Cement and Concrete Research 40 (2010) 1601–1608

Contents lists available at ScienceDirect

Cement and Concrete Research

journal homepage: http://ees.elsevier.com/CEMCON/default.asp

An investigation into the influence of inter-aggregate spacing and the extent of the ITZ on properties of Portland cement concretes

Prasada Rao Rangaraju [a], Jan Olek [b], Sidney Diamond [b,*]

[a] *Department of Civil Engineering, Clemson University, Clemson, SC 29634, USA*
[b] *School of Civil Engineering, Purdue University, 550 Stadium Mall Drive, West Lafayette, IN 47907, USA*

ARTICLE INFO

Article history:
Received 28 June 2008
Accepted 7 July 2010

Keywords:
Interfacial transition zone (B)
Mean inter-aggregate spacing
Compressive strength (C)
Elastic moduli (C)
Transport properties (C)

ABSTRACT

Conventionally-designed concretes were prepared with different sand particle size distributions, so as to systematically vary the extent of aggregate-cement paste interface and the mean spacing between sand grains. The range of fineness modulus of the sands fully encompassed the range of sands normally used in concretes. The concretes were batched at w/c ratios of 0.30 and 0.50 and cured for various periods before carrying out determinations of mechanical properties and of "rapid chloride permeability". The conventional notions of the effect of the ITZ on concrete properties would predict that a reduction in strength and an increase in chloride permeability would accompany increased ITZ interfacial area and closer spacing between sand grains. In general, no such influence was found. It appears from this research that the traditional notions of the adverse influence of the ITZ on the properties of conventional concretes may not be accurate, within the realms of conventional concrete and typical inter-aggregate spacings.

1. Introduction

Conventional Portland cement concrete is a multi-phase composite material containing appropriate proportions of coarse and fine aggregate embedded in a coherent matrix of hydrated cement paste. Cement hydration is almost always incomplete, and the cement paste consists of hydration products, residual unhydrated portions of cement grains, and pore space.

The cement paste fraction of this composite is often considered to consist of two distinct portions, i.e. a relatively porous interfacial transition zone (ITZ) extending generally to ca. 30 μm or more away from the contact with the aggregate surfaces, and less porous "bulk" paste beyond this zone. An authoritative overview of this concept has been provided by Scrivener [1] The ITZ is said to constitute the "weak link" in conventional concretes [2–4], and it has been suggested that overlap between adjacent porous ITZs for closely-spaced aggregates (so-called "percolation") provides preferential paths for transmission of water and ions [4–6]. Furthermore, several models have been developed in predicting the elastic modulus of concrete using a 3-phase composite material approach, in which the ITZ phase has been ascribed a uniform thickness with unique properties [7–9]. No distinction is usually made between ITZs associated with coarse aggregate and ITZs associated with sand grains; microstructural studies, for example that of Diamond and Huang [10], have found little difference between them. Since collectively the sand fraction of a conventional concrete exhibits much greater area in contact with cement paste than does the coarse aggregate fraction, most of the ITZ in concrete is necessarily associated with the sand grains.

If the conventional picture of the effects of the ITZ on concrete properties is correct, the proportion of "ITZ paste" volume to "bulk paste" volume, and the spacing between adjacent aggregate grains should be major determinants of concrete properties. The average distance between adjacent aggregate grains can provide something of a numerical index to these properties, since concretes with larger, more widely-spaced aggregates presumably possess proportionately less "ITZ paste" and proportionately more "bulk paste" per unit volume.

Some years ago Diamond et al. [11] measured inter-aggregate spacings on polished concrete surfaces for mixes characteristic of the range of normal field concretes. They found that the closest distance between adjacent aggregate grains on a planar surface averaged about 75 μm to about 100 μm, depending primarily on the fineness of the sand used. Since the traditional thickness ascribed to the ITZ is of the order of 30–50 μm, these findings suggested that in many normal concretes a significant proportion of the paste will fall close enough to an aggregate surface to be ascribed to "ITZ paste"; for concretes batched with finer sands the individual "ITZ paste" zones surrounding adjacent closely-spaced aggregates might effectively overlap each other.

It should be noted that (1) both "ITZ" and "bulk" paste are quite inhomogeneous on a fine scale, and that (2) on average, within the ITZ, the porosity exhibits a statistical gradient outward from the actual paste-aggregate interface. Nevertheless, it is often considered that the

---

* Corresponding author. Tel.: +1 765 494 5016; fax: +1 765 494 0395.
E-mail address: diamond@ecn.purdue.edu (S. Diamond).

0008-8846/$ – see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.cemconres.2010.07.002

paste in the ITZ zone has distinct properties different from the properties of the "bulk" paste.

It is possible to influence the proportions of "ITZ paste" to "bulk paste" (and the extent of overlap between adjacent ITZs) in concretes. One way of doing this is to incorporate sands of different particle size distributions (within the acceptable range for concrete sands) while at the same time keeping the paste content and the proportions of sand and coarse aggregate constant. Under these conditions concretes prepared with finer sands necessarily exhibit a greater extent of aggregate-paste interface per unit volume. At the same time, as the number of sand grains per unit volume increases, the average spacing between adjacent sand grains is necessarily reduced. If the traditional notions of the "weak-link effect" of the ITZ on mechanical properties are meaningful, and if percolation of closely-spaced ITZs affects the transmission of water and ions through the concrete, such variations should produce measurable effects on the properties of the concrete.

