A two-step approach for calculating chloride diffusion coefficient in concrete with both natural and recycled concrete aggregates

Wei-bin Yuan [a], Lixuan Mao [b,*], Long-yuan Li [b]

[a] College of Architecture and Civil Engineering, Zhejiang University of Technology, Hangzhou 310023, China
[b] School of Engineering, Computing and Mathematics, University of Plymouth, Plymouth, Devon PL4 8AA, UK

HIGHLIGHTS

- Analytical model is derived for predicting the diffusivity of recycled aggregate concrete.
- The model is validated using both experimental and numerical simulation results.
- The diffusivity of recycled concrete aggregate is modelled using spherical model.
- The diffusivity of three-phase concrete is modelled using effective medium approximation.
- The effect of natural aggregate replacement rate on concrete diffusivity is examined.

GRAPHICAL ABSTRACT
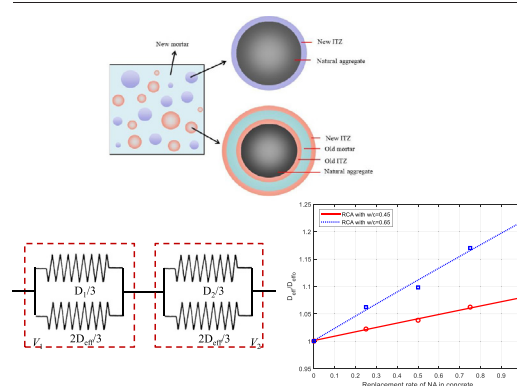
ABSTRACT

This paper presents an analytical approach to calculate the effective diffusion coefficient of chlorides in concrete with both natural and recycled concrete aggregates. In the approach the concrete is treated as a composite consisting of three phases, namely mortar, natural aggregate plus interfacial transition zone, and recycled concrete aggregate plus interfacial transition zone. The effective diffusion coefficient of chlorides in the composite is calculated through two steps. The first step is to calculate the effective diffusion coefficients of chlorides in the natural aggregate plus interfacial transition zone and in the recycled concrete aggregate plus interfacial transition zone by using multilayer spherical approximation, the results of which provide the information about the quality of recycled concrete aggregate in terms of chloride penetration resistance. The second step is to calculate the effective diffusion coefficient of chlorides in the three-phase concrete composite by using effective medium approximation, the results of which provide the information about the influence of recycled concrete aggregate on the diffusivity of recycled aggregate concrete. The analytical expression of the effective diffusion coefficient is derived and carefully compared with the results obtained from both the experiments and numerical simulations, which demonstrates that the present analytical model is rational and reliable. The analytical expression presented can be used to predict the service life of recycled aggregate concrete exposed to chloride environment.

1. Introduction

The construction industry produces a huge amount of waste that consists of unwanted materials created by construction and demolition industries. The construction waste increases the burden on landfill sites and has great impact on environment since the hazardous substances in the

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* Corresponding author.
   E-mail address: lixuan.mao@plymouth.ac.uk (L. Mao).

Notations

D_1	diffusion coefficient of chlorides in phase 1
D_2	diffusion coefficient of chlorides in phase 2
D_{eff}	effective diffusion coefficient of chlorides in concrete or two-phase composite
D_{effo}	effective diffusion coefficient of chlorides in concrete with 100 % NA
D_M	diffusion coefficient of chlorides in new mortar
D_{NA}	diffusion coefficient of chlorides in natural aggregate
D_{NA+}	effective diffusion coefficient of chlorides in natural aggregate plus interfacial transition zone
D_{nitz1}	diffusion coefficient of chlorides in new interfacial transition zone in the natural aggregate plus interfacial transition zone
D_{nitz2}	diffusion coefficient of chlorides in new interfacial transition zone in the recycled concrete aggregate plus interfacial transition zone
D_{oa}	diffusion coefficient of chlorides in old natural aggregate
D_{oa+}	effective diffusion coefficient of chlorides in the old natural aggregate plus interfacial transition zone
D_{oitz}	diffusion coefficient of chlorides in old interfacial transition zone in recycled concrete aggregate
D_{om}	diffusion coefficient of chlorides in old mortar in recycled concrete aggregate
D_{RCA}	effective diffusion coefficient of chlorides in recycled concrete aggregate
D_{RCA+}	effective diffusion coefficient of chlorides in the recycled concrete aggregate plus interfacial transition zone
EMA	effective medium approximation
FA	fly ash
h_{itz}	average thickness of interfacial transition zone
ITZ	interfacial transition zone
k	proportional constant
k_{itz}	proportional constant
MLSA	multilayer spherical approximation
NA	natural aggregate
NA+	natural aggregate plus interfacial transition zone
NAC	natural aggregate concrete
OPC	ordinary Portland cement
R_{NA}	average radius of natural aggregate or recycled concrete aggregate
RA	recycled aggregate
RAC	recycled aggregate concrete
RCA	recycled concrete aggregate
RCA+	recycled concrete aggregate plus interfacial transition zone
RCM	rapid chloride migration
RCP	rapid chloride permeability
RMA	recycled masonry aggregate
V_1	volume fraction of phase 1 in two-phase composite
V_2	volume fraction of phase 2 in two-phase composite
V_M	volume fraction of new mortar in concrete
V_{NA}	volume fraction of natural aggregate in the natural aggregate plus interfacial transition zone
V_{NA+}	volume fraction of the natural aggregate plus interfacial transition zone in concrete
V_{nitz1}	volume fraction of new interfacial transition zone in the natural aggregate plus interfacial transition zone
V_{nitz2}	volume fraction of new interfacial transition zone in the recycled concrete aggregate plus interfacial transition zone
V_{oa}	volume fraction of old natural aggregate in recycled concrete aggregate
V_{oitz}	volume fraction of old interfacial transition zone in recycled concrete aggregate
V_{om}	volume fraction of old mortar in recycled concrete aggregate
V_{RCA}	volume fraction of recycled concrete aggregate in the recycled concrete aggregate plus interfacial transition zone
V_{RCA+}	volume fraction of the ecycled concrete aggregate plus interfacial transition zone in concrete
w/b	water-to-binder ratio
w/c	water-to-cement ratio

waste can cause soil and water pollution. Therefore, it is extremely important to reduce, reuse and recycle the construction waste before disposal, which not only can conserve the planet's natural resources but also minimise the damage to environment. One of the activities in reducing construction and industrial waste is the recycling of waste materials, which can be used as the part of concrete materials (Vázquez et al., 2021; Adhikary and Ashish, 2022). The typical examples include the use of industrial waste to produce supplementary cementitious materials and geopolymer binders (Liu et al., 2022; He et al., 2022; Shehata et al., 2022), and the use of recycled aggregate (RA) to replace natural aggregate (NA) in concrete (Ma et al., 2022; Khushnood et al., 2020). The RA could be derived from crushed concrete, known as recycled concrete aggregate (RCA), or from masonry rubble, known as recycled masonry aggregate (RMA). To use RCA and/or RMA to partially or fully replace NA in concrete is not a new concept (Dhir and Lye, 2019). However, its practice is still not widely accepted in construction industry. The main reason for this is the variability and uncertainty in the quality and properties of the recycled concrete and/ or recycled masonry (Colangelo et al., 2021), which makes it difficult to control the grade of recycled aggregate concrete (RAC). In addition, the RCA or RMA may contain chlorides if the recycled concrete or recycled masonry are taken from marine environment, which may create the durability issue related to chloride-induced reinforcing steel corrosion.

