Systematic research on compucrete can shed light on some controversial issues in concrete technology

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Realistically simulating fresh and hardening cementitious materials renders possible understanding controversial issues existing in the field of concrete technology. The experimental studies on the impact of the interfacial transition zone (ITZ) on permeability of concrete reveal two controversial results. The first involves the concept of promoting the permeability by increasing the aggregate fraction of concrete that will lead to (more) ITZ percolation. This is supported by some experiments reported in the literature. However, contradictory data are also published by other researchers. This paper aims at explaining by an advanced modelling technique why these conflictive observations are experimentally obtained.

Key words: Permeability, concrete, saturation degree, interfacial transition zone

1 Introduction

Economy demands reduce the possibilities to experimentally lay down in a reliable way the major contours of complex phenomena in concrete technology. In such cases, it is easier nowadays, less time-consuming and more efficient to do this by compucrete, i.e., virtual concrete produced by computer software. This paper demonstrates how controversial experimental results in practice can be explained by modelling techniques.

ITZs overlap (or percolation) in concrete and thus should make concrete more permeable, since the ITZ is generally assumed to have larger pores and higher porosity than bulk paste. One can find experimental data supporting this logical concept (Halamickova et al., 1995). However, also contradictory findings can be found in the international literature (Garboczi and Bentz, 1996; Wong et al., 2009; Zheng et al., 2009). Thus, it would be interesting to investigate why these two conflictive results are both experimentally obtained. Advanced modelling techniques enable us to perform such studies in an effective way.

Concrete, generally recognized as a three-phase material, consists of aggregate, cement paste and the ITZ. Introducing aggregates into the cement matrix impacts transport properties by: dilution effect, increased tortuosity of transport channel and more porous ITZ. The former two factors contribute to a decline in permeability when increasing the aggregate fraction, while the ITZ is thought to facilitate the flow transport. Thus, the overall influence of added aggregates on permeability of concrete is determined by the factor that would be dominant. In Delagrave et al. (1997) and Shane et al. (2000), the diffusivity ratio between the ITZ and matrix is shown to be an important parameter and its value should be above a threshold such that it becomes dominant. In this case, the diffusivity of concrete goes up with the increased aggregate fraction. Otherwise, an opposite trend should be expected. In comparison to the conductivity study, the permeability is expected to show a similar phenomenon since they are both fundamentally transport-based properties. The permeability ratio of the ITZ to matrix is actually not a constant but it depends on the samples' states (i.e., hydration period, water saturation degree). This paper will show how the permeability ratio of the ITZ to the matrix varies with advancement of hydration and reveal its role in determining the permeability of concrete. It seems able explaining at least partly the two controversial experimental observations. Moreover, the specimens in practice are never fully saturated with water and this has been proven to have a severe effect on permeability. Therefore, the influence of water saturation degree is also taken into account in this work.

This are the questions on which we will focus in this paper, whereby the numerical approach will be applied. Since we deal with structure-sensitive properties, the approach is based on DEM (discrete element method) particle packing. An improved vector-based method (extended integrated particle kinetics model, XIPKM) is employed for hydration simulation. Input data for a network analysis are coming from a robotics-derived pore delineation algorithm (double random multiple-tree structuring, DRaMuTS) and a star volume method used in life sciences for pore measuring. The complete methodology has been outlined earlier in other international papers (Li et al., 2015; Stroeven et al., 2015) to which readers are referred for the methodological details. Herein only a brief introduction of the used methodology will be given in Section 2, so that readers can better appreciate what is pursued and how.

2 Brief outline of methodology for permeability estimation

Figure 1 illustrates the five stages in this normal approach to permeability estimation of fully saturated cement paste. The methodological modules or stages concern:

(1) A so called dynamic force-based DEM is employed instead of the quite popular RSA (random sequential addition) approaches in concrete technology. The latter are particle deposition techniques, whereas the particles are mixed by a dynamic stage in the used DEM package, HADES. Consequences are a more realistic particle dispersion and extended capabilities to produce high particle densities. The size range and distribution of the binder particle are set by the researcher. The first involves always a restriction compared with real cements because of computer capabilities. The second is generally - and also in the research described herein - governed by a Rosin Rammler function. To model the effect of the ITZ, two rigid boundaries are introduced in X-direction during the packing simulation while periodic boundaries are used on the other four directions.

