INTERFACIAL ZONE PERCOLATION IN CONCRETE: EFFECTS OF INTERFACIAL ZONE THICKNESS AND AGGREGATE SHAPE

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ABSTRACT

Previously, a hard core/soft shell computer model was developed to simulate the overlap and percolation of the interfacial transition zones surrounding each aggregate in a mortar or concrete. The aggregate particles were modelled as spheres with a size distribution representative of a real mortar or concrete specimen. Here, the model has been extended to investigate the effects of aggregate shape on interfacial transition zone percolation, by modelling the aggregates as hard ellipsoids, which gives a dynamic range of shapes from plates to spheres, to fibers. For high performance concretes, the interfacial transition zone thickness will generally be reduced, which will also affect their percolation properties. This paper presents results from a study of the effects of interfacial transition zone thickness and aggregate shape on these percolation characteristics.

INTRODUCTION

In recent years, the importance of the cement paste-aggregate interfacial transition zone (ITZ) in influencing concrete properties has been recognized, as witnessed by this symposium's topic. It has been shown that, even when air voids are not considered, concrete must still be considered as a three-phase material (paste, aggregate, and ITZ) to properly investigate elastic properties [1, 2], transport coefficients [3], and drying shrinkage [4]. Consideration of the ITZ is important because the microstructure of the ITZ in ordinary concrete is different than that of the bulk paste. It contains more and larger pores and less anhydrous cement and calcium silicate hydrate gel [5, 6]. Therefore, its response to mechanical and thermal loadings and chemical ingress will also differ from that of bulk paste.

The magnitude of the influence of these microstructural differences on the composite's properties depends on two parameters: the relative difference in properties between ITZ and bulk paste, and the volume fraction and connectivity (percolation) of the ITZ regions [3]. This paper addresses only the latter of these topics, studying the influence of ITZ thickness and aggregate particle shape on the percolation of ITZ in a mortar or concrete. Experimental evidence of this phenomenon of interfacial transition zone percolation has been provided by mercury intrusion porosimetry studies [7, 8, 9] and by experimental comparisons of the transport properties of mortars with those of their constituent cement pastes [10].

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COMPUTER MODEL

The computer model employed in this study is an extension of that employed in the study described in [8] in which particle shapes were limited to spheres. Based on recent analytical work [11], a new computer program was written to extend the model to triaxial ellipsoids, with semi-axes of lengths a, b, and c and any arbitrary orientation. Therefore, one may vary particle shapes from spheres (air voids) to cylindrical ellipsoids (fibers) to triaxial ellipsoids (aggregates). Given any size distribution of particles and scale factors a, b, and c, the program places a requested number of particles in a three-dimensional cubic volume, typically 10 mm on a side, such that no particles overlap. The program proceeds from the largest to the smallest particles. Interfacial transition zones are modelled as "soft shells" of a constant thickness t surrounding each "hard core" aggregate particle [8]. The ITZ may overlap one another and also may intersect the aggregate particles. After all particles have been placed, the program uses a burning algorithm [12] to determine the connectivity of the ITZ regions and volumetric point sampling to determine the volumes occupied by aggregates, bulk cement paste, percolated ITZ regions, and disconnected ITZ regions. By varying the number of particles placed in the box, the percolation behavior can be mapped out as a function of aggregate content.

The computer program was validated by determining the excluded volume of triaxial ellipsoids of different aspect ratios [13]. The excluded volume for an ellipsoid is the volume surrounding the ellipsoid where the center of an arbitrarily oriented second identical ellipsoid cannot be placed without some portion of the second ellipsoid overlapping a portion of the first one. For a variety of aspect ratios, the excluded volumes determined using the computer program, by sampling 500,000 random configurations, were within 0.5% of the exact analytical result [14].

In this study, three interfacial transition zone thicknesses (10, 22.5, and 50 μm) and five combinations of aspect ratios (1:1:1, 1.25:1:0.8, 1.5:1:0.667, 2:1:0.5, 1.59:0.794:0.794) were studied. The aspect ratios were chosen to maintain a constant particle volume ($a^*b^*c=1$) while varying the aggregate surface area. Surface areas were calculated using formulas presented in [15], which can also be found more recently in [16]. In a given execution of the simulation, the aspect ratio was held constant, but it could be easily varied to follow some distribution of particle shapes. The aggregate particle size distribution (PSD) for the mortars corresponded to that of a typical sand used in concrete, as measured experimentally in [17]. The particle diameters ranged between 75 and 4750 μm. For the 1000 mm^3 cubic volumes used in this study, between seven and twenty thousand aggregate particles were required to represent an actual mortar. Fig. 1 shows two-dimensional slices from three-dimensional microstructures for systems with 16,000 particles (aggregate volume fraction=43%), an ITZ thickness of 50 μm, and the two aspect ratios indicated.