To increase the use of RCA and RMA in concrete, efforts have been made in recent years to improve the quality of RCA and RMA by using various treatments. These include the use of accelerated carbonation (Pu et al., 2021), polymer (Spaeth and Tegguer, 2013), and silica fume slurry (Sasanipour et al., 2021) treatments to improve the quality of the adhered mortar and interfacial transition zone (ITZ) between the mortar and aggregate in the RCA. The results obtained from the tests carried out in laboratories demonstrated that the quality of the RCA can be significantly improved by using chemical or mechanical pre-treatments and the corresponding mechanical properties of the RAC mixed using pre-treated RCA are comparable with those of natural aggregate concrete (NAC) (Wang et al., 2021). Recently, Tam et al. (Tam et al., 2021) presented an excellent review article on the treatment techniques applied to enhance RCA quality and the processes utilized to remove adhered mortar in RCA.

The resistance of concrete to chloride penetration is one of the important durability properties of concrete structures. It has been demonstrated that RAC is more penetrable to chlorides than NAC (Duan and Poon, 2014) because the RCA not only has mortar and ITZ but also the microcracks generated during the crushing process of recycling concrete (Etxeberria et al., 2006; Oikonomou, 2005; Gutiérrez and Juan, 2004). It was reported that the strength improvement of parent concrete could reduce the water absorption of the mortar in RCA and thus increase the resistance of RAC to chloride penetration (Padmini et al., 2009; Pedro et al., 2014; Andreu and Miren, 2014). Similar to that of NAC, the chloride diffusivity of RAC is also affected by the water-to-cement ratio (w/c) used in the mixture of RAC. The higher the w/c ratio used in RAC, the quicker the penetration of chlorides in RAC (Kou, 2006; Qin et al., 2011). Moreover, the impact of the replacement of NA on chloride penetration in RAC was

found to increase with increased w/c ratio used in the mixture of RAC (Amorim Júnior et al., 2019; Bao et al., 2020), indicating that the w/c ratio used in RAC has some effect on the improvement of the diffusivity of the RCA. To quantify the effect of the use of RCA on the chloride penetration in concrete, analytical and numerical studies have been also carried out in recent decades. For example, Ying et al. (2013a) developed an analytical model for calculating the effective diffusion coefficient of chlorides in RAC, in which the concrete was treated as a five-phase composite consisting of mortar, ITZ, old mortar, old ITZ, and old aggregate. The model can be used to assess the effect of RCA on chloride diffusion in RAC. However, since it was developed based on the multilayer spherical approximation (MLSA), the model can be applied only to the concrete with fully replaced RCA. For the concrete with both NA and RCA the model may give inaccurate prediction because of the smear effect between RCA and NA. Damrongwiriyanupap et al. (2011) developed a multi-scale, multiphase and multi-species model to describe ionic transport in RAC, in which the concrete was treated as a multiphase composite and the ionic diffusion equations in different phases were solved numerically by using finite element method. Xiao et al. (2012), Ying et al. (2013b) and Hu et al. (2018) also investigated the chloride diffusion in RAC, in which the concrete was treated as a composite consisting of mortar matrix and randomly distributed NAs and RCAs of different sizes. Both the NAs and RCAs were surrounded by ITZs. The RCA was also treated as a composite consisting of old mortar, old ITZ and old aggregate. The diffusion equation of chlorides in the multiphase composite was solved by using finite element method, from which the effective diffusion coefficient of chlorides in the RAC was obtained. Wu and Xiao (2018) and Peng et al. (2019) investigated numerically the effect of micro-cracks on the chloride diffusion in RAC. The numerical model employed in their studies was an extension of that proposed by Ying et al. (2013b) by incorporating additional micro-cracks of different depths and widths in the RAC. Recently, Yu and Lin (2020) presented a multiphase numerical model to examine the effects of the shape of RCA and the distributions of old mortar and old ITZ in RCA on the chloride diffusivity of the concrete with both NA and RCA. Apart from the analytical and numerical studies, numerous experimental works have been also carried out to quantify how the incorporation of RCA in concrete would affect the chloride penetration resistance of the mixed RAC. For instance,

Adessina et al. (2019) provided an experimental and micromechanical investigation on the mechanical and durability properties of RAC. Otsuki et al. (2003) examined the influence of the use of RCA on the ITZ and chloride penetration in RAC. Villagrán-Zaccardi et al. (2008) studied the chloride penetration and binding behaviours of RAC. Silva et al. (2015) and Neves et al. (2018) reported the statistical analysis and corresponding statistical modelling on the resistance of chloride penetration in RAC based on the rapid chloride permeability (RCP) and rapid chloride migration (RCM) test results for various RAC. Recently, Liang et al. (2021) published a good review article on the chloride transport and chloride-induced steel corrosion in RAC.

The literature review described above shows that there are numerous studies on chloride diffusion in RAC. However, most of them were related to experimental tests or numerical simulations, and very few were on analytical modelling. In this paper, we present an analytical approach to predict the effective diffusion coefficient of chlorides in concrete with both NA and RCA. In the approach the concrete is treated as a composite consisting of three phases, namely mortar, NA plus ITZ (NA+), and RCA plus ITZ (RCA+), and the effective diffusion coefficient of chlorides in the concrete is calculated through two steps. One is to calculate the effective diffusion coefficients of chlorides in NA+ and in RCA+ by using MLSA, the results of which provide the information about the quality of RCA in terms of chloride penetration resistance. The other is to calculate the effective diffusion coefficient of chlorides in the three-phase concrete composite by using the effective medium approximation (EMA), the results of which provide the information about the influence of RCA on the diffusivity of RAC. The analytical formulations developed can be used to determine the NA replacement ratio in RAC in terms of its chloride penetration resistance and predict the service life of RAC exposed to chloride environment.