In the present study two groups of conventionally-designed concrete mixes were prepared to span the common range of w/c ratios, w/c ratio values of 0.30 and 0.50 being selected. Within each group the proportions of coarse and fine aggregates were kept constant as was the relative volume of paste to aggregate, but the particle size distribution of the sands employed was varied over the full range of conventional concrete sand size gradations. At one extreme, a coarse sand was used which would produce only limited sand surface areas, thus limiting the extent of the ITZs generated. At the other extreme, a fine concrete sand was used which would present much more total surface to the cement paste, and produce much more ITZs, and also smaller average distances between sand grains.

The concretes were mixed in a conventional laboratory concrete mixer, and specimens were compacted into steel molds and cured at 100% RH after being demolded at 1 day. After various periods of hydration a number of standard concrete tests were carried out to assess the effects of varying the particle size distribution of the concrete sand on the mechanical and other properties of the concretes.

It should also be noted that in these studies no supplementary cementitious materials such as silica fumes were used. Therefore, no modification of the ITZ due to the so-called "micro-filler" effect could have occurred.

2. Experimental details

2.1. Materials and mix designs

2.1.1. Cement

A single ASTM Type III Portland cement was used throughout. The chemical composition and associated properties provided by the cement manufacturer are: SiO_2 — 21.53%, Al_2O_3 — 5.07%, Fe_2O_3 — 2.31%, CaO — 65.69%, MgO — 1.14%, SO_3 — 2.5%, Na_2O — 0.07%, K_2O — 0.36%, LOI — 1.23%, insoluble residue — 0.4%, and Blaine fineness — 346 m^2/kg.

2.1.2. Coarse aggregate

The coarse aggregate used was a crushed dolomite of excellent strength-producing property; its measured specific gravity was 2.71, and the maximum aggregate size was 19 mm. Its external surface area was measured by a permeability method using the Kozeny–Carman equation [12]; the value obtained was 0.23 m^2/kg.

2.1.3. Sand

The sand fractions incorporated in the various concretes were derived from a natural sand by sieving it into five different size classes, and then blending the size classes so as to produce graded concrete sands of three different particle size distributions. The size gradations and other properties of the three blended sands used in the concretes (designated FA #1, 2, and 3, respectively) are provided in Table 1.

Table 1
Gradations, specific surface areas, and the calculated mean IAS values of the sands used in these concretes.

Sieve size range (mm)	Specific surface area, m^2/kg	Sand blend notation		
		FA #1% retained	FA #2% retained	FA #3% retained
#4–#8 (4.75–2.36)	1.00	0	0	0
#8–#16 (2.36–1.18)	2.08	0	19	50
#16–#30 (1.18–0.6)	4.30	11	38	35
#30–#50 (0.6–0.3)	7.54	36	33	15
#50–#100 (0.3–0.15)	13.54	53	10	0
Fineness modulus		1.58	2.65	3.35
Spec. surf. area, m^2/kg		10.36	5.87	3.67
Calculated inter-aggregate spacing, μm		60	100	160

The surface areas measured for these blended sands by the permeability method [12] were respectively 3.67 m^2/kg, 5.87 m^2/kg, and 10.36 m^2/kg. It is apparent that the surface area of the coarse aggregate (0.23 m^2/kg) is negligible in comparison to even the coarsest sand, and that nearly all of the paste-aggregate interfaces in the resulting concretes necessarily comprise interfaces with sand grains rather than with coarse aggregate particles.

The size gradation of sand for use in concrete is commonly characterized by a "fineness modulus" value, which is actually an index of coarseness. The usual range of fineness modulus values as given by Mindess et al. [2, p.130] is between 2.3 and 3.1. The fineness modulus values of the three blended sands used here were respectively 1.55, 2.65, and 3.35, thus encompassing (and indeed, somewhat exceeding) the range of gradations normally used in concrete.

2.1.4. Mix designs

Details of the mix designs of the various concretes studied here are provided in Table 2. As mentioned previously, concretes incorporating each of the sand blends were prepared at w/c ratios of 0.50 and 0.30 respectively. The mass ratio of coarse-to-fine aggregates was kept constant at 1.65 throughout, and the calculated percent paste by volume kept constant at 40%. Appropriate dosages of naphthalene sulfonate superplasticizer were used so as to generate slumps between 75 and 100 mm in each of the fresh concretes.

2.2. Mixing and specimen preparation

The concretes were mixed in a conventional laboratory mixer, and 76 mm × 152 mm cylinder specimens were compacted into steel molds and demolded at 1 day. They were subsequently cured in a fog room maintained at nominally 100% RH and approximately 23 °C until testing.

Table 2
Concrete mixture proportions.

