study and discriminate between each phase in terms of the effective chloride diffusion coefficient of RAC. Unlikely most existing research works on RAC, this study utilises the random distribution of RCA to model and analyse the effective chloride diffusion coefficient of RAC. The RAC is treated as a heterogeneous material composed of five phases, including the old and new ITZ as interphases, and the new mortar, adherent old mortar and original NCA as continuous phases. The chloride diffusion is assumed to take place in four phases, with the exception of the original NCA. Based on the proposed five-phase model, the given parameters (volume fraction of RCA, proportion and chloride diffusivity of adherent old mortar, thicknesses of old and new ITZs, and chloride diffusivity of old and new ITZs) are taken into consideration for their effects on the effective diffusion coefficient of chlorides in RAC. The present five-phase model reveals some important results regarding each phase's effects, which could not be found from the existing models or experiments.

Five-phase composite sphere prediction RAC model

As a multi-phase inhomogeneous material, RAC mainly incorporates RCA, new mortar and voids. As previously mentioned, the single RCA can be regarded as a three-component material, including the adherent old mortar, the original NCA and the ITZ between them. As the capillary pores and gel pores are both nano-sized compared with coarse aggregates, they can be roughly treated as homogeneous. However, because the microstructure of RAC is intrinsically variable and complex, in order to build up the RAC numerical model efficiently and integrally, some approximation is inevitable in order to model the effect of the RCA on the ionic diffusivity of RAC. In this paper, only the saturated case and the steady-state condition are taken into account when modelling RAC. The chloride ions principally diffuse through the mortar and ITZ phases by way of connected and water-filled pores, whereas the original NCA is regarded as an approximately impermeable phase. To simplify the shape of the aggregates, the RCA are modelled as polydispersed spheres. Although the aggregates in real concrete may not be perfectly circular, it has been verified that the effect of aggregate shapes on chloride diffusion in concrete is small (Li *et al.*, 2012; Liu *et al.*, 2017b). New and old ITZs are modelled as shells of different thickness that extend from the adherent old mortar and the original NCA, individually.

The physical properties of the multi-phase RCA and the interaction between each phase are represented by a partial schematic diagram of the five-phase composite sphere model of RAC in concrete, as shown in Figure 1(a). A two-dimensional series model is used to build a chloride diffusivity model of RAC, as shown in Figure 1(b). It is worth mentioning that, although the three-dimensional model can directly predict the chloride effective diffusion coefficient of concrete, with the higher number of dimensions, it is more time consuming and more expensive in terms of the computer simulation. In the one-dimensional case, Hobbs (1999) adopted simplified analytical prediction models including two-phase series and parallel models to predict the probable effect of the aggregate on chloride ion diffusion into saturated concrete within a given range water and cement ratio, neglecting the ITZ. Based on this prediction model, a modified two-dimensional and two-phase model proposed by Li *et al.* (2012), using the numerical simulation method, indicated the difference between the

effective diffusion coefficient for one dimension and two dimensions. Because the lower bounds of the effective diffusion coefficients for parallel and series models in one-dimensional and two-dimensional forms are dramatically different, it is essential to consider the chloride diffusion in a two-dimensional series model, which means the chloride ion diffusion can take place in any direction. Taking the random distribution of RCA into account, a two-dimensional mesoscopic structure RAC model of five phases is constructed herein. The finite-element mesh used in the simulation model of RAC with a volume fraction of RCA of 60% is displayed in Figure 2.

Effective diffusion coefficients of chlorides in RAC

The present paper considers only the diffusion process in RAC, and this process can be simplified into a single-component transport solely involving chlorides. In a two-dimensional RAC series model, the diffusion of chloride ions taking place in any direction through the cement paste areas in the RAC can be governed by Fick's second law, as follows

$$1. \quad \frac{\partial C}{\partial t} = D_k \nabla^2 C \quad (k = 1, 2, 3, 4, 5)$$

where C is the concentration of chloride ions in the cement paste matrix (moles per unit volume of paste bulk), particularly in the new mortar and the adherent old mortar; t is the time; ∇ is the Laplace differential operator; and D_k is the chloride diffusivity of each phase, where k stands for the k th phase ($k = 1, 2, 3, 4, 5$ represents the new mortar, new ITZ, adherent old mortar, old ITZ and original NCA, respectively).

In order to simplify the simulation process, initial conditions are assumed as follows

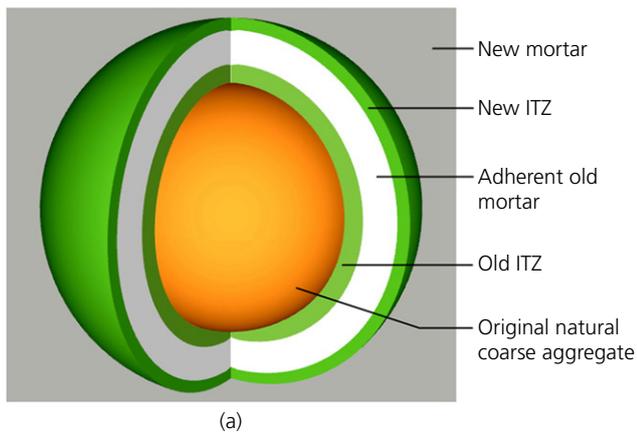
$$2. \quad C(0, x, y) = 0$$

The boundary conditions are given as

$$3. \quad C(t, 0, y) = C_1, \quad C(t, l, y) = 0$$

$$4. \quad \frac{\partial C(t, x, 0)}{\partial y} = 0, \quad \frac{\partial C(t, x, h)}{\partial y} = 0$$

where C_1 is the chloride concentration at the line $x=0$. The length (l) and height of the plain RAC (h) utilised in the numerical model are $l=50$ mm and $h=50$ mm, respectively. Using the given values of D_k and C_1 , the chloride concentration distribution profile at any time can be solved by Equation 1.



