Effects of Interfacial Transition Zone percolation on transport through concrete. Fact or myth?

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ABSTRACT

In this paper transport tests are carried out on concrete and mortars of gradually increasing Aggregate Fractions (AF) to examine the validity of ITZ percolation theory that blames the sudden increase in porosity of the mortar at a critical aggregate fraction on the joining up of the ITZs together to make a continuous path way through concrete. Test Results and their effects of the validity of the percolation theory are discussed. It is suggested that the jump in porosity at the critical aggregate fraction may be completely independent of ITZ altogether and two candidates of aggregate spacing and paste roughness have been proposed to replace ITZ percolation as the cause of this sudden increase at the critical aggregate fraction. The need for further tests have been established to elucidate the remaining ambiguities as to what exactly happens to the paste in concrete and its porosity as the volume of the aggregate is increased to and beyond critical value.

KEYWORDS

Interfacial Transition Zone; ITZ; Percolation; Transport Properties; Mercury Intrusion; Electrical Conduction; Carbonation; Microstructure; Oxygen Diffusion; Oxygen Permeation; Water Penetration.

INTRODUCTION

A key aspect in producing durable concrete structures is to produce concrete of high resistance to the transport processes. Concrete consists of discrete aggregate particles embedded in a continuous cement paste. It has long been appreciated that the existence of aggregate in concrete changes the microstructure of the paste in concrete from that of neat cement paste [1, 2, 3]. It is also known that the microstructure of the cement paste in the vicinity (between 20 and 50 μm thick) of an aggregate particle (Interfacial Transition Zone) in mortar or concrete differ from that further away from the aggregate [4, 5], where there are higher porosities and hydration products and lower anhydrous compared with the bulk paste [6].

More detailed features of ITZ are covered by different authors e.g. Scrivener [5], and Diamond [2, 3 and 7]

When very fine particles are present in concrete, such as silica fume or even inert particles such as carbon black, these particles pack preferentially close to the aggregate and there is little or no increase in porosity in the interfacial region [8, 9].

Snyder, et al.[10] at NIST have measured the porosity of mortars of different volumetric aggregate fractions of 0, 0.157, 0.384, 0.448, 0.486 and 0.554 by MIP (Fig 1) and their results show that at AF = 0.486 the cumulative pore volume in the paste shows 5.6 % increase compared with their mortar of AF = 0.448. They have assumed that the reason for this huge jump is that at a volumetric Critical Aggregate Fraction (CAF) the ITZs interconnect and percolate the system forming a continuous path through the mortar.

This study is designed to test the percolation theory explained above [10] and hence Electrical Conductions, Oxygen diffusions, Oxygen Permeations and Water Penetrations are measured in concretes of AFs 0.5, 0.2, 0.3, 0.4, 0.5 and 0.6 with W/C of 0.45 and also Electrical conductions and Carbonation Depths are measured in OPC mortars as well as SF (replacing 20 % of the volume of OPC cement) mortars of AFs 0.1, 0.2, 0.3, 0.35, 0.40, 0.45, 0.50, 0.55, 0.60 and 0.65 with the same W/C ratio to examine the validity of the ITZ percolation theory.

A critical AF is experienced in OPC as well as SF mortars at AF = 0.35, where Electrical Conduction and Carbonation depth are temporarily increased, but this increase is exacerbated rather than being eliminated in SF mortars. Also these increases have been shown to be only temporary at the CAF and they disappear beyond CAF.

It is proved that the ITZ percolation theory cannot justify the sudden huge jump of porosity at the critical AF. It has been suggested that this jump may be completely independent of ITZ altogether and two candidates of Aggregate Spacing and Paste Roughness are proposed to replace the ITZ percolation as the cause of the temporary jump in the paste porosity at the critical AF.

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EXPERIMENTAL PROGRAM:

AGGREGATE SIZE DISTRIBUTION FOR CONCRETE

The Chesil beach gravel was sieved into four different size ranges (within 2.36-10 mm) and then the more rounded particles were hand picked, washed, and dried. The Chelford sand was sieved into 17 different size ranges (0.063-2.36 mm). The sieved sands and gravels are recombined meticulously to produce a smooth grading curve typical of pumpable concrete (Fig.2).

