Cement and Concrete Research xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Cement and Concrete Research

journal homepage: www.elsevier.com/locate/cemconres

A numerical investigation into the influence of the interfacial transition zone on the permeability of partially saturated cement paste between aggregate surfaces

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ARTICLE INFO

Keywords:
Concrete (E)
Permeability (C)
Water saturation degree
Interfacial transition zone (B)
Modelling (E)

ABSTRACT

The interfacial transition zones (ITZs) are supposed to promote fluid transport through concrete. As a consequence, one would expect an increase in permeability with an increasing aggregate fraction. This has been shown in some experiments, however, the opposite effect is observed as well. The permeability ratio of ITZ to matrix seems to be a key parameter in interpreting this controversial phenomenon. A higher ratio favors the flow of water through the interface zone. This work aims at studying this ratio at various conditions (*i.e.*, hydration degree, water/cement ratio, particle size range and water saturation degree) using a numerical model, so that the influence of the ITZ on the permeability of cementitious composites can be better understood. The findings presented in this paper can provide a new perspective on controversial experimental results as to the effect of the ITZ on transport capacity.

1. Introduction

Concrete on meso-level is generally considered a three-phase material. Major components are the cement matrix and aggregate particles. However, modifications of the cement structure around aggregate surfaces referred to by the interface zone can be viewed as the third phase. This ITZ reveals significant compositional differences as compared to the bulk paste. These differences are most significant very close to the aggregate surface and gradually diminish away from the aggregate grain surface to become insignificant at a certain distance (15–50 μm) [1]. The ITZ differs from bulk paste in porosity, pore size and also in the complementary anhydrous cement and C-S-H (calcium silicate hydrate) contents. In general, the ITZ contains less anhydrous cement grains, larger pores and higher porosity, resulting in higher transport properties (*i.e.*, permeability, diffusivity and conductivity) in contrast to that of bulk paste. The ITZ fraction goes up and the spacing between adjacent ITZs decreases at increased aggregate content, both leading to a larger degree of ITZ overlap. According to the ITZ percolation theory [2], the ITZs will mutually connect and thus fast-conduction pathways will be formed through the material once the aggregate content reaches a threshold value. In theory, the permeability of concrete should rise at the increased aggregate content and a sudden increase can be expected at the percolation threshold [3]. This is confirmed by Halamickova's work [4]. Yet, it is not always the case in practice. Contradictory results are also experimentally obtained in permeability tests. As an example, the permeability of concrete is observed to go down at an increased aggregate fraction in [5]. A similar controversy exists regarding the influence of the ITZ on chloride diffusivity and electrical conductivity [6–8].

Although earlier studies [8–10] have been performed to better understand why such contradictory observations exist, the ITZ structures were in general simply treated as homogenous layers covering the surface of hydrating cement grains. Furthermore, the existing models [11–12] that have looked at the effect of technological parameters on the contrast ratio of ITZ to bulk cement paste are mostly based on diffusion rather than on permeation. The objective of this paper is to study the influence of several technological parameters (*i.e.*, hydration degree, water/cement ratio, cement particle size range and water saturation degree) on the permeability of the ITZ and bulk paste. For that purpose, the ITZ is numerically constructed using a DEM-based (Discrete Element Modelling) approach, as will be described in Section 2.1. It is expected that the presented work can provide a new perspective on this controversial experimental phenomenon.

In general, there exists a set of competing effects when adding aggregates to the cement matrix [5,9]. On the one hand, porous matrix is replaced by impermeable aggregate, resulting in a decline in the total porosity and thus permeability. This is denoted as the dilution effect. The enlarged amount of aggregates also leads to an increase in the

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http://dx.doi.org/10.1016/j.cemconres.2017.09.005
Received 21 July 2017; Received in revised form 25 August 2017; Accepted 5 September 2017