(2) Hydration simulation is a complicated operation. XIPKM formulates the different relevant hydration reactions of the major compounds of the cement and of the mineral admixture and their impact on the increasing volume of the respective particles (Le et al., 2013). The latter leads to interferences of the hydrating particles. To restrict complications, the binder particle are in general and also herein assumed spherical. A spatial pore network structure with complex geometric and topological properties is gradually arising. This process stops when all available water has been consumed at the ultimate degree of hydration (UDH).

(3) Topology of the pore network is assessed by DRaMuTS (Stroeven et al., 2012). Multiple "trees" are let to grow from seeds at the opposite surfaces, and forced to merge when in the same pore. The growing process makes use of the random points that are connected by straight lines. When impossible because of intersection with the pore network surface, the point is shifted along the connecting line. In this way, isolated pores and dead-end branches can be distinguished and eliminated from the present study set up.

(4) Point density governs porosity, however for assessment of pore size, Star Volume Measuring (SVM) is employed (Le and Stroeven, 2012). Since the earlier random point system is biased due to a shift of points, a second random point system is generated. Each

point is the nucleus of a "star" of which a large number of its pikes is used to measure local pore size. A sensitivity analysis resulted in using 10^5 points in pore space in a container of 100 µm.

(5) For constructing the tube network for flow simulation, the smallest pore cross section per point is additionally determined by 2D stars (yielding the so called throats). Moreover, the relationship between local pore shape and conductance is assessed for a relatively large but still limited number of points and approximated by regression analysis. This line is used to estimate conductance in all throats, serving as input parameters for establishing the tube system. Pore length in the tree structure is obtained by mathematically smoothing the zig-zag lines resulting from the robotics search procedure. All details can be found in Le (2015).

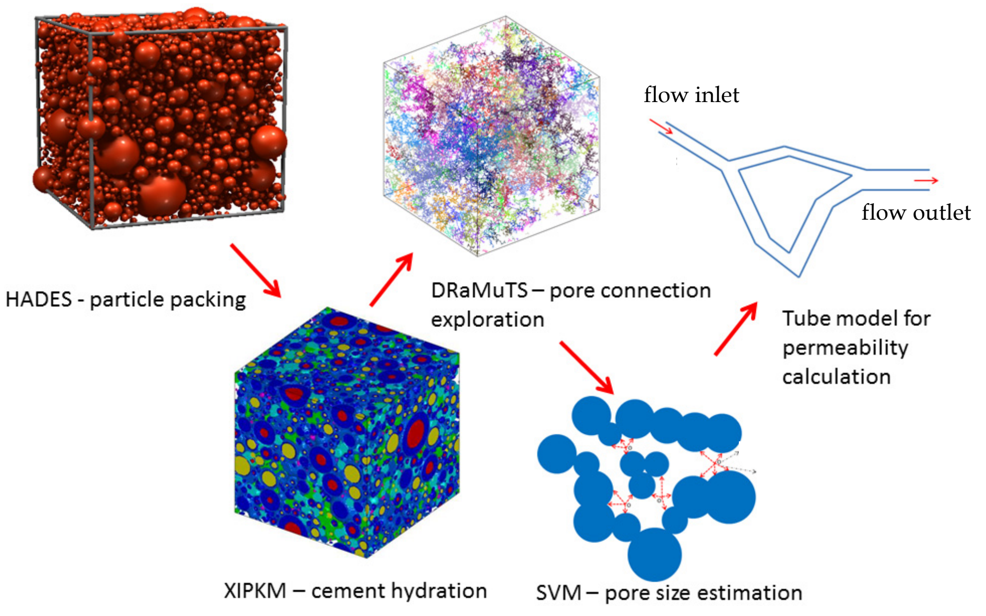

Figure 1. Successive stages in the normal approach to permeability estimation in fully water saturated cement paste. 1. HADES is a dynamic force-based discrete element system for particle packing; 2. XIPKM is a vector-based simulation system for hydration simulation of spherical binder grains; 3. DRaMuTS is a from robotics derived pore delineation system; 4. SVM is a method used in experimental approaches in life sciences for 3D pore size estimation; 5. Tube network modelling is one of the classical methods for permeability estimation of fully saturated cementitious materials (Li et al., 2016a). (Colour figures are available at www.heronjournal.nl.)