SPECIMEN MANUFACTURE OF CONCRETE

All concrete mixes were prepared in the same small pan mixer, the mixing procedure being essentially that described by BS 1881: part 125: 1986. At first the concretes were cast in cylindrical moulds of the arrangement shown in Fig 3 which were subjected to vibration, de-airing, inversion and “roll-vibration”, “roll-vibration” and then rotation for 24 hours at a speed of 6 revs/min. After setting, cylinders were sliced into several discs for check-up and unfortunately, two major problems were realized: 1) accumulation of air bubbles into one large bubble at the center of the specimens of low AF (about 2 or 3 cm in diameter) II) position dependency of the aggregate content in specimens with aggregate fractions less than 0.4. Therefore the moulds were heightened to 250 mm and the material of the tube was changed to plastic to reduce the weight and make the rotation possible. Also duration of the first vibration and de-airing were prolonged to extract as much air as possible. The duration of vibration phases was optimized for each concrete (i.e. for each AF). This eliminated the problem I, but could not totally eliminate the problem II. Although the aggregate content of specimens was not as position dependent as in the first mold, but, there was evidence that in the concrete with AF<0.4 the grading of the aggregate was. Despite this, it was decided to carry out the transport testing on four replicates. Since the replicates were all cut from one mold, the lower AFs and coarse/fine aggregate ratios of the top two replicates would be expected to somewhat compensate the higher AFs and coarse/fine aggregate ratios of the two bottom replicates and give the general trend of the results. Cylinders were sliced to produce the four replicates of 50 mm thick disc specimens; the end sections were discarded to avoid surface effects, each disc was tightly wrapped in more than 10 layers of cling film for curing.

TRANSPORT TESTS FOR CONCRETE

Considering the quality of the specimens it was obvious at this stage that the whole specimen production needed to be radically redesigned to overcome the problems encountered in this phase. Therefore tests for phase I were limited to the followings so that they could all be carried out on only one set of specimens:

- Electrical conduction
- Oxygen diffusion
- Oxygen permeation
- Water penetration.

The cut specimens (100 mm diameter x 50 mm depth) were unwrapped and soaked for 1 hour in distilled water.
before electrical conduction were carried out at the ages of 7, 14, 28 days. Then the same specimens were placed into desiccators to attain equilibrium at 75% RH in 2C before the rest of the tests. Specimens were placed back in desiccator after each test for one week to regain equilibrium.

The specimens took more than one year to reach the state of equilibrium stated above. This was due to large volume of specimens. Harsh drying procedures like 105 C oven drying were not acceptable in this project since they could result in huge microstructural damage that might mask the effect of ITZ on transport. The results are given as follows:

**TEST RESULTS:**

**ELECTRICAL CONDUCTION**

Fig.4 shows the continuous reduction of electrical conduction with increasing aggregate fraction at 3 different ages. The rate of change decreases, as the concrete become older. This is because all three lines have to intercept a single value when they are 100 % aggregates.

**OXYGEN DIFFUSION AND OXYGEN PERMEATION**

Fig.5 and 6 show the effect of AF on oxygen diffusion coefficient (diffusion, dry) and oxygen permeation coefficient (flow, dry) respectively for OPC concretes. As mentioned before the specimens were not homogeneous and maximum inhomogeneity has occurred in specimens concrete with AF=0.2 where the differences in coefficients of oxygen diffusion and permeation in the very top and the very bottom replicates were 134 % and 95 % respectively. Although there are nonuniformities in the graphs, but the general trend is one of decrease with AF.

**WATER PENETRATION**

The variation of water penetration with AF is plotted in fig.7 which shows a very little decrease in penetration depth with AF. Transport results can be analyzed in terms of the volume or the depth penetrated. The volume penetrated reduced dramatically with increasing AF, but the depth penetrated did not show significant variation. Specimens produced in this phase were not homogenous. This is because, the large aggregates sank faster in vibration due to their high volume/surface area ratio, (where there were less frictional force per unit weight) therefore the effect of ITZ percolation could not be detected in transport tests. Also the sizes of the specimens were very large and took about 15 months to dry.
In order to avoid the segregation of large aggregates encountered in the last phase, it was decided to produce mortars using OPC and OPC + 20% SF (20% of the volume of OPC cement was replaced with the same volume of SF) with the same w/c of 0.45 and aggregates of less than 2.36 mm.