RESULTS AND DISCUSSION

The ITZ thickness, t, of 22.5μm was chosen as an estimate of a typical thickness to be encountered in a mortar or concrete [8]. Fig. 2 shows a plot of the fraction of "ITZ paste" percolated vs. the aggregate content for the five aggregate particle aspect ratios investigated in this study. One clearly notices that as the aggregates become more elongated (higher aspect ratios), significantly lower aggregate contents are needed to form a percolated ITZ path across the microstructure. This result is in agreement with percolation studies of totally overlapping particles [13, 18]. Interestingly, when one plots these results against

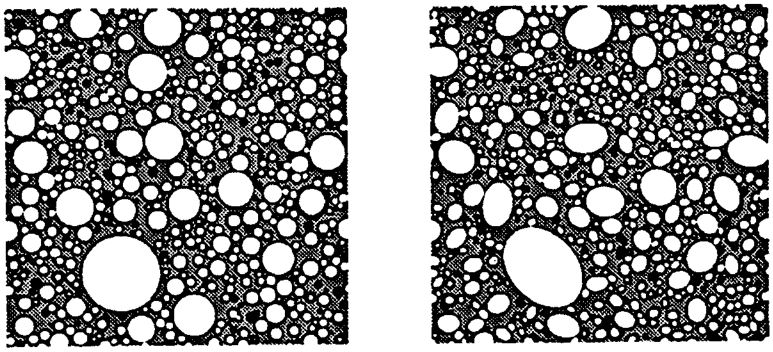

Figure 1: Two-dimensional slices from three-dimensional microstructures for aspect ratios of 1:1:1 (left) and 1.25:1:0.8 (right) (white: aggregate, grey: bulk cement paste, black: ITZ cement paste).

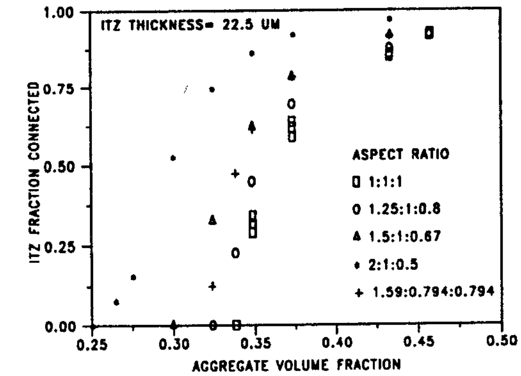

Figure 2: Percolated interfacial transition zone fraction vs. aggregate content. Three data sets are shown for the 1:1:1 aspect ratio system to provide some indication of variability.

the ITZ volume fraction (or the aggregate surface area), the results for the different aspect ratios appear to converge to a single curve as shown in Fig. 3. This suggests that for a given aggregate PSD and ITZ thickness, it is the volume fraction of ITZ paste, and not the aggregate content, which uniquely determines the percolation behavior of these systems.

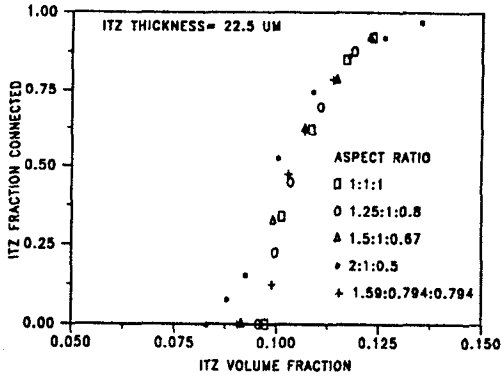

Figure 3: Percolated interfacial transition zone fraction vs. interfacial transition zone volume fraction.