(a)



(b)

Figure 1. (a) Partial schematic representation of five-phase composite sphere model of RAC. (b) Chloride diffusion in two-dimensional series model of RAC (arrows represent the chloride diffusion directions; chloride diffused in the cross-section normal to the axis of cylinders)

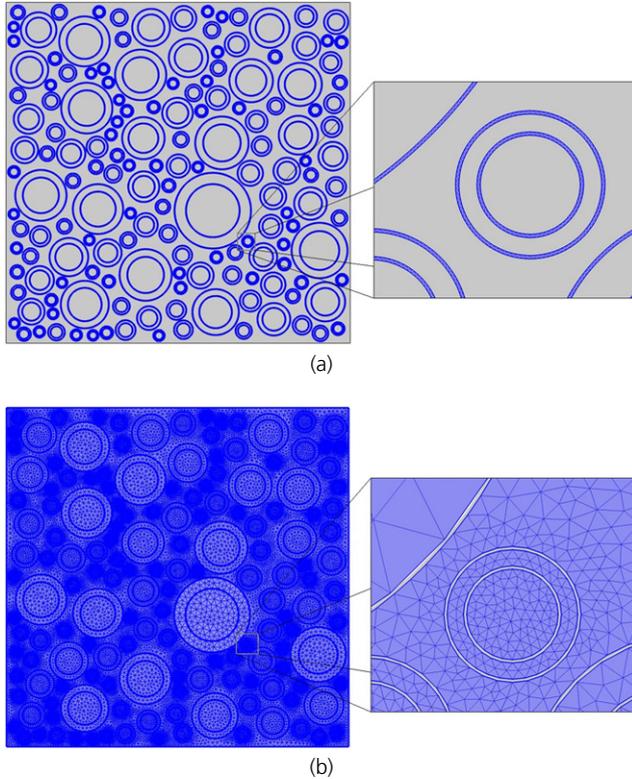


Figure 2. Finite-element mesh of the five-phase RAC model ($V_{RAC} = 0.6$, $R_{aom} = 0.3$, $T_{ITZ} = 40 \mu\text{m}$): (a) mesh of new and old ITZs; (b) mesh of ITZs free

Note that J_x is the x -component of the chloride diffusion flux at the steady state. The total chloride flux, J_t , along the right boundary $x = l$ at the steady state is given by

$$5. \quad J_t = \int_0^h J_x(\infty, l, y) dy = -D_k \int_0^h \frac{\partial C(\infty, l, y)}{\partial x} dy$$

Thus, the average flux at the right boundary $x = l$ is given by

$$6. \quad J_{x=l} = \frac{J_t}{h}$$

By taking the plain RAC as a representative elementary volume in a macroscopic structure model, the average flux, $J_{x=l}$, can also be expressed as

$$7. \quad J_{x=l} = -D_{\text{eff}} \frac{\partial C}{\partial x} = \frac{D_{\text{eff}} C_1}{l}$$

Substituting Equations 5 and 7 into Equation 6 yields

$$8. \quad D_{\text{eff}} = -\frac{D_k l}{C_1 h} \int_0^h \frac{\partial C(\infty, l, y)}{\partial x} dy$$

D_{eff} is the effective diffusion coefficient of chlorides in RAC and $D_c = D_{\text{eff}}$. If the chloride flux in the cement paste matrix is calculated by Equation 8 from the present two-dimensional mesoscopic structure model, D_{eff} of the chlorides in RAC can be evaluated.

Simulation results and discussion

Overview

From the above discussion, the volume fraction of RCA, the proportion and diffusivity of the adherent old mortar, and the thickness and diffusivity of the old and new ITZ are the main parameters that have a great impact on the chloride diffusivity of RAC. The geometric parameters taken in this study are given in Table 1. In this study, the volume fraction of RCA (V_{RCA}) refers to the volume of RCA used to prepare the RAC, which varies from 10 to 70%. The adhesive ratio of old mortar (R_{aom}) is defined as the ratio of the thickness of the adherent old mortar to the radius of each aggregate particle, which could be assigned to every RCA particle. For the purpose of simplicity, R_{aom} is approximately equal to 45% volume fraction rate of old mortar and RCA.