2. Three-phase model of concrete with both NA and RCA

Consider a concrete that is mixed with cement, water, sand, NA, and RCA. The hardened concrete thus can be treated as a three-phase composite, consisting of a mortar matrix, a large number of randomly distributed NA+ and RCA+ particles, as shown in Fig. 1. The penetration of chlorides in the concrete can be generally described by using diffusion equation, in
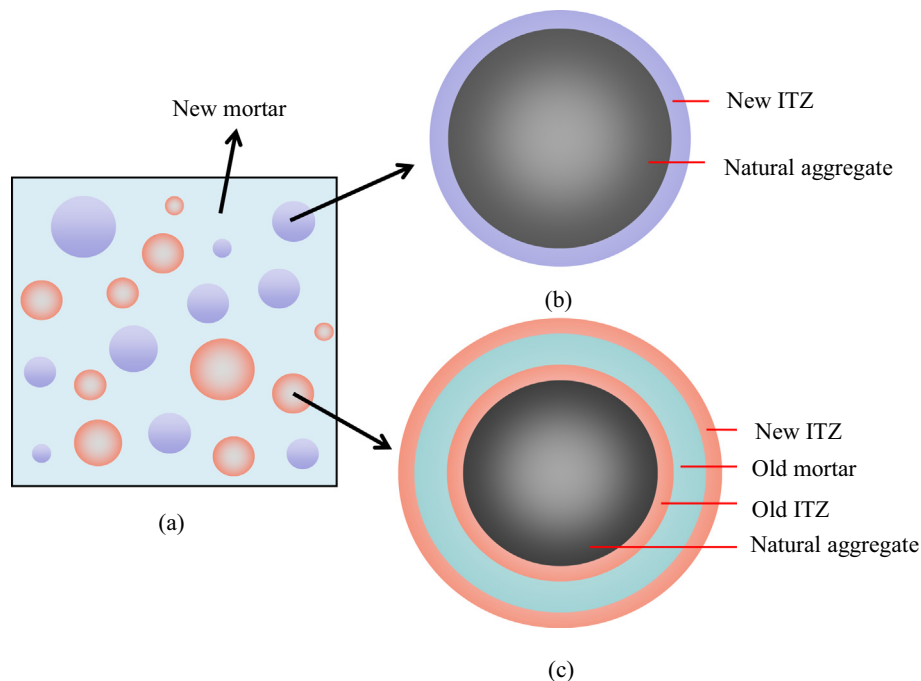

Fig. 1. The schematic representation of (a) concrete with both natural and recycled concrete aggregates, (b) natural aggregate plus interfacial transition zone, and (c) recycled concrete aggregate plus interfacial transition zone.

which the concentration of chlorides in the concrete is mainly controlled by the diffusivity of the concrete. To reflect the influence of individual components on the diffusivity of concrete the diffusion equation usually uses the effective diffusion coefficient. For the concrete with both NA and RCA it is anticipated that the effective diffusion coefficient of chlorides would depend on the volume fractions of mortar, NA+ and RCA+, and the diffusion coefficients of chlorides in the mortar, NA+ and RCA+. Since the NA and RCA are independently distributed in the concrete, they have independent contributions to the diffusivity of the concrete. Thus, the use of MLSA (Ying et al., 2013a) alone may not be suitable for this three-phase composite. In order to reveal the independent feature of NA+ and RCA+ on the diffusivity of concrete, herein, the EMA is utilized. EMA was first proposed by Bruggeman (1935) and Landauer (1952) independently for calculating the effective property of the two-phase composite. The EMA pertains to analytical modelling that describes the macroscopic properties of a composite material by averaging the multiple values of the constituents that directly make up the composite material. There are many different versions of EMA (Tinga et al., 1973), each of which is more or less accurate in distinct conditions. Mathematically, when it is applied to the diffusion problem, EMA can be expressed as follows,

$$\frac{D_{eff} - D_1}{2D_{eff} + D_1} V_1 + \frac{D_{eff} - D_2}{2D_{eff} + D_2} V_2 = 0 \tag{1}$$

where D_{eff} is the effective diffusion coefficient in the composite, D_1 and D_2 are the diffusion coefficients in phases 1 and 2, V_1 and V_2 are the volume fractions of phases 1 and 2, respectively, with $V_1 + V_2 \equiv 1$. Eq. (1) can be rewritten as follows,

$$\frac{V_1}{\frac{1}{3}D_1 + \frac{2}{3}D_{eff}} + \frac{V_2}{\frac{1}{3}D_2 + \frac{2}{3}D_{eff}} = \frac{1}{D_{eff}} \tag{2}$$

Eq. (2) indicates that the phase 1 of volume fraction V_1 and phase 2 of volume fraction V_2 can be considered as the series connection, provided that the diffusion coefficients in the two phases are taken as ($D_1/3$ and $2D_{eff}/3$) and ($D_2/3$ and $2D_{eff}/3$), respectively. This means that the EMA represents a combination of parallel and series models, as shown in Fig. 2. Eq. (2) can be easily extended from a two-phase composite to the three-phase concrete by adding-in one more phase in Eq. (2), as follows,

$$\frac{V_M}{\frac{1}{3}D_M + \frac{2}{3}D_{eff}} + \frac{V_{NA+}}{\frac{1}{3}D_{NA+} + \frac{2}{3}D_{eff}} + \frac{V_{RCA+}}{\frac{1}{3}D_{RCA+} + \frac{2}{3}D_{eff}} = \frac{1}{D_{eff}} \tag{3}$$

where V_M, V_{NA+}, and V_{RCA+} are the volume fractions of mortar, NA+, and RCA+ in the concrete with $V_M + V_{NA+} + V_{RCA+} \equiv 1$, D_M, D_{NA+}, and D_{RCA+} are the effective diffusion coefficients of chlorides in the mortar, NA+, and RCA+, respectively, and D_{eff} is the effective diffusion coefficient

of chlorides in the concrete. Eq. (3) indicates that if the volume fractions of the mortar, NA+ and RCA+ in the concrete and the corresponding effective diffusion coefficients of chlorides in the mortar, NA+ and RCA+ are known then the effective diffusion coefficient of chlorides in the concrete with both NA and RCA can be predicted. Note that one of the conditions in applying the EMA is the volume fraction of zero diffusion coefficient phase that must be <2/3. For the present three-phase concrete, neither NA+ nor RCA+ phase has the volume fraction >2/3. Moreover, since both NA+ and RCA+ phases involve the ITZs the diffusion coefficients in the NA+ and RCA+ phases would not be zero. Also, it should be mentioned herein that, in both the MLSA and EMA the inclusions are assumed to be spherical particles that are uniformly dispersed in the matrix phase of the composite. For concrete, aggregates are not exactly spherical. However, previous numerical simulations (Li et al., 2012) have demonstrated that the shape of aggregates has little effect on the overall diffusivity of the concrete. Thus, the use of EMA and MLSA is valid for the present three-phase concrete model.