Item	Mixture proportion					
	Series #1, w/c = 0.30			Series #2, w/c = 0.50		
	FA #1	FA #2	FA #3	FA#1	FA #2	FA #3
Cement, kg/m^3	590	590	590	451	451	451
Sand, kg/m^3	606	606	606	606	606	606
Coarse agg., kg/m^3	997	997	997	997	997	997
Water, kg/m^3	173.3	173.3	173.3	224.8	224.8	224.8
Air, %	3	3	3	3	3	3
Superplasticizer, L/m^3	7	7	7	2.5	2.5	2.5

2.3. ITZ characteristic and mean inter-aggregate spacings

If the average thickness of the ITZ around any sand grain does not vary with its size, as found, for example by Diamond and Huang [10], the differences in surface areas among the sand blends should result in proportional differences in content of "ITZ paste" per unit volume of concrete. Furthermore, since the total paste content per unit volume in these mixes is fixed, the corresponding differences in the *proportions* of "ITZ" paste" to "bulk paste" is necessarily considerably greater than this. Assuming the overall concrete porosity remains constant, the porosity of the "bulk paste" may be reduced as the proportion of the "ITZ paste" increases.

An index of the nominal proportion of "ITZ paste" to "bulk paste" in different concretes can be provided by calculating a mean inter-aggregate spacing (mean IAS) for each concrete. This can be done using the mix proportions, the dry-rodded unit weights of the aggregate blends, and the specific surface area of each of the individual fractions of aggregate. The procedure for this calculation is illustrated in Appendix A, and the necessary data for the present concretes is provided in Table 3. Since the paste-to-aggregate volume ratio is the same for both series of concretes the calculated mean IAS for concrete using a given sand is the same for concretes of both the w/c 0.50 and w/c 0.30 series.

The mean IAS so calculated is similar but not quite the same as the mean distance of closest approach measured by Diamond et al. [11]. It is similar in that it attempts to represent the average value of closest approach between adjacent grains. However, it is calculated from a simplified model and represents distances averaged over all directions in three dimensional space, rather than distance of closest approach measured on an arbitrary plane.

The results of these calculations indicated that the mean IAS in the concretes with the three different sand gradations would be approximately 160 μm, 100 μm, and 60 μm for the concretes containing the coarsest, medium, and finest sands. These values are of the same general magnitude as the measurements reported by Diamond et al. [11].

To verify the degree to which these calculated values of mean IAS were actually characteristic of the present concretes, a series of backscatter SEM micrographs were taken from flat polished specimens. Measurements were made of mean inter-aggregate spacings on the plane of observation. These were measured using a simple linear traverse procedure similar to that recommended in the ASTM C 457 standard procedure. In essence, this involved overlaying of a grid of closely-spaced lines on the SEM images and determining the respective total lengths traversed respectively on aggregate and paste fractions, at the same time counting the number of aggregate particles encountered during the traverse. The mean IAS on the plane was calculated simply by dividing the total length of paste encountered along a given traverse by the number of aggregates detected along the traverse line. Details of this procedure have been described elsewhere [13].

In Table 4 these measured mean IAS values are compared with the calculated values of mean IAS. In each case, the mean measured values of the IAS slightly exceed the calculated values. This would be expected, considering that the separations between aggregates were measured along an arbitrary line, rather than representing average distances of closest approach. Nevertheless, the agreement is quite satisfactory.

Table 3
Aggregate properties required for calculation of mean IAS.

	Specific gravity, ssd	Dry-rodded unit weight, kg/m^3	Specific surface area, m^2/kg
Coarse aggregate	2.71	1645	0.23
Fine aggregate			
Blend FA #1	2.63	1715	10.36
Blend FA #2	2.63	1735	5.87
Blend FA #3	2.63	1645	3.67
Mixture (CA/FA — 1.6)			
Blend FA #1 — CA	2.68	1908	4.12
Blend FA #2 — CA	2.68	1961	2.39
Blend FA #3 — CA	2.68	2016	1.55

3. Details of concrete testing

A number of standard concrete tests were carried out to investigate the degree to which difference in the ITZ contents and the mean spacings between aggregates might influence concrete properties, as suggested by conventional ideas of ITZ effects. These determinations included compressive strength according to ASTM C 39, tensile strength as evaluated by the split tensile test method (ASTM C 496), dynamic modulus of elasticity (as measured by impulse excitation method per ASTM E 1876), and measurement of relative permeance by the so-called "rapid chloride permeability" test (ASTM C 1202). Each of these determinations was carried out at various ages, on three replicate specimens, with the value reported being the mean of the set. Microscopic determinations of the air contents (ASTM C 457) and measurements of the water absorption (ASTM C 642) were also carried out for each of the concretes, in order to determine whether any differences found between them might be due to differences in air content or in water absorption.

4. Results

4.1. SEM observations illustrating inter-aggregate spacings

Representative SEM micrographs are provided in Fig. 1 to illustrate the differences in typical aggregate spacings between concretes made with the coarsest and the finest sand blends. The images were taken from 28 day-old concrete of w/c 0.30. The sand grains are of various shades of gray, reflecting compositional differences among different individual sand grains in this natural sand. The residual unhydrated cement particles are seen as smaller white grains, the cement hydration products (primarily C–S–H) are gray, and the air voids and other pores are black. It is evident that the great differences between the mean inter-aggregate spacings calculated for the two concretes do in fact reflect the substantial differences that exist in their internal structures.

4.2. Comparisons of concrete properties at 28 days

4.2.1. Compressive strength

Compressive strength values obtained at 28 days are plotted vs. the calculated mean IAS values in Fig. 2, for the both w/c 0.30 and 0.50 concrete series. The error bars are indications of the standard deviation.

The 28-day compressive strengths were relatively high, presumably due to the mix design, the use of Type III cement, and the quality of the aggregate. The values found were of the order of 50 MPa for the w/c 0.50 concretes and 85 MPa for the w/c 0.30 concretes, the

Table 4
Comparison of calculated vs. measured mean inter-aggregate spacings.