For the chloride diffusion coefficient in concrete, there is another significant element – aggregates – which also have a great impact on the diffusivity of chlorides (Hobbs, 1999). The chloride diffusivity of RCA is mainly determined by its constituent phases, especially the adherent old mortar and the old ITZ. In NAC, it is reasonable to assume that the aggregate is relatively impermeable compared with the highly permeable bulk paste. On the contrary, in RAC, the RCA is no longer assumed as impermeable due to its adherent old mortar and old ITZ, whereas the original NCA in the core area is considered as impermeable.

In order to accurately evaluate the effective diffusion coefficient of chlorides in RAC, the normalised diffusion coefficient from the diffusion model performed by Fick's laws is employed. The normalised chloride diffusion coefficient is calculated from the ratio of chloride diffusion coefficient of RAC (D_{RAC}) to that of the new mortar ($D_{\text{new,mor}}$)

Table 1. Geometric parameters

Volume fraction of RAC, V_{RCA} : %	Adhesive ratio of old mortar, R_{aom}	Thickness of ITZ, T_{ITZ} : μm	
		$T_{\text{new,ITZ}}$	$T_{\text{old,ITZ}}$
10	0.0	20	20
20			
30			
40	0.1	40	40
50			
60			
60	0.3	60	60
70			
	0.4		

(Liu *et al.*, 2017a). It should be noted that few precise investigations of the chloride diffusivity of adherent old mortar and the old ITZ related to RCA have been put forward at present. According to the cited literature, the ratio of chloride diffusion coefficient between adherent old mortar and new mortar ($M_1 = D_{old,mor}/D_{new,mor}$) varies in the range of 0.2–5 times (Xiao *et al.*, 2012, 2013); the ratio of chloride diffusion coefficient between old (new) ITZ and old (new) mortar ($M_2 = D_{old,ITZ}/D_{old,mor}$ and $D_{new,ITZ}/D_{new,mor}$) ranges from 1.3 to 16.2 times (Xiao *et al.*, 2012); the average thickness of old and new ITZ (T_{ITZ} , both $T_{old,ITZ}$ and $T_{new,ITZ}$) adopted in the literature varies in the range of 20–80 μm (Diamond and Huang, 2001; Gao *et al.*, 2005; Liu *et al.*, 2015b; Tam and Tam, 2007a; Zheng *et al.*, 2009). Similarly to what is found for NAC, the chloride diffusion coefficient in cement paste is well correlated to the ratio of water and cement (Atkinson and Nickerson, 1984; Díaz *et al.*, 2006; Xia and Li, 2013). When the water-to-cement ratio in old mortar is lower compared to new mortar, the chloride penetration resistance of the old mortar will be superior to that of the new mortar (Bo *et al.*, 2009; Kou and Poon, 2015). To evaluate the effective diffusion coefficient of chlorides in RAC, the initial values of these parameters are $R_{aom} = 0.3$, $T_{ITZ} = T_{new,ITZ} = T_{old,ITZ} = 40 \mu\text{m}$, $D_{new,mor} = 2 \times 10^{-6} \text{ mm}^2/\text{s}$ (Xiao *et al.*, 2013), $M_1 = D_{old,mor}/D_{new,mor} = 2$, $M_2 = D_{old,ITZ}/D_{old,mor} = D_{new,ITZ}/D_{new,mor} = 10$. When one of the parameters is under consideration, the others remain fixed or change within the scope of given values. From the foregoing, the chloride diffusion coefficients of each phase in RAC can be approximately set. The chloride diffusivity of each phase in RAC can be proved by the different ratios taken here. Then, by using a combination of the finite-element method and the partial differential Equation 8, with boundary and initial conditions established by Equations 2–4, the normalised chloride diffusion coefficient of RAC can be derived.

Parameter analysis

Effect of volume fraction of RCA

To investigate the influence of V_{RCA} on the normalised chloride diffusion coefficient of RAC for a given R_{aom} , the values of R_{aom} to be used are 0.4, 0.3, 0.2, 0.1 and 0.0. Figure 3 shows the normalised chloride diffusion coefficients of RAC plotted against different R_{aom} values, with the given values of V_{RCA} . As expected, the normalised chloride diffusion coefficient of every given V_{RCA} increases with the thickening of the adherent old mortar. The results herein are in agreement with the analytical results by Xiao *et al.* (2013). Moreover, it is interesting to notice from Figure 3 that, when R_{aom} varies from 0 to 0.1, the chloride diffusivity of the concrete increases more rapidly than during later periods. The reason for this rapid growth is mainly that the concrete changes from NAC to RAC when R_{aom} is varied from 0 to 0.1, and the appearance of the adherent old mortar and old ITZ will intensify the diffusivity of the concrete. In particular, when V_{RCA} increases