The aggregate size distribution was the same as concrete except that aggregates larger than 2.36 mm were omitted. They were mixed at different AFs (0, 0.10, 0.20, 0.30, 0.35, 0.40, 0.45, 0.50, 0.55, 0.60 and 0.65).

SPECIMEN MANUFACTURE OF MORTARS

As explained above it was found necessary to radically redesign specimen production so that homogeneous and reproducible specimens with minimum dissimilarities can be produced. It was decided to reduce volume of the specimens by reducing their diameter in order to have more control on the timing of mixing and vibration. Therefore 30 cc plastic syringes with 21.6 mm diameter were cast in three layers and then vibrated, de-aired, capped (sealed), back vibrated, roll vibrated and finally rotated for 24 hours. Large numbers of trial specimens were made to optimize the duration and the amplitude of the different vibrations for each AF and mortar type. Eventually entirely homogeneous specimens with no air pockets inside were produced. The specimens were demoulded after 28 days and 80-mm length cylindrical specimens were cut from each syringe discarding the ends to avoid the surface effects.

CONDITIONING THE MORTAR SPECIMENS

Electrical conduction test requires the specimens to be saturated surface dry. Transport tests concerned require the specimens to be conditioned to 75 % RH at 20 C. Therefore, before the conduction measurements the specimens were saturated. After the test, the specimens were cut to produce two 35-mm long specimens. They were then washed and submerged in water for one hour to regain saturation, and then placed in a 30 C fanned oven with plenty of saturated sodium di-cromate salt underneath the specimens, to stabilize the mortar to 55 % RH. This regime was chosen to speed up the drying process. Near the stability the specimens were moved to a 20 C desiccator with sodium chloride underneath and they were monitored until stability at 75 % RH. The stability of the specific humidity is assumed when weight loss in two successive weeks is less than 1 % of the total weight loss starting form saturated surface dry up to the state of equilibrium. The whole conditioning took more than two months.

TRANSPORT TESTS FOR MORTARS

Only electrical conduction and carbonation are considered for this paper.

RESULTS

Figures 8 to 11 show that both Electrical conduction and Carbonation depth decrease with increasing AF except at a critical aggregate fraction of, (in our OPC and SF mortars) 0.35 where there have been slight temporary increases in both transport mechanisms in OPC mortars and surprisingly much more sharply, but still temporarily in SF mortars.
CONCLUSION AND SUMMARY

Transport properties of mortars are primarily dependant upon their porosities, it is very interesting therefore to notice the followings:

1. Replacement of 20% of the OPC volume by Silica Fume has not only failed to eliminate this temporary increase in transport through our mortar, but it has also exacerbated it at the same critical AF.

2. This sudden increase in transport of mortars are only temporary and they disappear after the critical AF in OPC and SF mortars and reducing pattern of transport resume in all mortars.

Both of the above points cast serious doubts over the percolation theory. Had the theory been right, then replacement of 20% of OPC by SF in mortars should have eliminated the increase in transports at the critical AF of our mortars. Also the jump in the porosity of OPC mortars after the critical AF should have stayed similar to the level at the critical AF, if not higher. Yet in the above tests transport through all OPC and SF mortars (hence their porosities) resume their decreasing pattern after the critical AF.

It may be quite plausible to have somewhat disproportionate transport increase as the AF increases from just before to just after the ITZ percolation, owing to the connection of ITZs that may let the external molecules to permeate easier. It would however be inconceivable for the porosity of the concrete or its paste to incur a substantial volume increase just because the ITZs are now touching one another.

It is suggested therefore that the jump in porosity may be completely independent of ITZ, and that increasing the volume of aggregates to the critical value may create an environment in the mortar and in the entire paste in between the aggregates that leads to this jump in porosity.

It is not clear at this stage whether the porosity of the paste beyond the critical AF stays unaffected and similar to the paste porosity just before critical AF, or that also changes to better or worse with increasing AF.

Seeking some answer to the above ambiguities is the subject of my future papers in which some more results including the porosity measurements will be presented and discussed. The author is also currently working on two factors of spacing between neighboring aggregates or Average Paste Thickness (A.P.T) and paste roughness to see if they could have something to do with the onset of this sudden increase in porosity at critical aggregate fraction in our mortars.

REFERENCES