THE INTERFACIAL TRANSITION ZONE: "DIRECT" EVIDENCE ON COMPRESSIVE RESPONSE

DAVID DARWIN
Department of Civil Engineering, University of Kansas, Lawrence, KS 66045

ABSTRACT

There is little question that the strength of the interfacial transition zone (ITZ) between cement paste and aggregate affects the compressive strength of concrete. The key question, rather, is to what degree? It is difficult to directly measure the response of the overall composite to changes in interfacial properties, since it is difficult to isolate interfacial strength as the only variable.

Research on the effects of interfacial strength on the compressive response of concrete that comes the closest to providing direct evidence is summarized. The studies, dating to the 1950's, include both experimental and analytical efforts aimed at isolating the effects of the ITZ, as well as experimental efforts that are considered to provide strong indirect evidence. The research shows that the ITZ plays a measurable role in the response of concrete to compressive stress, but that its role is overshadowed by the properties of the cement paste and aggregate constituents of concrete and the heterogeneous nature of the composite.

INTRODUCTION

It is widely recognized that significant differences in structure exist between bulk cement paste and cement paste located in proximity to aggregate particles[1-9]. Paste near an aggregate surface exhibits a smaller fraction of unhydrated cement particles (out to about 40 μm) and greater porosity (especially out to about 10 μm) than cement paste in regions located farther from the aggregate[9]. This region is referred to as the interfacial transition zone (ITZ) and has an estimated thickness of 15 to 50 μm, depending on the method of estimation[3, 9, 10].

Early studies using fractured surfaces of mortar and concrete indicated that the zone consists of duplex layers, dominated by calcium hydroxide crystals with c-axes normal to the aggregate surface[11]. More detailed studies using flat polished surfaces indicate that the material in the interfacial zone is principally calcium silicate hydrate[8], with an average calcium hydroxide (CH) content only slightly higher than surrounding paste. The predominance of CH in earlier observations is likely due to the use of fractured surfaces, which are not representative of the true full cross-section. CH will appear more often on fractured surfaces due to the ease of cleavage along its basal plane.[8]

The observed differences in structure, combined with tests of interfacial strength[12, 13], has led to the conclusion that the interfacial transition zone is the "weak link" in the strength of concrete. It is often[6, 7, 14-17] but not universally[18-20] assumed that the ITZ plays a dominant role in the compressive as well as tensile strength of concrete. This concern has led to a continuing interest in the interfacial transition zone.

This paper summarizes research aimed at directly establishing the role of the interfacial transition zone in controlling the compressive strength of concrete. Experience has shown that it is difficult to measure changes in strength due to specific changes in interfacial properties. However, such measurements have been attempted in a number of studies, with various levels of success.

The studies used to evaluate the contribution of interfacial strength to the compressive strength of concrete can be placed in three categories:

1. Experimental studies in which the strength of the interface between aggregate and paste is artificially modified (interfacial properties may or may not be measured) and the effect on compressive strength of concrete is determined.

2. Finite element studies in which the properties of cement paste or mortar, aggregate, and the

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ITZ are modeled. Of particular interest are those studies in which the properties of the ITZ are varied and the effects on the analytical response are determined.

3. Experimental studies in which the paste constituent is modified (usually by replacement of cement by silica fume) in a way that will result in changes in the ITZ. The resulting changes in the properties of the concrete are measured.

The following evaluation of these studies is aimed at determining the degree to which changes in the interfacial region affect the compressive strength of concrete.

EXPERIMENTAL STUDIES MODIFYING INTERFACIAL BOND STRENGTH

There are two aspects of interfacial strength: 1) the strength of the paste in the interfacial transition zone itself and 2) the bond strength between cement paste and aggregate. The experimental studies discussed in this section have involved attempts to modify the latter without changing the properties of the paste within the ITZ.

Studies of the effect of paste-aggregate bond strength on the compressive strength of concrete date at least to the 1950's[21, 22]. Studies by Dantinne[22], Shah and Chandra[23], and Nepper-Christen-sen and Nielson[24] used relatively thick, soft coatings on coarse aggregate particles to reduce bond strength. Dantinne[22] used a thick bituminous coating; Nepper-Christensen and Nielson [23] used a 13 µm coating of a soft plastic; and Shah and Chandra[23] used a 66 µm coating of silicon rubber. Darwin and Slate[25, 26] analyzed the effect of these coatings and found that they isolated the aggregate from the surrounding mortar, resulting in behavior more representative of a material with a large number of voids in the concrete matrix than a material containing an aggregate with reduced interfacial strength. Thus, the apparently large impact of changes in interfacial properties observed in those studies[22-24] must be disregarded.