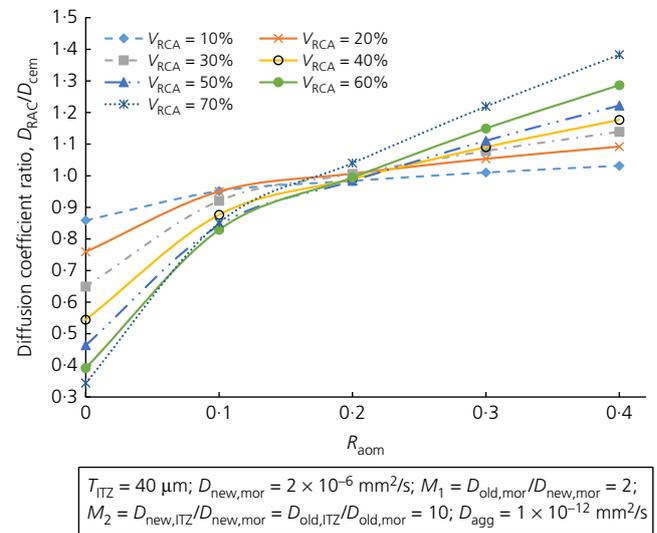


Figure 3. Effect of the volume fraction of RCA on chloride diffusivity of RAC

from 10 to 70%, the diffusion coefficient ratio increases by 20.1, 43.8, 75.4, 116.2, 163.4, 228.4 and 302.0% for R_{aom} from 0.0 to 0.4. One of the main reasons for this increase is that the content of adherent old mortar is increased with the increase of R_{aom} and V_{RCA} , which will distinctly increase the diffusivity of RAC. It has been proved that the adherent old mortar is usually more permeable than new mortar. This phenomenon definitely indicates that the content of adherent old mortar has a significant impact on the diffusivity of RAC. It also implies that it would be better to reduce the old mortar adhered to the surface of RCA for the purpose of making better use of RCA in practice.

Effect of adhesive ratio of old mortar

To investigate the influence of R_{aom} on the normalised chloride diffusion coefficient of RAC for a given V_{RCA} , the values of V_{RCA} are 10, 20, 30, 40, 50, 60 and 70%. When the R_{aom} tends to zero, this means that there is no adherent old mortar remaining on the surface of the aggregates, and the RAC will become NAC in this case. The normalised chloride diffusion coefficients of RAC plotted against different V_{RCA} with the given values of R_{aom} are shown in Figure 4. According to Figure 4, when R_{aom} is 0.1 and 0.2, it is evident that a similar fluctuation within a small range of diffusion coefficient ratios begins to happen. This slight fluctuation can be explained by the fact that, as a five-phase composite sphere model, the content of relatively highly permeable adherent old mortar and the amount of assumed impermeable original NCA have opposite effects on the chloride diffusivity property of the RAC. When the effect of the impermeable original NCA dominates, the normalised chloride diffusion coefficient of RAC will decrease (Caré, 2003). If V_{RCA} is small and R_{aom} is also lower,

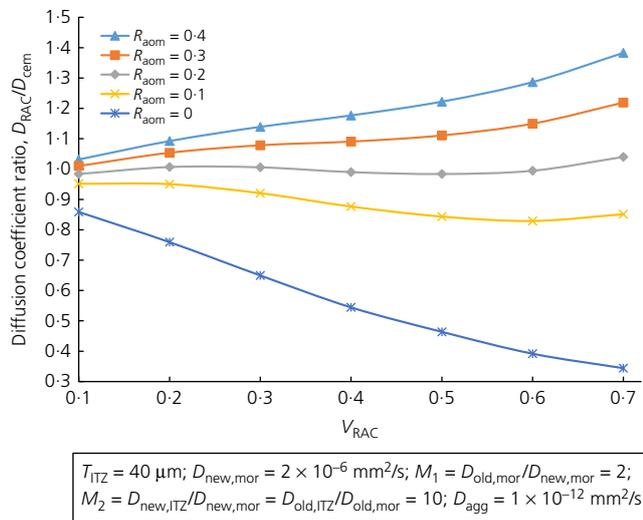


Figure 4. Effect of the adhesive ratio of old mortar on chloride diffusivity of RAC

this means there is just a flimsy shell of adherent old mortar remaining on every original NCA. Increasing V_{RCA} with lower R_{aom} may decrease the diffusion coefficient of the chloride ratio, as a result of increasing the impermeable original NCA. Conversely, when the effect of the adherent old mortar dominates, the normalised chloride diffusion coefficient of RAC will increase. Consequently, when the V_{RCA} is adequate in quantity from 50 to 70% with the given R_{aom} values of 0.1 and 0.2, the total amount of the flimsy shell of adherent old mortar dominates to increase the diffusivity of the RAC. Interestingly, when R_{aom} is 0.3 and 0.4, it can be seen from Figure 4 that the increase in diffusion coefficient ratio with the increase of V_{RCA} is noticeable and consistent. This tendency can be explained by the fact that when the adherent old mortar, which is more permeable than natural aggregates, takes up a certain part of every particle, its effect will predominate over the effect of the original NCA. When the R_{aom} of RCA is zero, this means that the adherent old mortar has vanished, and the chloride diffusivity of the RAC tends to be that of NAC. This implies that reducing R_{aom} to a great extent can

decrease the diffusivity of RAC and increase the chloride penetration resistance of RAC.