tortuosity of transport paths [13]. These two effects mainly result in reduced transport properties. This is counteracted by the enlarged area of the more porous ITZ, facilitating the ingress of fluid and thus promotes the transport properties. As a consequence, the effects of aggregates on concrete's transport properties depend on which factor will dominate the scene. According to Garboczi and Bentz [14], the conductivity ratio of ITZ to matrix paste ($\sigma_{ITZ}/\sigma_{matrix}$) is an important parameter in determining the relationship between mortar conductivity and the sand volume fraction. It is found that a value of the $\sigma_{ITZ}/\sigma_{matrix}$ of at least 10 to 20 is required to compensate for detrimental effects so that the conductivity of mortar can be enhanced at increased sand content. Later, Shane et al. [15] further pointed out that $\sigma_{ITZ}/\sigma_{matrix}$ varies with the hydration time and is actually a function of the degree of hydration. The peak with a value of approximately 7 only occurs when the degree of hydration is around 0.7. This value is below 10, so it is not likely that the ITZ will significantly enhance the conductivity of mortar [15]. From permeability studies it is already known that permeability is much more sensitive to pore size than conductivity is [4–5]. Due to the large pores in the ITZ region, the permeability ratio between ITZ and matrix paste ($\kappa_{ITZ}/\kappa_{matrix}$) is expected to be much larger than the conductivity ratio $\sigma_{ITZ}/\sigma_{matrix}$. In reality, the difference may be even larger because of microcracks in concrete. Nevertheless, the threshold value for $\sigma_{ITZ}/\sigma_{matrix}$ and $\kappa_{ITZ}/\kappa_{matrix}$ that is used to evaluate the influence of the ITZ on transport properties of cementitious material is supposed to be similar. When $\kappa_{ITZ}/\kappa_{matrix}$ is exceeding the threshold value, the ITZ effect will outweigh the other effects of the aggregate, so that the ITZ is supposed to enhance the permeability of concrete. Otherwise, the dilution and tortuosity effects become dominant, which leads to a reduced permeability at an increased aggregate content. Fig. 1 shows the different effects of the ITZ on the conductivity of mortar at increasing sand volume fraction. It seems that the conductivity ratio between the ITZ and the bulk matrix should exceed a threshold so that the ITZ would promote the conductivity of mortar. However, relatively little is known about the quantitative differences between the permeability of the ITZ and the bulk paste. Additionally, cementitious materials in service are seldom fully saturated due to water evaporation. Yet, the saturation degree of specimens has been proven an important factor governing permeability [16–19]. It should likewise affect the value of $\kappa_{ITZ}/\kappa_{matrix}$. Unfortunately, experimental research on the $\kappa_{ITZ}/\kappa_{matrix}$ relationship to the permeability of concrete has not been conducted yet. It will constitute therefore the main content of the presented work. Since experimental measurements on permeability of cementitious materials usually require specialized equipment and long periods of time to complete, a modelling approach is employed in the present case due to its economic and reliable characteristics. Drying-induced micro-cracking is another crucial factor affecting the transport properties of concrete and thus the permeability ratio ($\kappa_{ITZ}/\kappa_{matrix}$), as shown by recent experimental and 3D modelling studies [20–23]. Due to the nature of ITZ, cracks in practice tend to initiate in this zone. Once these cracks coalesce, the water ingress process could be promoted, resulting in a higher $\kappa_{ITZ}/\kappa_{matrix}$. Moreover, air voids may exist in mortars or concretes in real experiments due to inadequate compaction and thus exaggerate the ITZ effect. This may be the reason why the measured permeability in [4] goes up with the increasing aggregate content. However, the influence of micro-cracks on water transport of cementitious materials is not within the scope of this study.

In this paper, a systematic study is performed to evaluate the influence of several parameters (*i.e.*, degree of hydration, water/cement ratio, particle size range and saturation degree) on the value of $\kappa_{ITZ}/\kappa_{matrix}$. A brief introduction on the developed numerical approach will be given in Section 2. Since the full methodology has been presented in detail earlier, the interested readers are referred to the relevant publications for more information [18,24]. The respective effects of degree of hydration, water/cement ratio, particle size range and saturation degree on the permeability ratio of ITZ to matrix paste will be presented in Section 3. The value of $\kappa_{ITZ}/\kappa_{matrix}$ is found to strongly depend on the sample's conditions (degree of hydration, water/cement ratio, particle size range and saturation degree). The findings presented in this work are compared with the results from [15].

2. Numerical approach

Since the developed modelling technique has been presented in full detail in [18,24], only a brief introduction is given here. Nevertheless, enough information will be provided in this section to enable the readers to understand how the modelling operation is performed.

2.1. Generation of the bulk material and ITZ

It is well known that the origin of the ITZ derives from the cement particles packing against the aggregate surface [1]. Since the aggregate size is in general much larger than that of the cement grains, the aggregate surface can actually be considered flat. When the cement

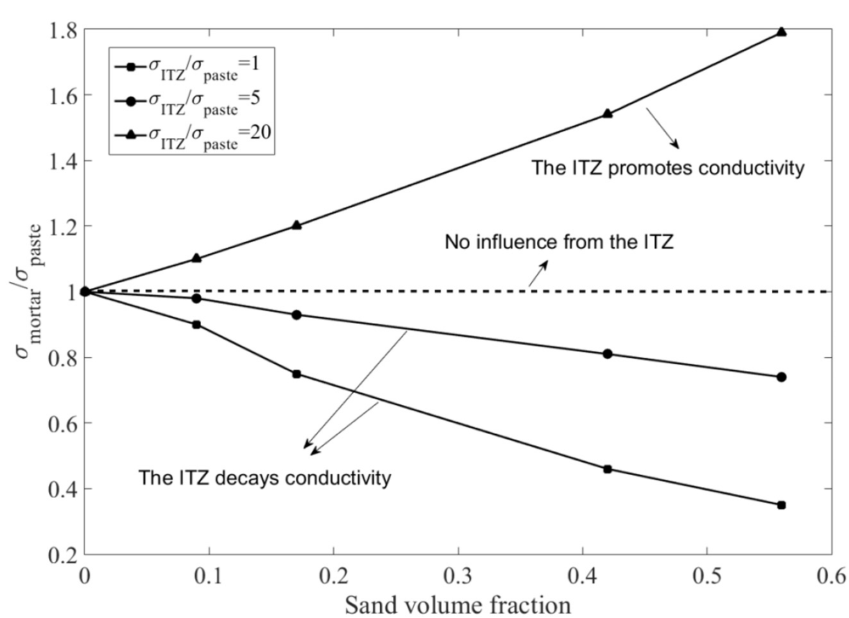

Fig. 1. Conductivity ratio between mortar and cement paste (σ_m/σ_p) is shown as a function of sand concentration for several values of the interfacial zone conductivity to bulk matrix ($\sigma_{ITZ}/\sigma_{matrix}$). Dashed line indicates no difference in conductivity between mortar and cement paste at a certain threshold of $\sigma_{ITZ}/\sigma_{matrix}$. The data shown in this figure are obtained from [8].