Aware of problems in the earlier studies, Darwin and Slate[25, 26] and Perry and Gillott[27, 28] designed studies to determine the effect of paste-aggregate strength on compressive strength that would not isolate the coarse aggregate from the surrounding mortar. Darwin and Slate coated natural coarse aggregate particles with a 10 µm coating of polystyrene and demonstrated that the thickness and stiffness of the coating were such that the aggregate particles were not isolated from the surrounding mortar matrix. They ran separate tests to determine the tensile and shear bond strength of coated aggregate using sawed pieces of sandstone and limestone. Their tests demonstrated that they were able to reduce the tensile strength to about a third and the shear strength to about a fifth of that obtained at a typical mortar-aggregate boundary[12, 13]. They evaluated the stiffness and strength of concrete mixes and measured microcracking in concretes with and without coated coarse aggregate. Concrete strengths ranged from 21 to 31 MPa. In the load tests, Darwin and Slate observed no change in the initial stiffness, but about a 10 percent reduction in compressive strength for concrete containing coated aggregate compared to concrete with uncoated aggregate (Fig. 1). Using both microscopic and x-ray analyses, they observed no appreciable difference in interfacial microcracking in the two concretes at compressive strains of 0.0010, 0.0018, and 0.0022. They did, however, observe a small, yet significant increase in mortar microcracking in the concrete containing the coated coarse aggregate.

Perry and Gillott[27, 28] cast a series of concrete mixes containing glass marbles and artificially prepared quartzite coarse aggregate particles with different degrees of surface roughness. The glass marbles had either smooth or roughened surfaces. The roughened surfaces were obtained by placing the particles in ball mill containing either 80 or 1000 mesh silicon carbide grit. The quartzite particles were manufactured from solid rock and had a smooth barrel shape. The particles had polished surfaces or surfaces roughened in a manner similar to that used for the glass spheres. The maximum roughness for both aggregates corresponded to a "center line average" value of 4 to 5 µm. Concrete strengths ranged from 25 to 57 MPa. For each aggregate type, the rougher the coarse aggregate surface, the higher the compressive strength. The differences in strength, however, were low, matching the differences observed by Darwin and Slate[25, 26]. The smooth-surfaced glass spheres produced a compressive strength equal to 91.6 percent of that obtained with the 80 mesh roughened glass spheres, and the polished quartzite particles produced a concrete with

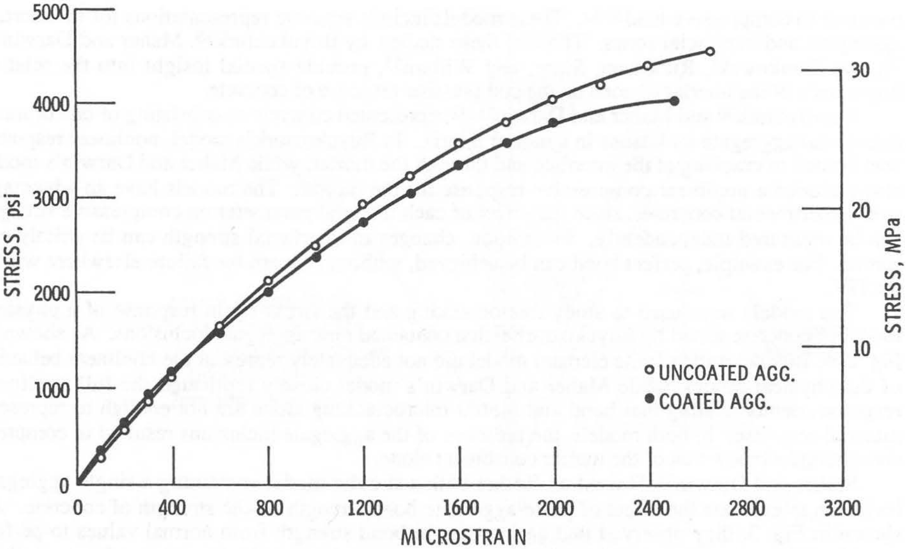

Fig. 1 Stress-strain curves for concrete comparing the effects of uncoated and coated coarse aggregate[25,26]

a strength equal to of 92 percent of that obtained with the roughened quartzite particles.

In another study, involving polymer-impregnated concrete, Carino[29] measured the effects of polymer impregnation on the bond between mortar and aggregate. Measuring interfacial tensile strength using modified tension briquets and interfacial shear strength using inclined aggregate slabs in mortar prisms, he observed that polymer impregnation does not increase interfacial bond strength, but does increase the tensile strength of mortar. Carino attributed the increase in the compressive strength in concrete obtained from polymer impregnation to the increased mortar strength, downgrading the importance of interfacial bond.