Effect of chloride diffusivity of adherent old mortar

To further demonstrate the variation of adherent old mortar with the normalised chloride diffusion coefficient of RAC, the influence of V_{RCA} on the normalised chloride diffusion coefficient of RAC with a given M_1 and T_{ITZ} is explained. The values of M_1 are 0.2, 2 and 10, set from evaluation in the literature using analytical methods (Xiao *et al.*, 2013). This means that the diffusivity of adherent old mortar tends to be increasingly inferior to new mortar. The geometric parameters and chloride diffusivity of each phase in this section are given in Table 2. The thicknesses of both old and new ITZs are 40 μm in Figure 5(a) and 20 μm in Figure 5(b). Figures 5(a) and 5(b) show that when M_1 is 0.2 and T_{ITZ} is a certain constant, the diffusivity of RAC absolutely decreases with the increase of V_{RCA} , which shows a trend consistent with that of NAC. The gap between RAC ($M_1=0.2$, $T_{ITZ}=40 \mu m$ or 20 μm) and NAC ($T_{ITZ}=40 \mu m$ or 20 μm) is small. The reason for this behaviour is that, when M_1 is 0.2, this means that the chloride penetration resistance of old mortar is superior to that of new mortar, and as a result RCA as a whole can be treated as a kind of relatively low-permeability NCA. As expected, when M_1 is 10, the diffusion coefficient ratio increases with the increase in V_{RCA} due to the increasing content of highly permeable adherent old mortar. Intriguingly, when $M_1=2.0$ and $T_{ITZ}=20 \mu m$, the diffusivity of adherent old mortar is also inferior to that of new mortar, whereas the diffusivity of RAC remains at a stable level around 1.0. This implies that the diffusivity of RAC in this kind of property ($M_1=0.2$, $M_2=10$, $T_{ITZ}=20 \mu m$) can be equivalent to cement paste without aggregate, which can be explained by the effects of both the adherent old mortar and the original NCAs. As shown in Figure 5(c), it can be observed that the tendency of the two cases is completely consistent. The gap between $T_{ITZ}=40 \mu m$ and $T_{ITZ}=20 \mu m$ with the same M_1 increases with the increase of V_{RCA} . A thicker ITZ will more effectively increase the diffusivity of RAC; the curves of $T_{ITZ}=20 \mu m$ always remain below those of $T_{ITZ}=40 \mu m$. It can be speculated that both the thickness and the diffusivity of ITZ play an

Table 2. Geometric parameters and chloride diffusivity of each phase in section entitled 'Effect of chloride diffusivity of adherent old mortar'

$T_{new,ITZ}$ μm	$T_{old,ITZ}$ μm	M_1	M_2	$D_{new,mor}$ $10^{-6} mm^2/s$	$D_{old,mor}$ $10^{-6} mm^2/s$	$D_{new,ITZ}$ $10^{-6} mm^2/s$	$D_{old,ITZ}$ $10^{-6} mm^2/s$	D_{agg} $10^{-6} mm^2/s$
					$M_1 \times D_{new,mor}$	$M_2 \times D_{new,mor}$	$M_2 \times D_{old,mor}$	
40	40	0.2	10	2	0.4	20	4	10^{-6}
40	40	2	10	2	4	20	40	10^{-6}
40	40	10	10	2	20	20	200	10^{-6}
20	20	0.2	10	2	0.4	20	4	10^{-6}
20	20	2	10	2	4	20	40	10^{-6}
20	20	10	10	2	20	20	200	10^{-6}