(6) For partly water saturation conditions, a blocking algorithm is developed in Li et al. (2016b). Starting from the largest pores that are likely to empty first due to water evaporation, the relevant places are successively blocked. This can easily be accomplished because information is readily available in the pore data system.

3 ITZ percolation and its influence on permeability of concrete

When the aggregate fraction is increased, the disturbed boundary zones in the binder particle packing around the aggregate grains (also referred to as ITZs) come closer and will also overlap to an increasing degree. At a certain threshold, the ITZ becomes interconnected and a direct path for fluid transport forms. This is the phenomenon of ITZ percolation. Due to the fact that the ITZ contains larger pores and is more porous than the bulk phase, it seems logic assuming that the resulting growing ITZ overlap (i.e., percolation) at densification of the aggregate skeleton will promote higher permeability outcomes. Indeed, supporting evidence by experimental studies can be found in the literature (Halamickova et al., 1995). However, this is not always the case in practice since a reversed trend is also reported (Wong et al., 2009; Zheng et al., 2009). In fact, it should be mentioned that a higher volume fraction of aggregate automatically causes a reduction of the paste volume, and its contribution to permeability. This is referred to as the particle insulation effect. Additionally, the pore tortuosity that is found proportional to aggregate volume fraction (Stroeven, 2000) is increasing, and so delaying the water flow through the material. This is a competing set of parameters, among which ITZ overlap can only be expected the dominating one under certain conditions. The ITZ overlap fraction can be theoretically assessed by the hard core-soft shell model, as presented in Zheng and Zhou (2007). However, the model of hard core-soft shell underlying such discussions gives an incorrect impression on the matter. When larger pores are found in the ITZ, they are supposed to follow the aggregate surface and unite with such pores in the overlapping neighbouring ITZ. Only then the formation of major continuous pore channels in the pore network structure will be promoted. The pore network system is a result however of the packing of cement particles in the fresh state in bulk and near the rigid surfaces of aggregate grains. These processes are governed by ordering and by stochastic influences, inevitably leading to patch formation (Diamond, 2003; Stroeven et al., 2009). Although this was challenged by Wong and Buenfeld (2006) as an artefact of specimen preparation, the patch formation imposes the pore channels to meander in an erratic way though the material (Stroeven, 2000). So, overlap of boundary zones around aggregate grains will not automatically lead to increased pore continuity of the larger pores. Moreover, pore tortuosity will also be much higher than estimated by the hard core-soft shell approach. So an increased packing density of the aggregate may have initially a smaller effect on permeability and will be more easily counteracted by increased tortuosity at higher aggregate packing density. The dispute on opposite effects of ITZ percolation on permeability can therefore be settled readily by systematic research on DEM-based compucrete, as discussed herein.

In this paper, we will primarily focus on the permeability ratio of the ITZ and bulk matrix, since this parameter is the only factor promoting permeability. In Shane et al. (2000), this ratio is estimated to be of the order of 10-20, so that ITZ overlap will easily compensate for the decreased permeability due to the particle insulation effect and to an increased tortuosity in the aforementioned case. It is investigated as a function of hydration time on the basis of a DEM-produced particular compucrete quality, revealing a quite dramatic peak at an advanced period, as will be presented in Section 4.1. The differences in the permeability ratio of the ITZ and matrix at different hydration stages and for various material parameters (data published in Li et al., 2016c and Li et al., 2017) will contribute to the conflicting behaviour in experimental settings.