3. Effective diffusion coefficients of chlorides in NA+ and RCA+

In geometry, the NA+ phase consists of natural aggregate and ITZ surrounding the aggregate. The effective diffusion coefficient of chlorides in the NA+ phase thus can be calculated by using the two-layer spherical approach (Caré and Hervé, 2004), as follows,

$$D_{NA+} = \frac{(3 - 2V_{Nitz1})D_{NA} + 2V_{Nitz1}D_{Nitz1}}{V_{Nitz1}D_{NA} + (3 - V_{Nitz1})D_{Nitz1}} D_{Nitz1} \tag{4}$$

where D_{NA} is the diffusion coefficient of chlorides in NA, D_{Nitz1} is the diffusion coefficient of chlorides in new ITZ surrounding NA, V_{Nitz1} is the volume fraction of new ITZ in the NA+ phase, V_{NA} is the volume fraction of NA in the NA+ phase, and $V_{Nitz1} + V_{NA} \equiv 1$. Note that the NA can be generally considered to be impermeable and thus $D_{NA} = 0$ can be assumed. Also, since the order of magnitude of ITZ thickness is much smaller than that of the radius of aggregate, the following approximation can generally be taken,

$$V_{Nitz1} = \frac{(R_{NA} + h_{itz})^3 - R_{NA}^3}{(R_{NA} + h_{itz})^3} \approx \frac{3h_{itz}}{R_{NA}} \ll 0.024 \tag{5}$$

where $R_{NA} \approx 5$ mm is the assumed average radius of NA and $h_{itz} \approx 40$ μm is the average thickness of ITZ in NA+ phase, which was based on the results obtained from scanning electron microscopy analysis (Hu et al., 2018; Yu and Lin, 2020). Note that the values of R_{NA} and h_{itz} may vary depending
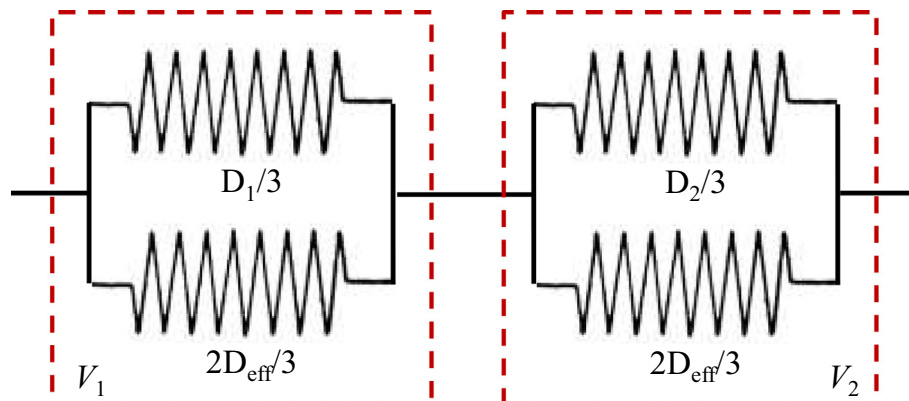

Fig. 2. The schematic representation of effective medium approximation.

on the actual sizes of aggregate and the w/c ratio used in the concrete mixture. Hence, Eq. (4) can be simplified as follows,

$$D_{NA+} = \frac{2V_{Nitz1}}{3 - V_{Nitz1}} D_{nitz1} \approx \frac{2}{3} V_{Nitz1} D_{Nitz1} \qquad (6)$$

Similarly, for the RCA+, we have

$$D_{RCA+} = \frac{(3 - 2V_{Nitz2})D_{RCA} + 2V_{Nitz2}D_{Nitz2}}{V_{Nitz2}D_{RCA} + (3 - V_{Nitz2})D_{Nitz2}} D_{Nitz2} \approx \frac{3D_{RCA} + 2V_{Nitz2}D_{Nitz2}}{V_{Nitz2}D_{RCA} + 3D_{Nitz2}} D_{Nitz2} \qquad (7)$$

where D_{RCA} is the effective diffusion coefficient of chlorides in RCA, D_{Nitz2} is the diffusion coefficient of chlorides in new ITZ surrounding RCA, V_{Nitz2} is the volume fraction of new ITZ in the RCA+ phase, V_{RCA} is the volume fraction of RCA in the RCA+ phase, and $V_{Nitz2} + V_{RCA} \equiv 1$. Note that, unlike the natural aggregate that can be assumed to be impermeable, the RCA is permeable and thus $D_{RCA} \neq 0$.

The RCA can be modelled as a three-phase composite, consisting of old NA, old ITZ, and old mortar (see Fig. 3). However, the configuration of the three phases in RCA may vary significantly from one to another. As an approximation, herein the RCA is treated as a three-layer sphere where the old NA is the inner core of the sphere, the old ITZ is the middle layer of the sphere, and the old mortar is the outer layer of the sphere (Hu et al., 2018; Yu and Lin, 2020). Similar to Eq. (6), by assuming the old NA is impermeable, the effective diffusion coefficient of chlorides in the two-phase material consisting of the old NA and old ITZ can be expressed as follows,

$$D_{oa+} = \frac{2V_{oitz}D_{oitz}}{3(V_{oitz} + V_{oa})} \qquad (8)$$

where D_{oa+} is the effective diffusion coefficient of chlorides in the two-phase material consisting of old NA and old ITZ, D_{oitz} is the diffusion coefficient of chlorides in old ITZ, V_{oitz} is the volume fraction of old ITZ in RCA, V_{oa} is the volume fraction of old NA in RCA, and $V_{om} + V_{oa} + V_{oitz} \equiv 1$ where V_{om} is the volume fraction of old mortar in RCA.