Sand blend	Inter-aggregate spacing, μm		
	Calculated	Measured (SEM method)	% difference
FA #1	60	68	13
FA #2	100	122	22
FA #3	160	200	25

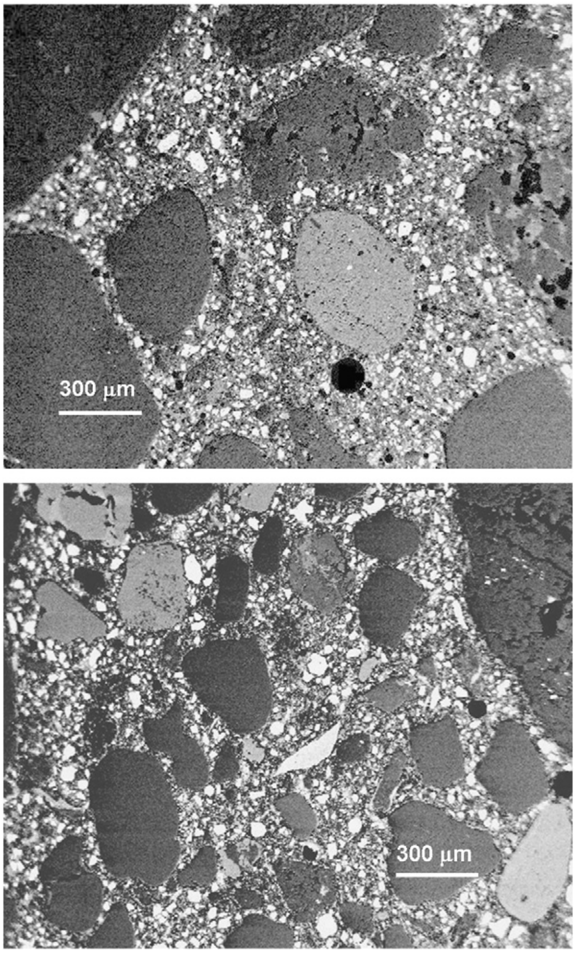


Fig. 1. SEM micrographs illustrating the difference between typical inter-aggregate spacings for concrete specimens made with the coarsest sand (above) and with the finest sand (below).

difference illustrating the overriding effect of w/c ratio on strength. It should be recalled that no silica fume or other potential “ITZ filler” was employed.

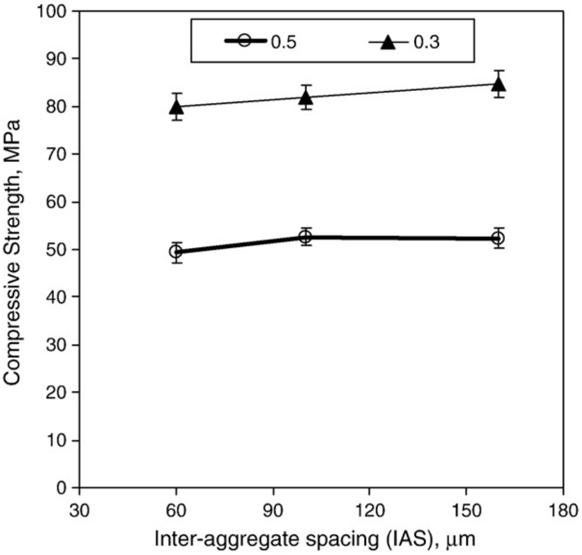


Fig. 2. Compressive strengths of concrete at 28 days vs. calculated mean inter-aggregate spacing. The range bars indicate one standard deviation.

It is clear from Fig. 2 that there is no appreciable effect of the differences in inter-aggregate spacing on compressive strength for the w/c 0.50 concretes. For the w/c 0.30 series, there is a slight apparent effect, i.e. the mean compressive strength of the concrete with the finest sand (and thus the greatest proportion of “ITZ paste” to “bulk paste”) was a little lower (at about 80 MPa) than the mean compressive strength for the coarsest sand concrete (at close to 84 MPa). However, we consider that this small difference is of dubious significance, and, as shown later in Table 6, similar differences were not found at either earlier or later ages.

4.2.2. Split tensile strength

The split tensile strength values determined at 28 days are presented in Fig. 3. The values obtained were between about 5 to 6 MPa for the w/c 0.50 series and about 8 to 9 MPa for the w/c 0.30 series. Here there seems to be a fairly obvious trend toward reduced tensile strength for the concretes with the finer (and more closely-spaced) sands. However, this trend is not found either at earlier or at later ages, as seen later in Table 6. To illustrate the difference, Fig. 3B shows the split tensile