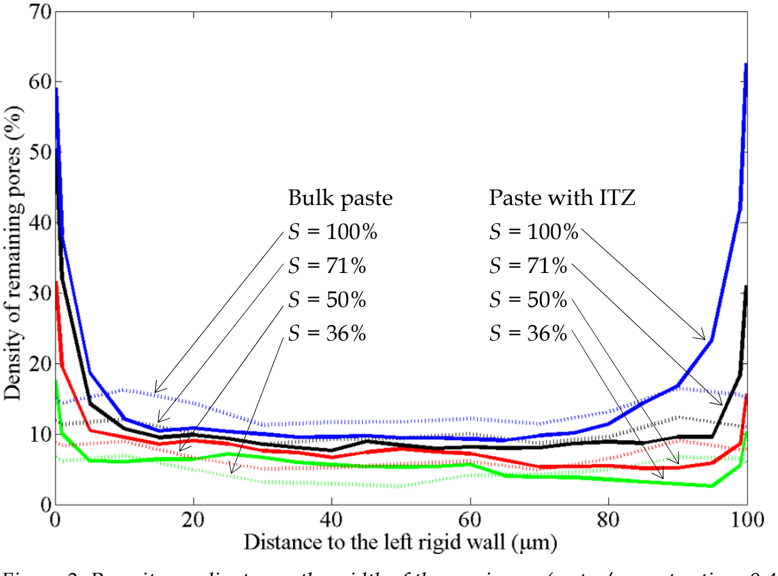

Figure 2. Porosity gradient over the width of the specimens (water/cement ratio = 0.4, hydration age: 28 days, PSD:1-30 μm); remaining pores after blocking due to water evaporation

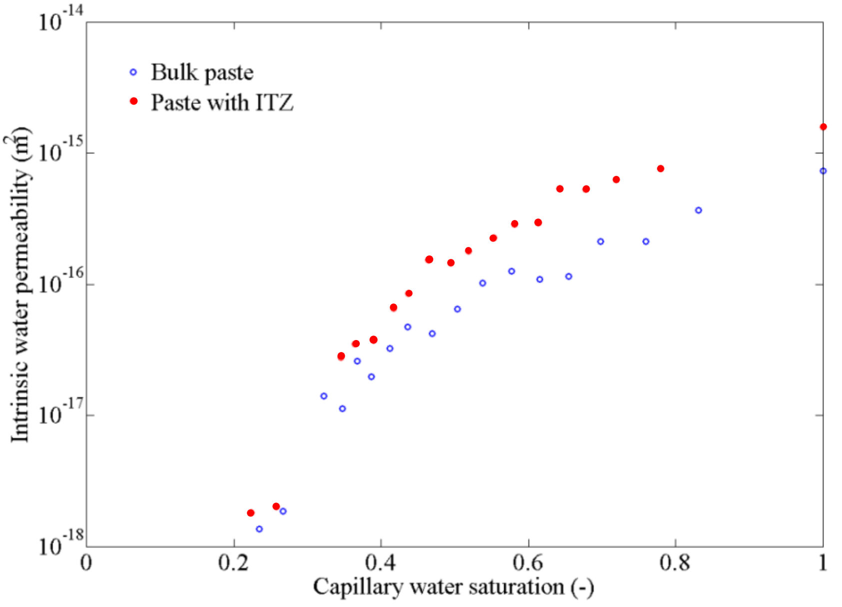

Figure 3. Influence of ITZ on water permeability of cement pastes (water/cement ratio = 0.4, hydration age: 28 days, PSD:1-30 µm)

Still a major factor that influences the permeability is the water saturation degree of the specimens (Kameche et al., 2014; Li et al., 2016b). In practice, concrete will never be completely saturated with water. In research, even concrete specimens stored before testing under submerged conditions, will contain air bubbles in the pore network system that reduce water transport through the material. The influence of the water saturation degree, *S*, on the permeability of specimens containing the ITZ or not has been systematically studied by the authors (Li et al., 2016c), as illustrated in Figs. 2 and 3. Specifically, Fig. 2 convincingly demonstrates how the significant difference in porosity between ITZ and bulk paste dramatically declines with reducing *S*. As a result, the influence of the ITZ on permeability also vanishes at low values of *S*, as reflected by Fig. 3. This mostly ignored (or incorrectly interpreted!) factor dominates the scene. The balance between the earlier mentioned three factors is significantly disturbed at diminishing value of *S*, leading to negative effects of the ITZ percolation influence on permeability. The permeability ratio between the ITZ and cement matrix is found to be an important factor in studying the effects of the ITZ on the permeability of concrete, as discussed in another publication (Li et al., 2017) of the authors. However, some findings are used in this paper as well and will be presented in Section 4.

4 ITZ to bulk permeability ratio versus hydration time

4.1 *Fully saturated concrete*

For a fully saturated concrete (S = 100% and water/cement ratio = 0.4), the permeability of the ITZ and of the cement matrix at various hydration ages was calculated by our numerical approach, as described in Section 2. The outcomes are shown in Fig. 4. Degree of hydration (DOH) is defined as the reacted cement at a certain hydration time divided by the total amount of cement at initial stage. Thus, the permeability ratio between the ITZ and matrix is straightforwardly obtained and plotted in Figure 5.