By further using the two-layer spherical model for the old mortar and old ITZ plus old NA, we have the following expression,

$$D_{RCA} = \frac{(3 - 2V_{om})D_{oa+} + 2V_{om}D_{om}}{V_{om}D_{oa+} + (3 - V_{om})D_{om}} D_{om} \qquad (9)$$

where D_{om} is the diffusion coefficient of chlorides in old mortar of RCA. Substituting Eq. (8) into Eq. (9), it yields,

$$D_{RCA} = \frac{2(3 - 2V_{om})V_{oitz}D_{oitz} + 6V_{om}(V_{oitz} + V_{oa})D_{om}}{2V_{oitz}V_{om}D_{oitz} + 3(V_{oitz} + V_{oa})(3 - V_{om})D_{om}} D_{om} \qquad (10)$$

Eqs. (10) and (7) can be used to calculate the effective diffusion coefficients of D_{RCA} and D_{RCA+}, whereas Eq. (6) is used to calculate the effective diffusion coefficient of D_{NA+}. After the D_{RCA+} and D_{NA+} are calculated, the effective diffusion coefficient of chlorides in the RAC with both NA and RCA can be calculated using Eq. (3). It can be seen from Eq. (3) and Eqs. (7) and (10) that, the effective diffusion coefficient of chlorides in the RAC depends on the volume fractions of the mortar, ITZ, NA, and the old mortar, old ITZ and old NA in RCA, as well as the diffusion coefficients of chlorides in individual phases. The volume fractions of the mortar, NA and RCA are related to the mix design of RAC; whereas the volume fractions of old mortar, old ITZ and old NA are related to the parent concrete used for making RCA. The individual diffusion coefficients of chlorides in mortars and ITZs are dependent on their porosities which are controlled by the type and content of the cement and the w/c ratio used for making the concrete. Note that the effective diffusion coefficient is a macroscopic parameter to describe the ionic diffusion through the pore space of concrete. In the present model the effect of pore structure of concrete such as the porosity of concrete on the effective diffusion coefficient is considered through the volume fractions of individual phases involved as well as the diffusion coefficients of chlorides in the individual phases.

4. Results and discussion

The effect of RCA on the diffusivity of RAC is mainly due to its permeable feature caused by the adhered mortar and ITZ in the RCA. Thus, it would be of interest to see how the effective diffusion coefficient of RCA varies with the volume fraction of old mortar in the RCA. Fig. 4 shows graphically the variation of the effective diffusion coefficient of RCA with the volume fraction of old mortar calculated using Eq. (10), in which the volume fraction of old ITZ in RCA was assumed to increase linearly with the volume fraction of old mortar with the proportional factor of 1/5 until it reaches to its upper limit value of 0.1, and thereafter it remains unchanged, which covers the RCA with varying old mortar volume fraction. It can be seen from the figure that the relative effective diffusion coefficient of RCA is almost linearly proportional to the volume fraction of old mortar in the RCA. The proportional factor increases slightly with the increased diffusion coefficient of chlorides in the ITZ of RCA. However, owing to the fact that the volume fraction of the ITZ in RCA is rather small and the difference in diffusion coefficient between the mortar and ITZ in RCA is also small, the effect of ITZ in RCA on the effective diffusion coefficient of chlorides in RCA seems to be limited.

It would be anticipated that the resistance of concrete to chloride penetration will increase with the increased volume fraction of aggregate. This is due to the impermeable feature of NA and the increased tortuosity when more aggregates are incorporated in the concrete (Li et al., 2012; Li et al., 2015). However, when RCA is used to partially or fully replace the NA in concrete, the diffusivity of the mixed concrete will be dependent on the volume fractions and diffusion properties of not only the mortar but also the RCA used in the mixture. The latter is dependent on the properties of the parent concrete used to make the RCA and the quality of RCA. As an example, Fig. 5 shows the variation of the effective diffusion coefficient of chlorides with the replacement ratio of NA (zero means no RCA and one
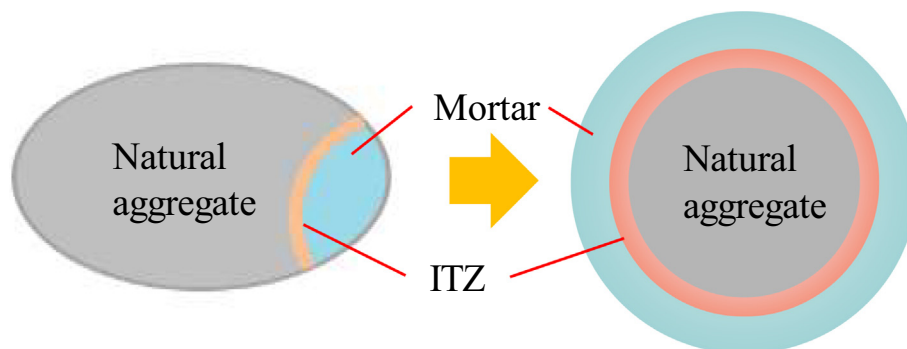

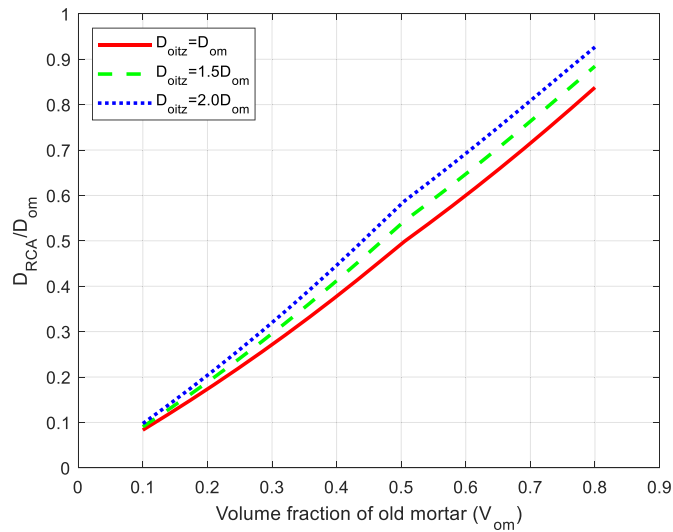**Fig. 3.** Multilayer spherical diffusion model of recycled concrete aggregate.

Fig. 4. Variation of effective diffusion coefficient of chlorides in RCA with volume fraction of mortar ($V_{oitz} = 0.2V_{om}$ and $V_{oitz} \leq 0.1$).

Table 1
Parametric values employed in calculations.