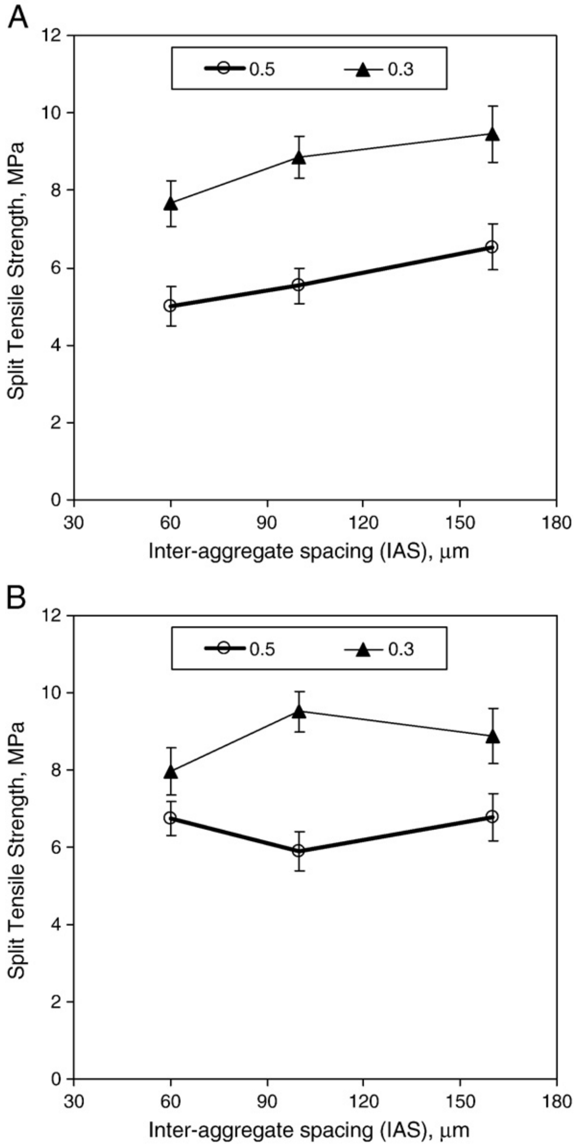


Fig. 3. A. Split tensile strength of concrete at 28 days vs. calculated mean inter-aggregate spacing. B. Split tensile strength of concrete at 56 days vs. calculated mean inter-aggregate spacing.

strength values at 56 days, this apparent trend at 28 days is not substantiated at 56 days or at 7 days.

4.2.3. Dynamic modulus of elasticity

Results of the measurements of the dynamic modulus of elasticity at 28 days are provided in Fig. 4. The values obtained were consistently 43 GPa for the w/c 0.50 series and 53 GPa for the w/c 0.30 series. Here there clearly is no detectable effect of the inter-aggregate spacing on the results, which are remarkably consistent for a given w/c ratio.

4.2.4. "Rapid chloride permeability" test results

The "rapid chloride permeability test" (ASTM C 1202), while not actually a measure of permeability, provides an acceptable comparative index of the relative permeance of a given concrete, i.e. the relative ease of movement of water and ions through it.

One of the fundamental assumptions of the conventional picture of the effects of the ITZ is that the movement of water and ions through the "ITZ paste" is much more rapid than through the "bulk paste". More specifically, it is usually considered that when aggregates are sufficiently closely-spaced that the adjacent ITZs "percolate", i.e. overlap in space, fluid and ion transport is significantly enhanced [5,6].

The results of the "rapid chloride permeability" tests at 28 days are provided in Fig. 5. The values obtained were of the order of 6200 coulombs for the w/c 0.50 concretes, and about 3500 coulombs for the w/c 30 concretes. In neither series is there any indication of a greater permeance for concrete with even the smallest inter-aggregate spacing. Indeed, the values for the w:c 0.30 series show an apparent trend in the *opposite* direction from that expected, i.e. the concrete with the more widely-spaced aggregates exhibited slightly higher "rapid chloride permeability" values. If real, this small difference may reflect greater interference in the flow path by the more closely-spaced impervious sand grains, i.e. a more tortuous flow path.

4.2.5. Air content and water absorption determinations

Measurements of the air contents of the hardened concretes (ASTM C 457), and of water absorption (ASTM C 642) were carried out at 28 days; the results are presented in Table 5. These tests were carried out to establish whether or not incorporation of the sands of different gradations might have produced differences in amount of entrapped air (or in water absorption characteristics) that might have offset or overcome the effects of the differences in ITZ characteristics. No such effects were found. The air contents for both series were all reasonably close to the expected mix design value of 3%. The measured absorption values were approximately 6% for the w/c 0.30 series and slightly more than 7.5% for the w/c 0.50 series, irrespective of the gradation of sand used.

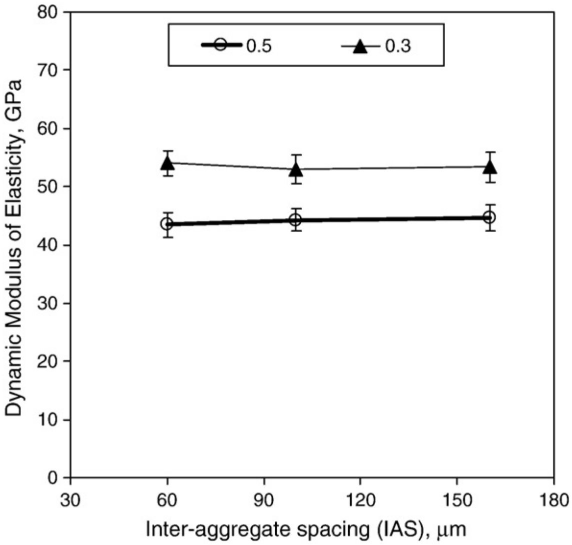


Fig. 4. Dynamic modulus of elasticity of concretes at 28 days vs. calculated mean inter-aggregate spacing.