It is found that permeability of ITZ and bulk regions decline as hydration proceeds. On a log scale this has been shown a similar phenomenon. Since the permeability in ITZ and bulk paste are almost proportionally declining with hydration time, the ratio of both is almost constant, with permeability of the ITZ as the larger one in this not heavily compacted structure. At an advanced hydration stage, the pore de-percolation starts leading to a sharp decline in permeability in both phases. Yet, the pore de-percolation effect can be expected starting earlier in bulk paste because of the finer pore network structure. For hydration values at which pore de-percolation will start in bulk and ITZ, respectively, the ratio of ITZ and bulk permeability will therefore reveal a distinct peak (DOH is about 0.76). This has been found in satisfactory agreement with work by Shane et al. (2000). The peak value of the curve shown in Figure 5 is around 52. This value is larger

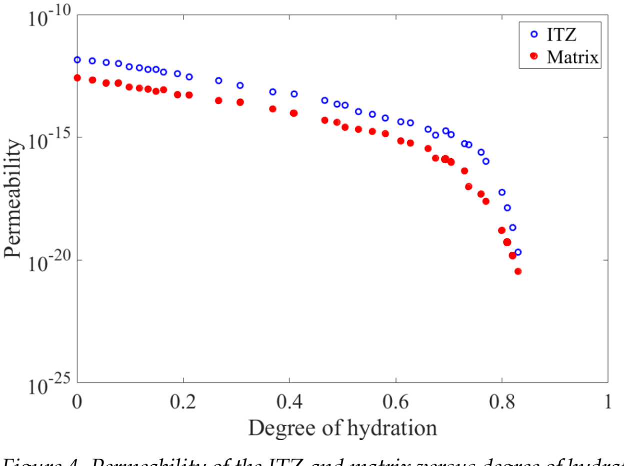

Figure 4. Permeability of the ITZ and matrix versus degree of hydration of fully saturated concrete (water/cement ratio = 0.4) (Li et al., 2017)

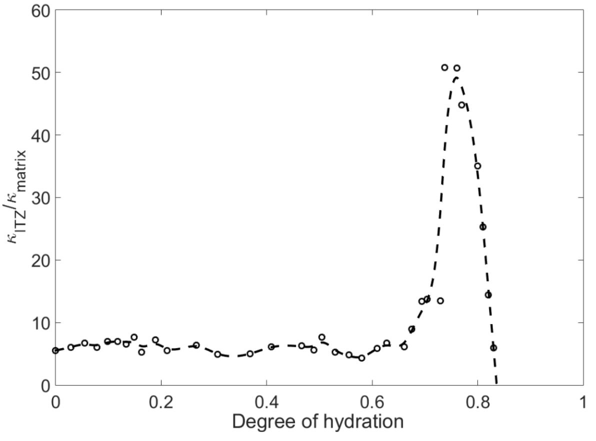

Figure 5. Permeability ratio of the ITZ to matrix versus degree of hydration of fully saturated concrete (water/cement ratio = 0.4). Dashed line follows the original observations in a smooth way (Li et al., 2017).

than the reported one (i.e., 20) by Shane et al. (2000). Nevertheless, permeability is expected to be promoted under such conditions. However, the permeability ratios at other hydration stages are mostly below 20. So, the particle dilution effect and increased tortuosity become more dominant factors influencing the permeability value.

4.2 *Partially saturated concrete*

So far, all the samples used in this particular study were fully saturated with water. However, concrete in practice will inevitably loose water by evaporation. This has been proven having a significant influence on permeability. So, a blocking algorithm was developed to represent this state of virtual concrete, as presented in Li et al. (2016b). The same methodology as earlier described is used to produce virtual specimens at different saturation degrees. Afterwards, the permeability of the ITZ and cement matrix can be separately calculated and then the permeability ratio is straightforwardly obtained. The results are presented in Fig. 6. Three different water saturation degrees are taken into account in this work. Curves in black, blue and red represent S = 100%, S = 67% and S = 44%, respectively.