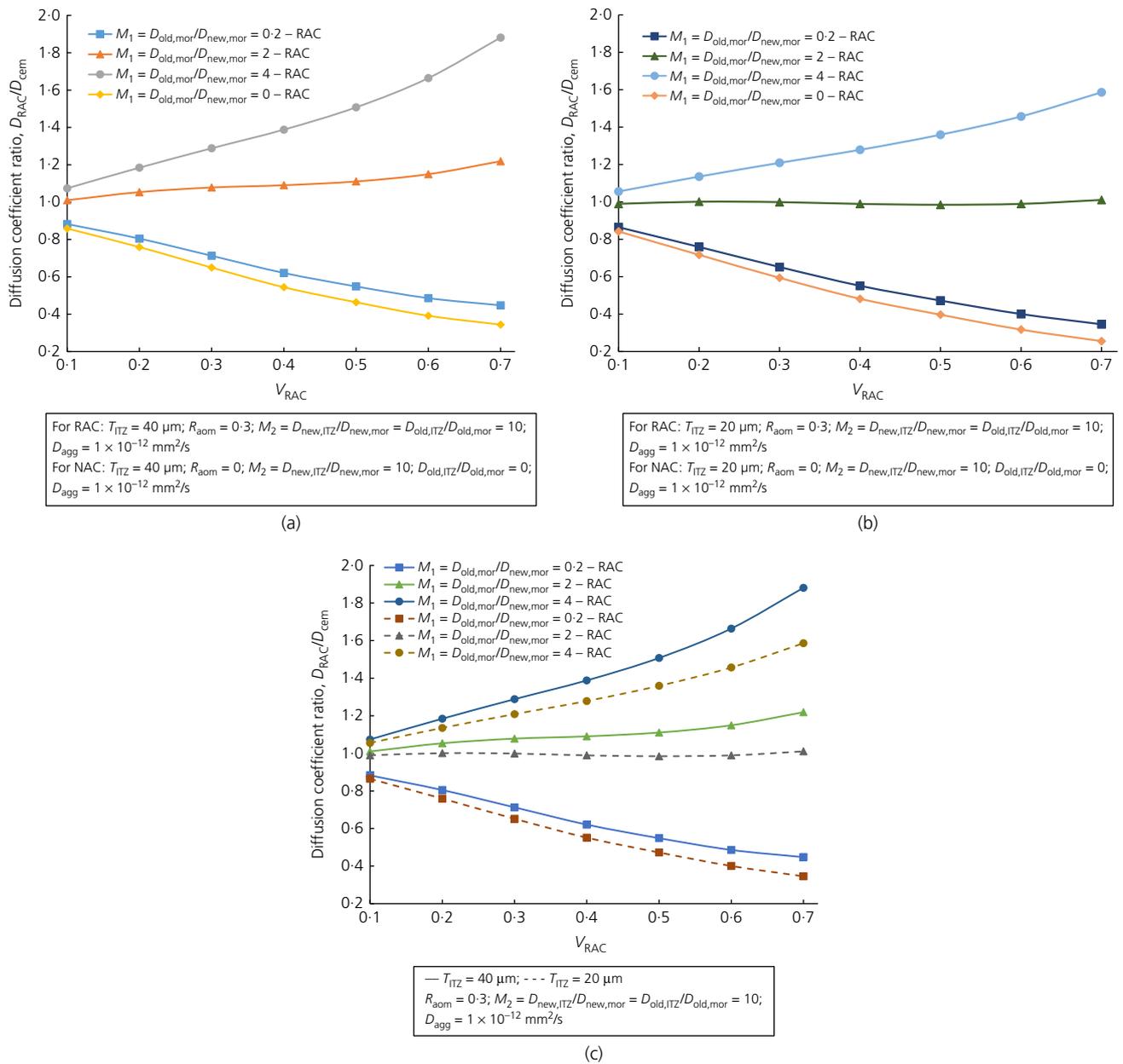


Figure 5. Effect of chloride diffusivity of adherent old mortar on chloride diffusivity of RAC: (a) $T_{ITZ} = 40 \mu m$; (b) $T_{ITZ} = 20 \mu m$; (c) combined figure: $T_{ITZ} = 40 \mu m$ and $T_{ITZ} = 20 \mu m$

important role in the diffusivity of RAC, which should be discussed. This also implies that the higher the quality of the adherent old mortar on RCA, the better the chloride penetration resistance of RAC. Thus, the quality and the source of RCA should be considered carefully and improved in practice.

Effect of ITZ thickness

To investigate the influence of V_{RCA} on the normalised chloride diffusion coefficient of RAC for a given T_{ITZ} value (both old ITZ and new ITZ), the values of T_{ITZ} were 20,

40 and 60 μm . The normalised chloride diffusion coefficients of RAC plotted against the different V_{RCA} with the given values of ITZ thickness are shown in Figure 6. From the figure, it is apparent that the normalised chloride diffusion coefficients of RAC are almost stable when the T_{ITZ} is 20 μm , but increase with V_{RCA} when the T_{ITZ} is 40 or 60 μm . When the T_{ITZ} is 20 μm , the effect of T_{ITZ} is too small to change the diffusivity of RAC, and the slight undulation of data is mainly caused by the randomness of RCA distribution in the RAC numerical simulation model. From this, it can be inferred that the increasing thickness of both new and old ITZ will have a

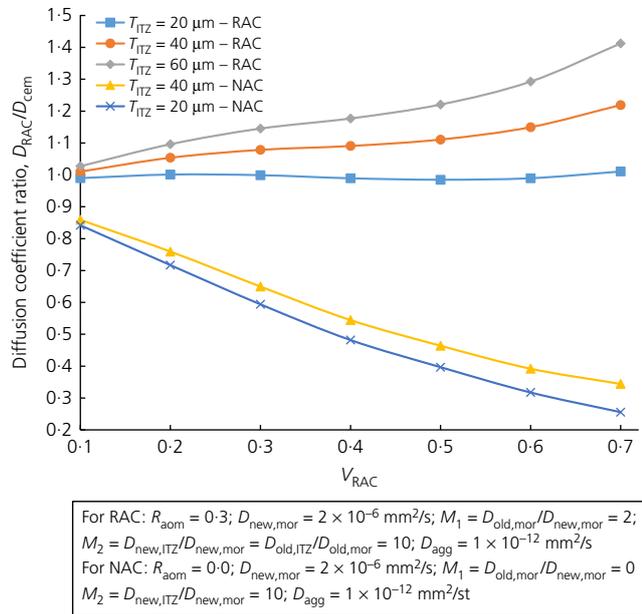


Figure 6. Effect of ITZ thickness on chloride diffusivity of RAC

more noticeable influence on the chloride diffusivity of RAC due to its naturally poor properties, and the greater the ITZ thickness, the more the content of the ITZ fraction. The gap between the curve of $T_{ITZ} = 20 \mu m$ and the curve of $T_{ITZ} = 40 \mu m$, and between the curve of $T_{ITZ} = 40 \mu m$ and the curve of $T_{ITZ} = 60 \mu m$ also increases with the increasing V_{RCA} as a result of the increasing volume fraction of ITZ and adherent old mortar. The chloride diffusivities of ITZs and adherent old mortar are usually higher than those of new mortar. These phenomena indicate that both the thickness and the properties of old and new ITZ have a significant impact on the normalised chloride diffusion coefficient of RAC.