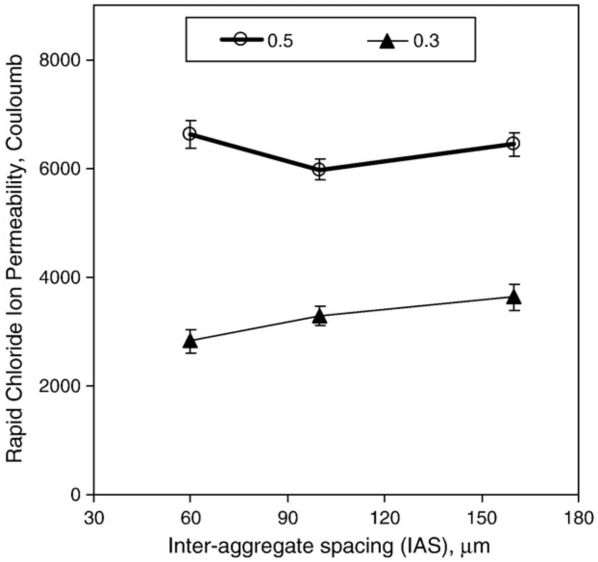


Fig. 5. "Rapid chloride permeability" of concretes at 28 days vs. calculated mean inter-aggregate spacing.

4.3. Concrete properties measured at earlier and later ages

The results cited previously were all obtained at a standard age of 28 days. It is axiomatic that concrete properties change over time; strength increases, and fluid and ion transport properties decrease, as hydration proceeds. Accordingly, in this study tests of concrete mechanical properties were conducted at ages earlier, and also later, than 28 days. The results obtained at 7 days and generally at 70 days are summarized in Table 6.

Examination of the results in Table 6 indicates that for comparisons at ages earlier and later than the conventional 28 day, the concretes of a given w/c ratio did not show any consistent differences in compressive strength, split tensile strength, dynamic modulus of elasticity, or "rapid chloride permeability" with progressive differences in sand gradations. In particular, the seeming trend found for split tensile tests at 28 days did not appear either at 7 days or at 56 days. Table 6 also indicates that the concretes all gained strength and showed reduced values of "rapid chloride permeability" with age as expected.

Table 5
Results of air content and water absorption determinations.

	Fine aggregate blend used in the mix	ASTM C-457 Air Content, %	ASTM C 642 Absorption, %
w/c 0.30	FA #1	2.70	5.85
	FA #2	2.94	6.05
	FA #3	3.03	6.10
w/c 0.50	FA #1	3.5	7.52
	FA #2	3.0	7.86
	FA #3	3.6	7.73

Table 6
Measured properties of concretes at ages other than 28 days.

Age, days	Property	w/c ratio	Calculated IAS		
			60 µm	100 µm	160 µm
7	Compressive strength, MPa	0.3	68.4	70.0	69.5
		0.5	34.0	36.5	36.8
	Split tensile strength, MPa	0.3	7.80	7.93	7.27
		0.5	5.85	5.94	5.83
	Dynamic elastic modulus, GPa	0.3	51.0	49.5	50.0
		0.5	39.1	38.8	40.8
	Chloride permeability, Coul.	0.3	3300	3400	3800
		0.5	7500	7200	7400
70[a]	Compressive strength, MPa	0.3	88.6	90.7	94.0
		0.5	55.0	59.0	59.0
	Split tensile strength, MPa	0.3	7.97	9.51	8.88
		0.5	6.75	5.89	6.78
	Dynamic elastic modulus, GPa	0.3	55.8	54.7	55.4
		0.5	45.2	46.0	47.0
	Chloride permeability, Coul.	0.3	2380	2600	2490
		0.5	4230	3590	4250

[a] Except for the split tensile strength determinations, which were secured at 56 days.

5. Discussion

The results obtained here are interpreted as casting doubt on the widely-accepted concept that the ITZ paste acts as a significant "weak link" with respect to concrete properties, since varying the extent of the interface and the presumed degree of overlap of adjacent ITZs does not seem to affect them, at least over the range of conventional concretes.

It is possible that some details of the ITZ itself may be influenced by inter-aggregate spacing; for example the average gradient of porosity between aggregate interface and "bulk" paste may be greater as the proportion of "bulk" paste is reduced. Nevertheless, the almost complete lack of measured effect on concrete properties is significant.

A number of so-called 'three phase' models of concrete have been developed in recent years, for example [7–9,14,15]. These models treat the concrete as a system composed of aggregates, "normal" cement paste, and "ITZ paste", each treated as a separate component with different properties. In these models significantly reduced properties, particularly elastic modulus values, are usually assigned to the "ITZ paste" component. We found no indication of any reduction in dynamic elastic modulus associated with the presumed increase in proportion of ITZ associated with fine sands. The results provided here appear to be in agreement with the comments of LeRoy and deLarrard [16] casting doubt on the necessity for such "three phase model" treatments, at least within the range of normal concretes.