Results for the three ITZ thicknesses are summarized in Table I. As would be expected, decreasing t increases the aggregate content necessary to achieve percolation, but decreases the interfacial transition zone volume needed. While plotting ITZ fraction connected against ITZ volume fraction for different aspect ratios gives a universal curve for all three ITZ thicknesses studied, plotting against aggregate surface area (Fig. 4) generates universal curves only for the two thicker interfacial transition zones. For the 10 μm ITZ thickness (dotted lines in Fig. 4), the plot against aggregate surface area exhibits a greater degree of dispersion which contrasts with the results obtained for t values of 22.5 and 50 μm. Since to a first order, ITZ volume is proportional to aggregate surface area, this suggests that the volume fraction occupied by regions composed of two or more overlapping ITZ may be greater for the systems with the smallest ITZ thickness. For example, if the average width of overlapping interfacial transition zone regions remains relatively constant as t varies, the fraction of total ITZ volume contained in overlaps will increase with decreasing ITZ thickness (e.g., 2 μm out of 10 μm vs. 2 μm out of 50 μm).

TABLE I: Volume Fractions at Percolation vs. Interfacial Transition Zone Thickness

ITZ Thickness (μm)	Agg. Vol. Frac. Range(%)[a]	ITZ Vol. Frac.(%)[a]
10	44-50	6
22.5	30-37	10
50	16-24	16

[a]Values for fraction connected = 50%

A third scenario which can be addressed using this simulation model is the effect of aggregate PSD on ITZ percolation. Using spherical particles and a t value of 22.5 μm, Fig. 5 shows the effects of removing the smallest particles (< 150 μm in diameter) from the original aggregate PSD. Physically, this would be equivalent to sieving the aggregate and discarding

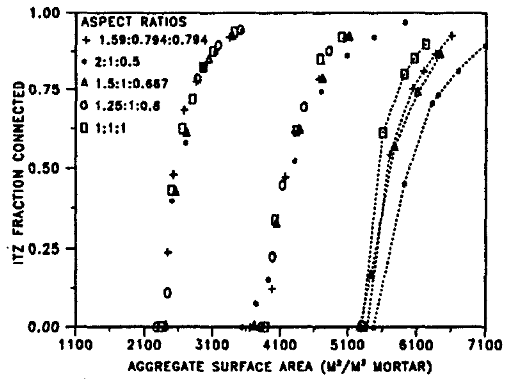

Figure 4: Percolated interfacial transition zone fraction vs. aggregate surface area for t=50, 22.5, and 10 (dotted lines) μm from left to right.

the portion which passed through a sieve with an opening size of 150 μm. In coarsening the aggregate PSD, one finds that a greater volume of aggregate, but a smaller volume of ITZ paste (indicated by the points connected by dotted lines in Fig. 5) is needed to percolate the system. In this case, one can imagine maintaining percolation by replacing an already percolated cluster of small aggregate particles by a single larger aggregate particle, which would indeed increase aggregate volume while somewhat decreasing ITZ volume.

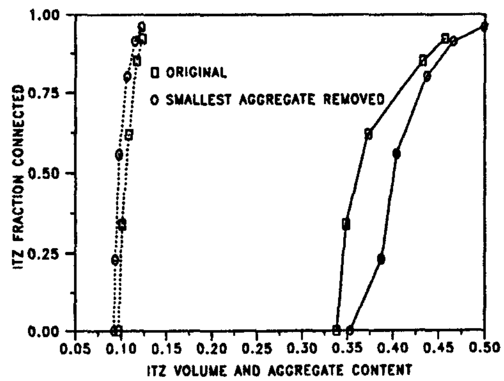

Figure 5: Percolated interfacial transition zone fraction vs. ITZ volume (dotted lines) and aggregate volume fraction (solid lines) for t=22.5 μm showing effect of removing finest aggregate particles.

CONCLUSIONS

A computer experiment has been performed to study the effects of interfacial transiti zone thickness and aggregate shape on the percolation of ITZ in mortars. Both paramete can have significant effects on the percolation characteristics of these systems. For a giv aggregate particle size distribution and ITZ thickness, the curve of ITZ fraction connected ITZ volume fraction was found to be relatively independent of particle shape, while elongat aggregate shapes resulted in lower aggregate contents being required for ITZ percolation th spherical ones. Future experimental studies will concentrate on a systematic assessment the effects of ITZ connectivity on transport coefficients, such as ionic diffusivity, permeabili and sorptivity, both for mortars made with typical sands and with spherical aggregates.

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