In another attempt to increase the compressive strength of concrete by increasing interfacial bond, Popovics[15] used a series of surface treatments on coarse aggregate. His procedures resulted in increases in the compressive strength of some mixes, but in a reduction in compressive strength in the majority of cases. Popovics pointed out that his lack of success was probably due to a failure at the interface at some distance from the actual contact region between paste and aggregate. The result was that the surface treatments did not adequately increase the actual interfacial strength, which was in fact governed by the surrounding cement paste, raising the point that to improve bond strength, the strength of the full interfacial transition zone must be increased, not just the strength at the contact surface between paste (or mortar) and aggregate.

Overall, the four studies described above demonstrate that 1) significant decreases and moderate increases in interfacial strength at the paste-coarse aggregate boundary, compared to values obtained in normal strength concrete, have a measurable but relatively low effect on compressive strength, and 2) increases in compressive strength can be obtained by increasing the strength of the mortar constituent (including the ITZ) alone.

FINITE ELEMENT STUDIES

Finite element models have been used to gain an improved understanding of the response of concrete to compressive load[30-35]. These models include separate representations for the mortar, aggregate, and interfacial zones. Three of these studies, by Buyukozturk[30], Maher and Darwin[31, 32], and Stankowski, Runesson, Sture, and Willam[35], provide special insight into the relative importance of the interfacial zone on the compressive response of concrete.

Buyukozturk[30] and Maher and Darwin[31, 32] represented concrete as consisting of one or more cylindrical aggregate inclusions in a mortar matrix. In Buyukozturk's model, nonlinear response was limited to cracking at the interface and through the mortar, while Maher and Darwin's model also included a nonlinear compressive response for the mortar. The models have an advantage over experimental concretes, since the effect of each material parameter on compressive strength can be measured independently. In addition, changes in interfacial strength can be strictly enforced. For example, perfect bond can be achieved, without concern for failure elsewhere within the ITZ.

The models were used to study microcracking and the stress-strain response of a physical model of concrete tested by Buyukozturk[30] that contained nine aggregate inclusions. As shown in Fig. 2 [36], Buyukozturk's finite element model did not adequately represent the nonlinear behavior of the physical model, while Maher and Darwin's model closely replicated the full nonlinear response, demonstrating that bond and mortar microcracking alone are not enough to represent material response. In both models, the presence of the aggregate inclusions resulted in compressive strengths below that of the mortar constituent alone.

Maher and Darwin[31, 32] went on further with a simpler model containing a single aggregate inclusion to evaluate the effect of paste-aggregate bond strength on the strength of concrete. As shown in Fig. 3, they observed that an increase in bond strength from normal values to perfect bond (no failure at the interface) resulted in only a 4 percent increase in the compressive strength of the model. A decrease to zero interfacial strength resulted in a decrease in compressive strength of just 11 percent.

In a more recent study, Stankowski et al.[35] represented concrete as a series of polygonal aggregate particles in a mortar matrix (Fig. 4). Using this realistic representation of concrete, they obtained only a 7 percent increase in compressive strength for a model with perfect interfacial bond

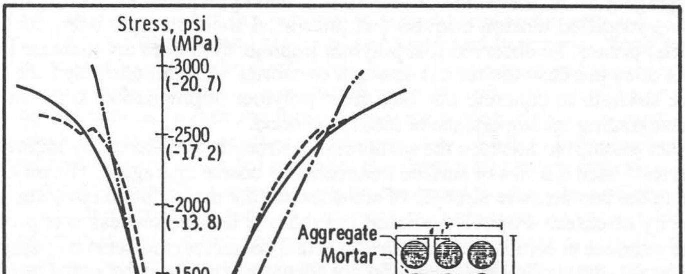

Fig. 2 Stress-strain curves for concrete model [36]