6. Conclusions

Two series of concretes were prepared over the conventional range of concrete w/c ratios (0.30 and 0.50) to test the concept that the extent of the ITZ (and degree of overlap of adjacent ITZs) significantly influences concrete properties. Within each series, concretes were prepared at constant paste content, but with different sand particle size distributions, collectively covering the full range of sand size distributions normally used in concrete. At one extreme, the coarse sand used had only a small surface area, thus limiting the content of "ITZ paste"; with this sand the mean spacing between sand grains was large, and little overlap could have existed between the ITZs of adjacent sand grains. At the other extreme, the fine sand used had a large surface area and consequently much of the cement paste present was "ITZ paste" as conventionally described; since the average spacing between sand grains was so small, extensive overlap should have existed between the ITZs surrounding neighboring sand grains, at least under conventional ITZ assumptions.

It was found that measured compressive strengths, dynamic elastic moduli, and "rapid chloride permeability" values were all unaffected by these extreme variations in ITZ contents and with extent of overlap of neighboring ITZs. This was true at all ages tested (ranging from a week to more than two months), and was equally true for both w/c 0.30 and w/c 0.50 concretes. The only exception found was a modest reduction in split tensile strength with reduced inter-aggregate spacing which was observed at 28 days, but not at 7 days or 56 days. Thus varying the ITZ content by varying the fineness of the concrete sand between extremes consistent with normal concrete practice did not appreciably influence most measured concrete properties.

These results are interpreted as casting doubt on the conventional notion that ITZs act as significant 'weak links' in conventional concretes. The use of so-called "three component" models of concrete, in which the ITZ paste is treated as a separate phase and assigned different properties from the remainder of the cement paste does not appear to be justified.

Acknowledgments

The results presented here were taken from the Ph. D. thesis of the first-named author at Purdue University [13], which was carried out under the direction of the second-named author. The authors acknowledge the financial support provided by Purdue Research Foundation. Thanks are also extended to Ms. Janet Lovell, Mr. Luis Graf-Noriega, Mr. Bing Tian and Dr. Jingdong Huang, for technical assistance.

Appendix A. Method of calculation of mean inter-aggregate spacings

This calculation involves a number of assumptions, applied to measured parameters, specifically the dry-rodded unit weight of the aggregates, the specific surface areas of the sand and coarse aggregate fractions, and their mass proportions. All calculations are made on the basis of 1 m^3.

The simplifying assumption is first made that all of the aggregate grains are spherical.

In the first step of the calculation the total volume (Vdry) that would be occupied by the dry-rodded aggregate (coarse aggregate and sand blended in proper proportions) is calculated. This is done by dividing the sum of the aggregate weights by the dry-rodded unit weight. This dry-rodded volume includes the actual volume of aggregates in 1 m^3 plus the total of the spaces between the aggregate particles in the dry-rodded condition. The inter-aggregate pore space in the dry-rodded condition is calculated by Eq. (1), viz:

$$\eta = [Vdry-(W/SGssd)]/Vdry \tag{1}$$

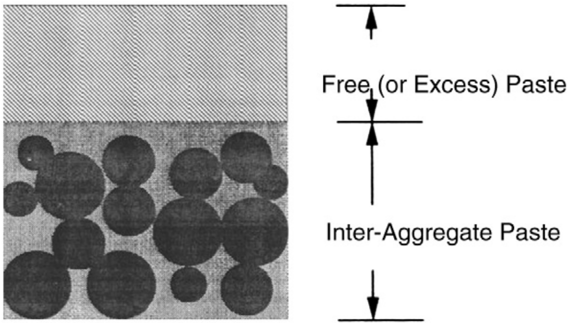
where:

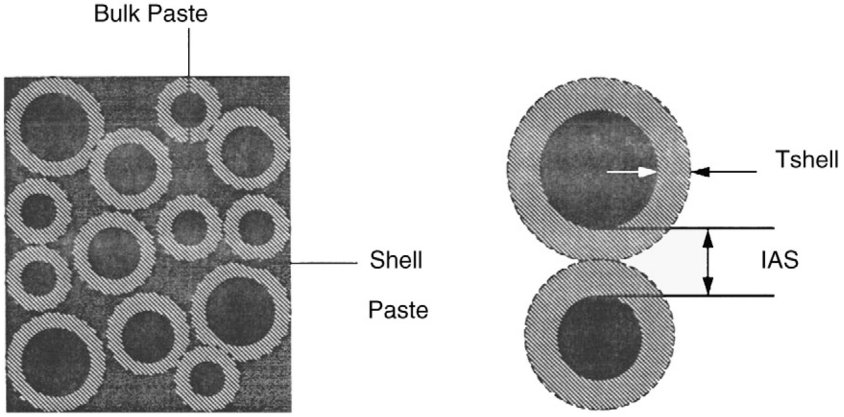
η	is the total pore space between the aggregate in the dry-rodded condition,
Vdry	is the total volume occupied by the dry-rodded blended aggregate,
W	is the weight of the aggregate (i.e. combined coarse aggregate and sand),

S is the specific surface area of the combined coarse aggregate and sand, and
SGssd is the specific gravity of the blended aggregate in the saturated surface dry condition.

Conceptually the volume of cement paste (i.e. cement plus water) is then introduced into the dry-rodded aggregate, in two steps. In the first step, the cement paste just fills the pore space between the aggregate grains in the dry-rodded condition, the volume being. "η" as calculated above. In a second step, the "excess" paste beyond this volume serves to push the aggregate grains apart until all of the excess paste has been introduced and the 1 m^3 total concrete volume attained. It is assumed that all of the aggregate grains (regardless of size) are enveloped by a shell of paste of uniform thickness, as indicated in the figure below. This shell thickness ("T_{shell}") is simply the volume of the excess paste divided by the total surface area of the aggregate. The mean inter-aggregate spacing is twice the thickness of the shell that results from introducing the excess paste between the aggregate grains.