Effect of chloride diffusivity of ITZ

In order to further examine the effect of the variable diffusivity of the ITZ on the normalised chloride diffusion coefficient of RAC, the influence of V_{RCA} on the normalised chloride diffusion coefficient of RAC with a given M_2 ($M_2 = D_{old,ITZ}/D_{old,mor}$ and $D_{new,ITZ}/D_{new,mor}$) and T_{ITZ} is examined herein, shown as Figure 7. The geometric parameters and chloride diffusivity of each phase in this section are given in Table 3. The values of M_2 are 2, 6 and 10, adopted from the literature, which means that the diffusivity of the old (new) ITZ tends to be even more inferior to that of the old (new) mortar. It can be seen from Figure 7(a) ($T_{ITZ} = 40 \mu m$) that when $M_2 = 6$, the diffusion coefficient ratio remains at a stable value around 1.0, in the middle of the three curves, whereas, with the increase of V_{RAC} , the curve of $M_2 = 10$ maintains an upwards trend and the curve of $M_2 = 2$ exhibits a downwards tendency. Furthermore, interestingly, it can also be seen from Figure 7(b) ($T_{ITZ} = 20 \mu m$) that when $M_2 = 10$, the diffusion coefficient

ratio remains at a stable value around 1.0 and the curves of both $M_2 = 6$ and $M_2 = 2$ are below it, and also both keep a slow downward trend with the increase of V_{RCA} . From Figure 7(c), when $M_2 = 2$, there is a consistent tendency with the thickness of ITZ 40 μm and 20 μm for the diffusivity of RAC to decrease with the increase of V_{RCA} . Specifically, the curve of $M_2 = 6$ and $T_{ITZ} = 40 \mu m$ follows the same trends as the curve of $M_2 = 10$ and $T_{ITZ} = 20 \mu m$, and the results are very close to each other in these two cases. This indicates that the thickness of ITZ and the diffusivity of ITZ play the same important role and both have significant effects on the chloride diffusivity of RAC.

Conclusions

In this paper, RAC is considered as an inhomogeneous composite material with the random distribution of RCA in a two-dimensional simulation. A five-phase composite numerical model, which includes three continuous phases (new mortar, adherent old mortar and original NCA) and two interphases (new ITZ and old ITZ), is proposed to predict the effective diffusion coefficient of chlorides in RAC. Considering the properties of each phase and their individual impacts on the effective diffusion coefficient of chlorides in RAC, the critical factors including the volume fraction of RCA, the adhesive ratio of old mortar, the chloride diffusivity of adherent old mortar, the thickness of the old and new ITZ and the chloride diffusivity of old and new ITZ were investigated by quantitative evaluation. Based on the numerical simulation model of RAC combined with parametric research, the following observations can be made.

- The effective diffusion coefficient of chloride in RAC increases with the increase of the volume fraction of RCA. However, when the chloride penetration resistance of adherent old mortar is superior to that of new mortar, the chloride diffusivity of RAC decreases with the increase of the volume fraction of RCA, which is similar to the NAC. The higher the quality of RCA, the better the chloride penetration resistance of the RAC. Note that the quality and the sources of RCA should be considered and improved.
- The effective diffusion coefficient of chloride in RAC increases with the increase in the adhesive ratio of old mortar, whereas this relationship will be volatile when the adhesive ratio of the old mortar is lower. For better utilisation of RAC in practice, it should be mentioned that even a flimsy shell of old mortar adhered onto every piece of RCA can cause dramatic variations in the chloride diffusivity of RAC.
- The effective diffusion coefficient of chloride in RAC tends to be that of NAC when the adhesive ratio of the old mortar is zero, which means that the adherent old mortar has vanished. To make better use of RCA, it is suggested that the adherent old mortar should be decreased and at