Component	Volume fraction	Diffusion coefficient
RCA		
Old mortar	0.25	$1.5 \times 6.0 \times 10^{-12}$ m²/s
Old ITZ	0.05	$2.0 \times 6.0 \times 10^{-12}$ m²/s
Old NA	0.70	0
RCA+		
New ITZ (NA)	$0.05k_{itz}$	$1.6 \times 6.0 \times 10^{-12}$ m²/s
RCA	$1-0.05k_{itz}$	Eq. (10)
NA+		
New ITZ (RCA)	$0.05k_{itz}$	$1.6 \times 6.0 \times 10^{-12}$ m²/s
New NA	$1-0.05k_{itz}$	0
Concrete with NA+ and RCA+		
New mortar	0.5	6.0×10^{-12} m²/s
NA+	$0.5(1-x)$	Eq. (6)
RCA+	$0.5x$	Eq. (7)

means 100 % RCA) in the concrete with a fixed volume fraction of new mortar of 0.5. The results were obtained by using Eq. (10) for D_{RCA}, Eqs. (6) and (7) for D_{NA+} and D_{RCA+}, and Eq. (3) for D_{eff}. The parametric values employed for the volume fractions and diffusion coefficients for individual phases are given in Table 1, in which the k_{itz} represents the effect, for example, w/c on the volume fraction of new ITZ formed in concrete. The data used here in Table 1 are similar to those used in literature (Hu et al., 2018; Yu and Lin, 2020). It can be seen from the figure that the effective diffusion coefficient of chlorides in concrete increases almost linearly with the replacement ratio of NA. The larger the volume fraction of new ITZ, the higher the effective diffusion coefficient of chlorides in the concrete. For the assumed diffusion properties of RCA, the relative effective diffusion coefficient of chlorides in the concrete could increase by 65–90 % when the 100 % replacement ratio of NA is used.

Fig. 6 shows the variation of the effective diffusion coefficient of chlorides with the volume fraction of new mortar for three different replacement ratios of NA, in which the diffusion properties of RCA given in Table 1 are used. As it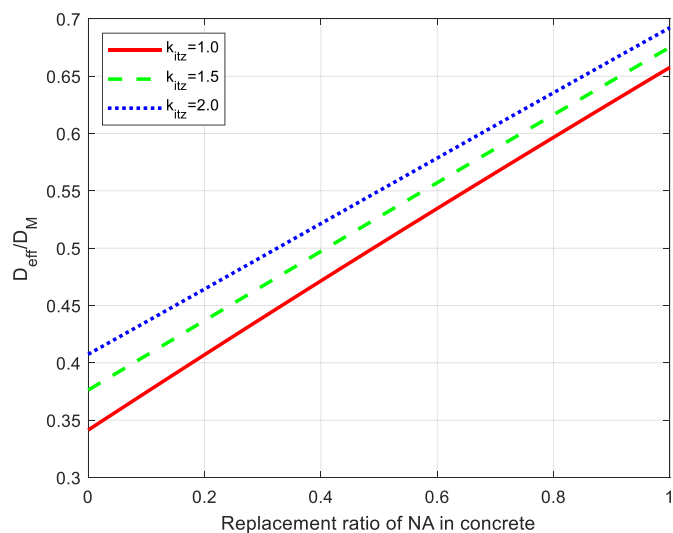 is to be expected, the effective diffusion coefficient decreases with the increase of the total volume fraction of NA and RCA, but the rates of the decrease are significantly different when different NA replacement ratios are used. The concrete with higher replacement ratio of NA decreases much slowly than the concrete with lower replacement ratio of NA. For the case of $V_M = 0.4$, for instance, the relative effective diffusion coefficient of chlorides in the concrete with $V_{RCA} = 3V_{NA}$ is almost 1.6 times that in the concrete with $V_{RCA} = V_{NA} / 3$.

5. Experimental and numerical validation of model

To validate the present prediction model, the comparison between the calculated effective diffusion coefficient of chlorides in concrete using Eqs. (3), (6), (7) and (10) and that measured in experiments is made for the concrete mixed with 75 % ordinary Portland cement (OPC) and 25 % fly ash (FA) (in weight), the results of which are shown in Fig. 7. The experimental data employed herein were taken from Refs. Ying et al. (2013a) and Kou and Poon (2006). The details of the experiments were reported in Kou and Poon (2006) and Kou and Poon (2012). In the experiments, the mixture proportions used were 410 kg/m³ of binder (75%OPC + 25%FA), 611 kg/m³ of sand, and 1017–1048 kg/m³ of natural and recycled coarse aggregates. Two mixtures with water-to-binder ratios (w/b) of 0.45 and 0.55 were used in the experiments. In each mixture the RA (combined stone, RCA and RMA) was used as 0 %, 20 %, 50 %, and 100 % by volume
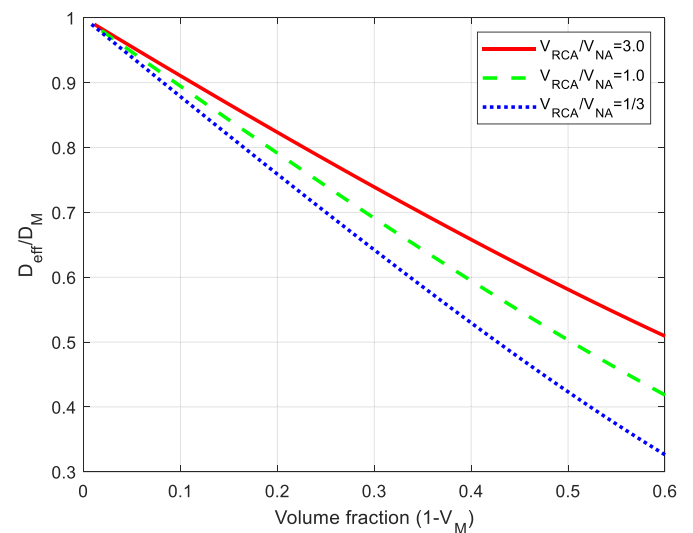

Fig. 5. Variation of effective diffusion coefficient of chlorides with replacement ratio of NA ($V_M = 0.5$, $V_{nitz1} = V_{nitz2} = 0.05k_{itz}$).

Fig. 6. Variation of effective diffusion coefficient of chlorides with volume fraction of RCA + NA ($V_{nitz1} = V_{nitz2} = 0.05$).
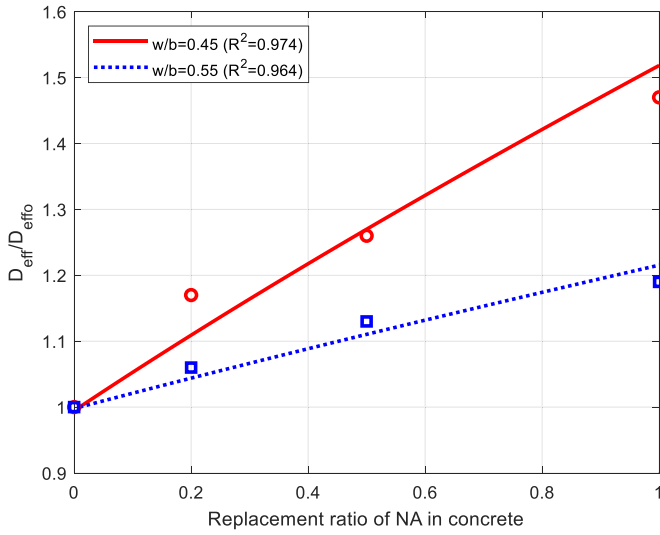
Fig. 7. Comparison of effective diffusion coefficient between predictions and experiments for concrete with different NA replacement ratios (D_{effo} is the effective diffusion coefficient of chlorides in concrete with 100 % NA).