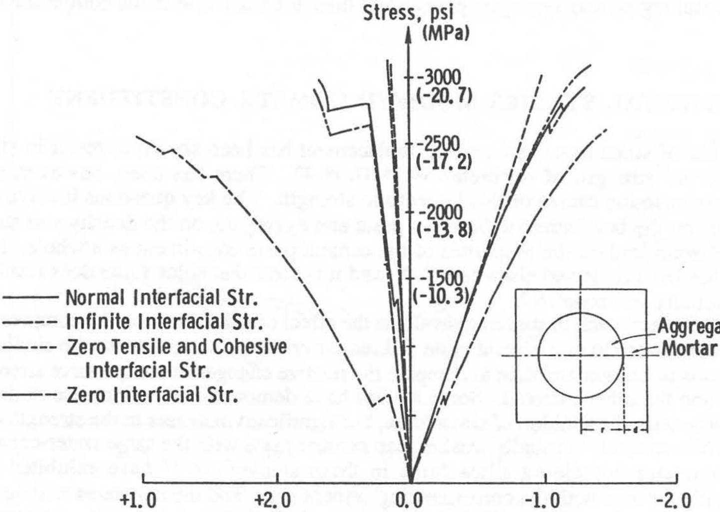

Fig. 3 Stress-strain curves for finite element models of concrete with different values of mortar-aggregate bond strength[31, 32]

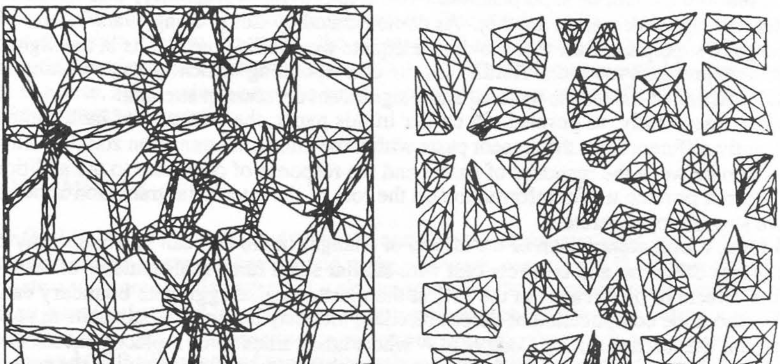

Fig. 4 Finite element model of mortar and aggregate[35]

strength compared to one with typical values of tensile and shear strength at the interface. Not surprisingly, the same change in interfacial strength produced a 29 percent increase in tensile strength.

The key observations from the analytical studies measuring the effects of direct changes in interfacial properties match the experimental observations indicating that the bond strength between

the interfacial region and aggregate plays a less than dominant role in the compressive strength of concrete.

EXPERIMENTAL STUDIES MODIFYING PASTE CONSTITUENT

The use of silica fume as a cement replacement has been shown to result in significant increases in the strength of concrete[7, 14, 16, 17, 19, 20]. There has been, however, a continuing controversy as to the causes of this increase in strength. The key questions involve the effect of silica fume on the bond strength between paste and aggregate, on the density and strength of the interfacial zone, and on the properties of the cement paste constituent as a whole. Much of this research has been reviewed elsewhere[19, 20], and it is clear that silica fume does result in a denser interfacial transition zone[4, 6, 7].

A popular approach in studies to evaluate the effect of silica fume on the compressive strength of concrete has been to cast cement paste and mortar or concrete specimens with similar silica fume replacements of cement and then to compare the relative changes in compressive strength obtained for paste and the other material. Some studies have demonstrated no increase in the strength of cement paste with the addition of silica fume, but significant increases in the strength of concrete[7, 16, 17]. While concrete is usually weaker than cement paste with the same water-cement ratio[7, 17, 37], the concretes containing silica fume in these studies[7, 16, 17] have exhibited compressive strengths higher than both the corresponding cement paste and the concretes without silica fume. These observations have led the investigators to conclude that the increase in compressive strength with the addition of silica fume has been due to an increase in the strength of the ITZ.

In contrast, other studies have demonstrated increases in the compressive strength of cement paste, mortar and concrete with the partial replacement of cement by silica fume[19, 20], leading to the conclusion that increases in the strength of the cement paste constituent are the primary reason for increases in the strength of concrete and mortar.

A key drawback in comparisons of cement pastes, mortars and concretes containing silica fume is that the amount of superplasticizer (used to provide workability) that works well in one material will not work well in another. As demonstrated by Cong, Gong, Darwin, and McCabe[20], the dosage of superplasticizer that provides adequate workability and results in the highest strength for concrete provides too much fluidity for the corresponding cement paste, resulting in segregation and bleeding in the plastic material and a significant decrease in strength.

More importantly, as pointed out earlier in this paper, the structure of bulk cement paste is significantly different from the cement paste within the interfacial transition zone[1-9]. Thus, a direct comparison between the response of pastes and the response of concretes to the addition of silica fume will not provide useful information on the role of the interfacial transition zone in the compressive strength of concrete.