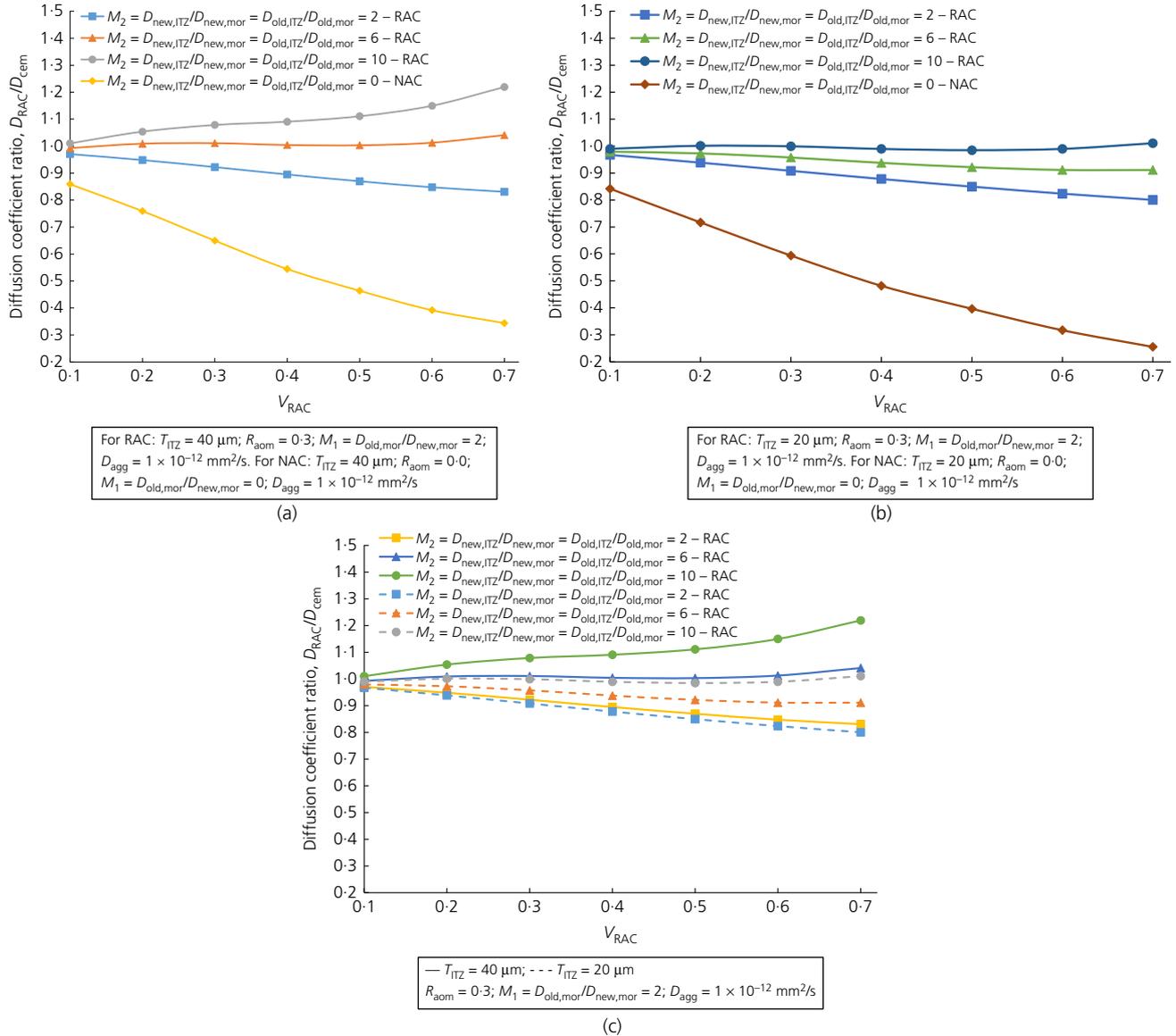


Figure 7. Effect of chloride diffusivity of ITZ on chloride diffusivity of RAC: (a) $T_{ITZ} = 40 \mu m$; (b) $T_{ITZ} = 20 \mu m$; (c) combined figure: $T_{ITZ} = 40 \mu m$ and $T_{ITZ} = 20 \mu m$

Table 3. Geometric parameters and chloride diffusivity of each phase in section entitled 'Effect of chloride diffusivity of ITZ'

$T_{new,ITZ}$ μm	$T_{old,ITZ}$ μm	M_1	M_2	$D_{new,mor}$ $10^{-6} \text{ mm}^2/\text{s}$	$D_{old,mor}$	$D_{new,ITZ}$	$D_{old,ITZ}$	D_{agg} $10^{-6} \text{ mm}^2/\text{s}$	
					$10^{-6} \text{ mm}^2/\text{s}$	$10^{-6} \text{ mm}^2/\text{s}$	$10^{-6} \text{ mm}^2/\text{s}$		
					$M_1 \times D_{new,mor}$	$M_2 \times D_{new,mor}$	$M_2 \times D_{old,mor}$		
40	40	2	2	2	4	4	8	10^{-6}	
40	40	2	6	2	4	12	24	10^{-6}	
40	40	2	10	2	4	20	40	10^{-6}	
20	20	2	2	2	4	4	8	10^{-6}	
20	20	2	6	2	4	12	24	10^{-6}	
20	20	2	10	2	4	20	40	10^{-6}	

the same time the chloride penetration resistance of the adherent old mortar should also be improved.

- The effective diffusion coefficient of chloride in RAC increases with the increase of the thickness and the diffusivity of the ITZ. As the thickness of the ITZ increases, the effect of ITZ thickness on the diffusivity of RAC increases. For the same thickness of the old ITZ, there will be better chloride penetration resistance of the old ITZ, and an improved chloride penetration resistance in the RAC.

Acknowledgements

The work was supported by the Natural Science Foundation of China (51508324), the Shanghai Pujiang Program, China (15PJ1403800) and the Shanghai Chenguang Program, China (16CG06). The third author would also like to thank the Natural Science Foundation of China (51408537) for its support. The seventh author would like to gratefully acknowledge the Open Research Fund of the State Key Laboratory of Simulation and Regulation of Water Cycle in River Basin, China Institute of Water Resources and Hydropower Research (IWHR-SKL-201705).

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