replacement of NA. The diffusion coefficients of chlorides in the RAC were determined in accordance with ASTM C1202 (1997) using a 50 mm thick × 100ϕ mm concrete disc cut from the 100ϕ × 200 mm concrete cylinder. In the prediction model the parametric values used are: $V_{om} = 0.1$, $V_{oitz} = 0.01$, $D_{om} = 6.0 \times 10^{-12}$ m/s^2, $D_{oitz} = 1.5D_{om}$, $V_M = 0.6$, $D_M = 2k \times 10^{-12}$ m/s^2, $D_{nitz1} = D_{nitz2} = 1.5D_M$, $V_{nitz1} = V_{nitz1} = 0.005k$ where k is a constant with $k = 1.0$ for w/b = 0.45 and $k = 2.75$ for w/b = 0.55. These parametric values were assumed based on the experimental data provided in Refs. Kou and Poon (2006, 2012). It is evident from the comparison shown in Fig. 7 that there is a considerably good agreement between the predicted and measured effective diffusion coefficients for the two concrete mixtures with different w/b ratios (both have high coefficient of determination R^2 values). A larger k value reflects the effects of w/b on the volume fraction of new ITZs and the chloride diffusion coefficient in new mortar, which is to be expected. This demonstrates that the use of combined MLSA and EMA in the present model is appropriate.

As the second example of the validation, Fig. 8 shows the comparison of the predicted effective diffusion coefficient of chlorides in concrete using
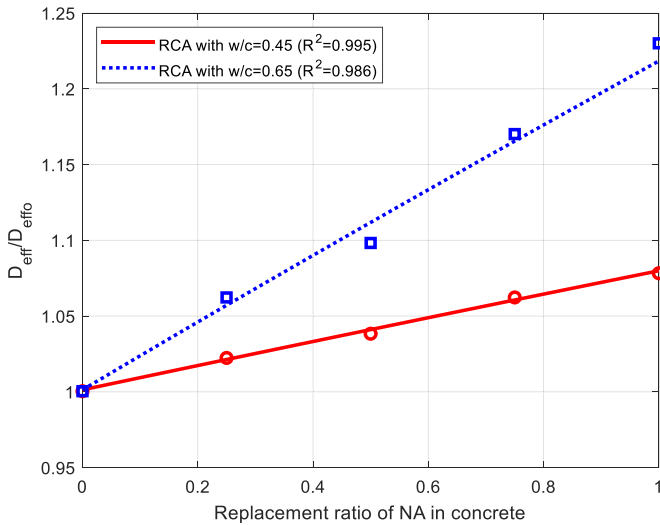

Fig. 8. Comparison of effective diffusion coefficient between predictions and numerical simulations for concrete with different NA replacement ratios (D_{effo} is the effective diffusion coefficient of chlorides in concrete with 100 % NA).

Eqs. (3), (6), (7) and (10) and that calculated using multiphase, mesoscale numerical simulation model (Yu and Lin, 2020). The latter are taken directly from Ref. Yu and Lin (2020). In the multiphase model the concrete was treated as a composite with a matrix (new mortar) embedded with a large number of randomly distributed NAs and RCAs. ITZs with a thin layer were assumed to be surrounding each aggregate. The RCAs of different sizes were also assumed as the composite consisting of old mortar, old ITZ and old NA. Different diffusion coefficients were used for different phase materials, which are generally dependent on the w/c ratios used in the mixture of the concrete and the parent concrete used for making RCA (Yu and Lin, 2020). The diffusion equations were solved numerically in different phases by considering the continuity conditions of both the concentration and its gradient at the interface between jointed phases. However, owing to the significant scale difference between ITZs and other phase materials, the numerical scheme for such analyses need be implemented carefully and it may need to use special finite element methods or multi-scale models (Li et al., 1995). In the present prediction model the parametric values used are: $V_{om} = 0.35$, $V_{oitz} = 0.01$, $D_{om} = 2.25k \times 10^{-12}$ m/s^2, $D_{oitz} = 1.5D_{om}$, $V_M = 0.78$, $D_M = 6.0 \times 10^{-12}$ m/s^2, $D_{nitz1} = D_{nitz2} = 1.5D_M$, $V_{nitz1} = V_{nitz1} = 0.01$ where k is a constant with $k = 1.0$ and 3.25 for w/b = 0.45 and 0.65, respectively, for the two types of RCA used. These parametric values were assumed based on the numerical simulation model employed in Ref. Yu and Lin (2020). The results shown in Fig. 8 illustrates the effect of the w/c ratio in the parent concrete making RCA on the relative effective diffusion coefficient of chlorides in the RAC with different NA replacement ratios. It is evident from the comparison shown in Fig. 8 that there is very good agreement between the analytically predicted and numerically simulated effective diffusion coefficients for the two concretes mixtures with two types of RCA represented by different w/b ratios (both have very high coefficient of determination R^2 values). This again demonstrates that the present analytical model is reliable and accurate in predicting the effective diffusion coefficient of chlorides in concrete with both NA and RCA. The results also show how the effective diffusion coefficient of chlorides in concrete can be reduced by the improvement of RCA (e.g. reducing the w/b ratio in RCA) (Ozbakkaloglu et al., 2018; Vázquez et al., 2014).