At decreasing water saturation degree, the peak value is found to gradually go down and finally completely vanish. This is because the large pores existing in the ITZ region are first to be blocked in the process of water evaporation according to the Kelvin-Laplace equation. So, at a low water saturation degree, the difference (in terms of pore structure) between the ITZ and matrix phase is not so significant. This makes the permeability ratio of the ITZ and matrix at S = 44% almost a constant at a level of about 1-2. In this case, the permeability of concrete is not governed by the ITZ. The tortuosity effect and the particle insulating effect win over the increased permeability of the ITZ. Therefore, it will be likely to observe in practice a reduction in permeability of concrete due to these ITZ properties, as experimentally observed in Wong et al. (2009).

5 Discussion and conclusions

This paper aims at explaining controversial observations reported on in the international literature in the field of permeability research on concrete. To produce concrete, aggregates are mixed with cement paste and generally compacted by vibration. This introduces boundary zones in the packing of binder grains near all aggregate particles. Herewith, ITZs are introduced as a model concept in the system. Although the more porous ITZ supposedly promotes the water flow through concrete, whether the ITZ significantly enhances the overall permeability of concrete is still an issue for dispute. Opposite tendencies based on experimental approaches have been reported in the literature.

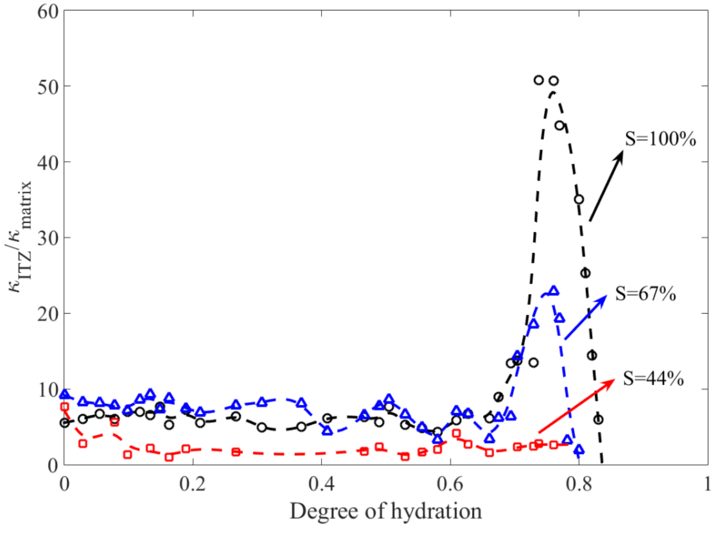

Figure 6. Permeability ratio of the ITZ to matrix versus degree of hydration of partially saturated concrete (water/cement ratio = 0.4). Dashed line is a smooth approximation of the indicated original observations (Li et al., 2017).

Analysis of the case shows that attributing factors are the ITZ overlap, on the one side, and particle insulation and pore tortuosity, on the other side. The presented study on compucrete learns that the erratically meandering pores in the network structure will decline the likelihood of pore channel continuity due to ITZ percolation. In parallel, it also yields the estimate for the pore tortuosity from the hard core-soft shell model a dramatic underestimate. Depending on material parameters, the outcome of these competing influences on permeability will thus be different. The permeability ratio of ITZ and bulk paste is shown to be a function of hydration time and a significant peak (of 52 or so) is found for fully saturated samples with a *w/c* ratio of 0.4 at *DOH* = 0.76. For such conditions, ITZ percolation may be the dominating factor. However, for other material parameters (i.e., *w/c* and particle size range), this influence will be different, as will be discussed elsewhere. Additionally, herein it is demonstrated that the water saturation degree, which is governed by environmental conditions, has also significant influence on the permeability ratio. With a decrease in the water saturation degree and given material parameters, it is observed that the peak value of the permeability ratio gradually goes down at the same *DOH* and finally completely vanishes under lower water saturation conditions. At other hydration degrees, the ratio is maintaining an almost constant value of 1-2. Hence, even the case of *w/c* = 0.4 does not lead to a significant influence of the ITZ on permeability for lower saturation degrees. Since concrete in practice has certainly a reduced saturation degree, so is not fully saturated as assumed in modelling, it is not likely that the ITZ will *significantly* enhance the permeability of real (partially saturated) concrete and neither will have ITZ percolation such an effect.

Hence, the systematic studies on DEM-produced compucrete discussed in this paper can provide insight in the background of controversial permeability results available in the literature.

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