The concepts are illustrated in the figure below. The upper figure shows the dry-rodded volume, incorporating part of the paste (equivalent in volume to "η"), with the remainder of the paste undistributed, i.e. in a layer above the dry-rodded volume. In the lower figure, the "excess" paste is distributed as uniform shells surrounding each aggregate grain.





Sample calculation

To illustrate the calculation procedure, a sample calculation is provided for the case of concrete containing the finest sand (FA#1):

Coarse aggregate content = 997 kg/m3
Fine aggregate content = 606 kg/m3
Total surface area of coarse aggregate in 1 m^3 of concrete = 0.23 m^2/kg × 997 kg = 229.31 m^2
Total surface area of fine aggregate in 1 m^3 of concrete = 10.36 m^2/kg × 606 kg = 6278.16 m^2
Combined surface area of aggregates (coarse + fine) in 1 m^3 of concrete = 6507.47 m^2
The calculated excess paste volume from Eq. (1) above was 0.20 m^3
The calculated shell thickness = 0.20 m^3/6507.47 m^2 = 30.73 µm
The mean inter-aggregate spacing = 2 × 30.73 µm = 61.46 µm.

References

[1] K.L. Scrivener, Characterization of the ITZ and its quantification by test methods, in: M.G. Alexander, G. Arliguie, G. Ballivy, A. Bentur, J. Marchand (Eds.), Engineering and Transport Properties of the ITZ in Cementitious Composites, RILEM Publications S.A.R.L., Paris, 1999, pp. 3–18, RILEM Report 20.
[2] S. Mindess, J.F. Young, D. Darwin, Concrete, 2nd ed, Prentice Hall, Upper Saddle River, NJ, 2003, pp. 305–306.
[3] J.J. Zheng, C.Q. Li, X.Z. Zhou, Characterization of microstructure of interfacial transition zone in concrete, ACI Materials Journal 102 (4) (2005) 265–271.
[4] J.J. Zheng, C.Q. Li, X.Z. Zhou, Thickness of interfacial transition zone and cement content profiles around aggregates, Magazine of Concrete Research 7 (7) (2005) 397–406.
[5] D.P. Bentz, J.T.G. Hwang, C. Hagwood, E.J. Garboczi, K.A. Snyder, N. Buenfeld, K.L. Scrivener, Interfacial zone percolation in concrete: effects of interfacial zone thickness and aggregate shape, in: S. Diamond, S. Mindess, F.P. Glasser, L.W. Roberts, J.P. Skalny, L.D. Wakeley (Eds.), Microstructure of Cement-Based

Systems/Bonding and Interfaces in Cementitious Systems, MRS Symp, Vol. 370, Materials Research Society, Pittsburgh, PA, 1995, pp. 437–442.

[6] D.N. Winslow, M.D. Cohen, D.P. Bentz, K.A. Snyder, E.J. Garboczi, Percolation and pore structure in mortars and concrete, Cement and Concrete Research 24 (1) (1993) 25–37.

[7] Z. Sun, E.J. Garboczi, S.P. Shah, Modeling the Elastic Properties of Concrete Composites, Cement and Concrete Composites, Vol. 29, No. 1, 2007, pp. 22–38.

[8] J. Zheng, J. Lu, Z. Wu, Prediction of the elastic modulus of concrete with inhomogeneous ITZ, Fuhe Cailiao Xuebao/Acta Materiae Compositae Sinica 25 (5) (2008) 141–146.

[9] Z. Hashin, P.J.M. Monteiro, An inverse method to determine the elastic properties of the interphase between the aggregate and the cement paste, Cement and Concrete Research 32 (8) (2002) 1291–1300.

[10] S. Diamond, J. Huang, The interfacial transmission zone: reality or myth, in: A. Katz, A. Bentur, M. Alexander, G. Arliguie (Eds.), Proc. 2nd International Conference on the ITZ in Cementitious Composites, Haifa, RILEM Proc 35, E and FN Spon, London, 1998, pp. 3–39.

[11] S. Diamond, S. Mindess, J. Lovell, On the spacing between aggregate grains in concrete and the dimension of the aureole de transition, Liaisons Pates de Ciment Materiaux Associes, Proceedings Colloque RILEM, Toulouse, 1982.

[12] A.G. Loudon, The computation of permeability from simple soil test, Geotechnique 3 (1952) 165–183.

[13] P. Rangaraju, Mixture proportioning and microstructural aspects of high-performance concretes, Ph. D. Thesis, Purdue University, West Lafayette, IN (1997) pp. 155–164.

[14] A.J. Nilson, P.J.M. Monteiro, Concrete: a three phase material, Cement and Concrete Research 23 (1) (1993) 147–151.

[15] K.M. Lee, J.H. Park, A numerical model for elastic modulus of concrete considering interfacial transition zone, Cement and Concrete Research 38 (3) (2008) 396–402.

[16] R. LeRoy, F. deLarrard, A discussion on the paper “Concrete: a three phase material” by A.U. Nilsen and P.J.M. Monteiro, Cement and Concrete Research 24 (1) (1994) 189–193.