Alternatively, the RAC can be interpreted as the modification of the concrete with 100 % NA by adding RCA and simultaneously taking out the NA of the same volume. In this case, the application of EMA leads to the following equation,

$$\frac{1}{\frac{1}{3}D_{effo} + \frac{2}{3}D_{eff}} - \frac{V_{RCA+}}{\frac{1}{3}D_{NA+} + \frac{2}{3}D_{eff}} + \frac{V_{RCA+}}{\frac{1}{3}D_{RCA+} + \frac{2}{3}D_{eff}} = \frac{1}{D_{eff}} \quad (11)$$

Eq. (11) can be further expressed as follows,

$$\frac{1}{1 + 2\left(\frac{D_{eff}}{D_{effo}}\right)} - \frac{V_{RCA+}}{\left(\frac{D_{NA+}}{D_{effo}}\right) + 2\left(\frac{D_{eff}}{D_{effo}}\right)} + \frac{V_{RCA+}}{\left(\frac{D_{RCA+}}{D_{NA+}}\right)\left(\frac{D_{NA+}}{D_{effo}}\right) + 2\left(\frac{D_{eff}}{D_{effo}}\right)}$$
$$= \frac{1}{3\left(\frac{D_{eff}}{D_{effo}}\right)} \quad (12)$$

Eq. (12) indicates that the normalised effective diffusion coefficient, D_{eff}/D_{effo}, depends on three variables, V_{RCA+}, D_{NA+}/D_{effo}, and D_{RCA+}/D_{NA+}. Note that D_{effo} can be calculated using Eq. (10) if (V_{om}, V_{oitz}, V_{oa}, D_{om}, D_{oitz}) are replaced by (V_m, V_{itz}, V_a, D_m, D_{itz}), that is,

$$D_{effo} = \frac{2(3 - 2V_m)V_{itz}D_{itz} + 6V_m(1 - V_m)D_m}{2V_{itz}V_mD_{itz} + 3(1 - V_m)(3 - V_m)D_m}D_m \quad (13)$$

By using Eq. (6) for D_{NA+} and Eq. (7) for D_{RCA+}, we have,

$$\frac{D_{NA+}}{D_{effo}} = \frac{V_{Nitz1}D_{nitz1}[2V_{itz}V_mD_{itz} + 3(1 - V_m)(3 - V_m)D_m]}{3(3 - 2V_m)V_{itz}D_{itz}D_m + 9V_m(1 - V_m)D_m^2} \quad (14)$$

$$\frac{D_{RCA+}}{D_{NA+}} = \frac{(9D_{RCA} + 6V_{Nitz2}D_{Nitz2})D_{Nitz2}}{2(V_{Nitz2}D_{RCA} + 3D_{Nitz2})V_{Nitz1}D_{Nitz1}} \tag{15}$$

For a given concrete if the data for RCA is not available, one can calculate the value of D_{NA+}/D_{effo} using Eq. (14), while the value of D_{RCA+}/D_{NA+} could be assumed. As an example, Fig. 9 shows the prediction curves of D_{eff}/D_{effo} against the replacement ratio of NA calculated using Eqs. (12) and (14), in which all data were taken from Table 1 except for the data for RCA that are assumed. To demonstrate the present predictions, experimental data from literature for different types of concrete with different RCA are also superimposed in the figure. The figure shows that the effect of RCA on the effective diffusion coefficient is largely dependent on the quality of the RCA. The higher value obtained in some tests is probably due to the poor quality of RCA; for example, cracks may exist in the old mortar in RCA. The figure demonstrates that how the RCA quality and corresponding replacement ratio could affect the diffusivity of the RAC.

6. Conclusions

This paper has presented a two-step model for predicting the chloride diffusion in concrete with both NA and RCA. The model uses the combined MLSA and EMA. The former is used to predict the effective diffusion coefficients of chlorides in RCA, RCA + and NA + phases; whereas the latter is used to predict the effective diffusion coefficient of chlorides in concrete mixed with both NA and RCA. The appropriateness and accuracy of the model are demonstrated by the comparisons of the predicted results with those obtained from both the experimental tests and numerical simulations. From the present study, the following conclusions can be drawn.

- The present analytical model can be used to calculate not only the diffusivity of RCA but also the effect of RCA on the diffusivity of RAC when the RCA is used to partly replace the NA in concrete.
- The replacement of NA using RCA in concrete will increase the diffusivity of concrete because the RCA is generally not impermeable. The increase of the effective diffusion coefficient of chlorides in concrete when it incorporates RCA is dependent on the quality of the RCA, which includes the volume fractions of the adhered mortar and ITZ in the RCA as well as the diffusion coefficients of chlorides in them. The present example showed that the effective diffusion coefficient could increase by 10 %,

70 %, and 130 % for concrete with fully replaced RCA with $D_{RCA+}/D_{NA+} = 2$, 10 and 20, respectively.
- For a given type of RCA, the relative effective diffusion coefficient of chlorides in the concrete increases almost linearly with the replacement ratio of NA in the concrete. The rate of the increase, however, is dependent on the properties of the mixture of the concrete such as w/c or w/b ratio. The higher the w/c or w/b ratio is, the smaller the rate of the increase is.
- The effective diffusion coefficient decreases with the increase of the total volume fraction of NA and RCA regardless the NA replacement ratio used. However, the rates of the decrease are significantly different when different NA replacement ratios are used. The concrete with higher replacement ratio of NA decreases much slowly than the concrete with lower replacement ratio of NA.
- The comparison of the predicted effective diffusion coefficient with those obtained from experimental tests and numerical simulations demonstrates that the use of MLSM for the calculation of diffusion coefficients in RCA, RCA + and NA +, and EMA for the calculation of effective diffusion coefficient in concrete with NA + and RCA + is rational and appropriate.

Data availability

No data was used for the research described in the article.

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.

Fig. 9. Comparison of effective diffusion coefficient between predictions and experimental data for different types of concrete with different NA replacement ratios (D_{effo} is the effective diffusion coefficient of chlorides in concrete with 100 % NA. Test data are taken from Amorim Júnior et al. (2019) for (a), Adessina et al. (2019) for (b), Bao et al. (2020) for (c) and Liang et al. (2021) for (d)).

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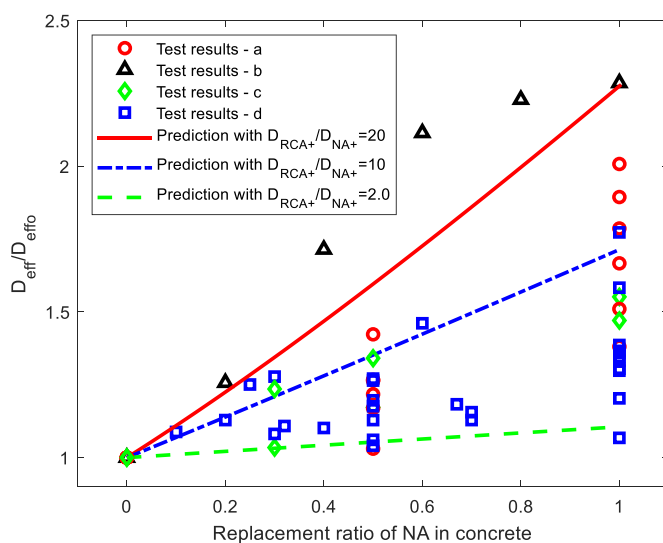
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