A more useful comparison of the effects of changes in the ITZ can be obtained by comparing the strengths of mortar and concrete cast with similar silica fume replacements of cement. In this case, the effects of differences in the ITZ at the mortar-coarse aggregate boundary can be determined without the complication of excess bleeding that may occur with bulk cement paste. Such a comparison was carried out by Cong et al.[20] who studied silica fume replacement in cement paste, mortar, and concrete. One method of comparison that they used to establish the role of the ITZ was to compare the ratio of concrete strength to mortar strength (f'_c/f'_m) as a function of mortar strength. Thus, any changes brought about in the compressive strength of mortar due to silica fume replacement of cement would be considered separately from changes in the interfacial transition zone between the mortar and the coarse aggregate.

As illustrated in Fig. 5, Cong et al.[20] observed that the ratio f'_c/f'_m is virtually independent of the degree of silica fume replacement and the water-cementitious material ratio. The figure shows the results for concretes and mortars with water-cementitious material ratios of 0.33 and 0.39 tested at ages of 3, 7, and 28 days. The mixes involved contained 1) no admixtures, 2) superplasticizer alone, or 3) superplasticizer and a 15 percent cement replacement by silica fume. All of the mortars had identical volumes of fine aggregate and all of the concretes had identical vol-

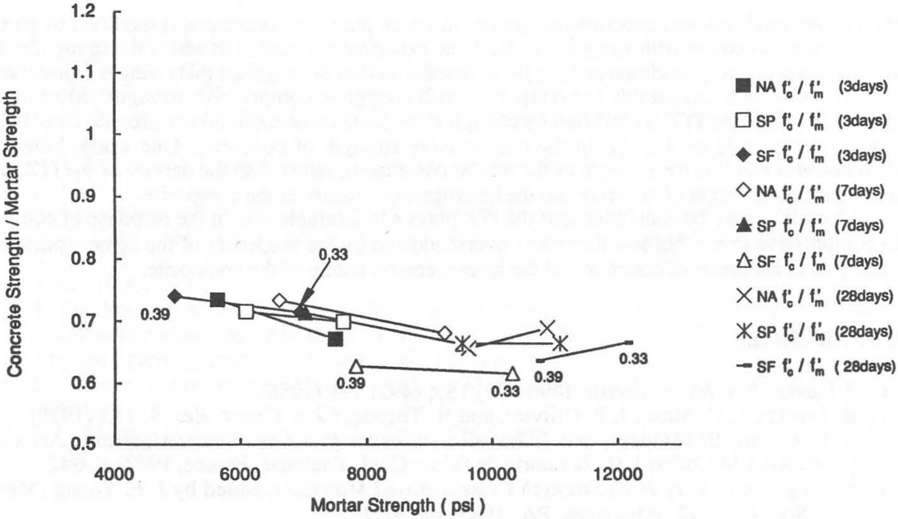

Fig. 5 Ratios of concrete strength to mortar strength as a function of mortar strength (NA = no admixtures; SP = superplasticizer; SF = superplasticizer and silica fume)[20]

umes of fine and coarse aggregate. The presence of coarse aggregate results in a drop in strength of 26 to 39 percent. The results plot in a narrow range and, if the results for the materials containing silica fume (SF) tested at 7 days are removed, the results could be characterized as falling in a *very* narrow range. The principal factor controlling the ratio f'_c/f'_m is mortar strength, independent of age, water-cementitious material ratio or admixture. The results in Fig. 5 lead to the conclusion that, for the materials used in the study, the increases in the density of the ITZ around coarse aggregate particles obtained through the use of silica fume had no measurable impact on the strength of concrete, again pointing out the small impact of changes in interfacial properties on the strength of the composite material.

FINAL COMMENTS

A few final points can be obtained from the early microcracking studies in which the tensile and shear bond strengths between cement and aggregate were measured[12, 13]. The tensile strength between paste and aggregate decreased at a slower rate than the compressive strength of cement paste with an increase in water-cement ratio. The tensile and shear strengths between mortar and coarse aggregate were largely insensitive to water-cement ratio for water-cement ratios ranging between 0.36 and 0.75. This lack of sensitivity in bond strength to changes in water-cement ratio provides strong support for the matrix, rather than the interface, as the principal controlling factor in the strength of concrete.

SUMMARY

The research summarized in this paper emphasizes studies designed to directly measure the effects of the properties of the interfacial transition zone on the compressive response of concrete. The research involves experimental studies in which the interfacial strength between aggregate and paste is artificially modified, finite element studies in which the properties of the constituents and the ITZ are modeled, and experimental studies in which the paste constituent is modified by partial replacement of cement with silica fume. Both the experimental studies in which the strength of the interface is artificially modified and the finite element studies demonstrate that changes in interfacial strength provide a measurable but relatively small change in compressive strength. Most of the studies in which the ITZ is modified by changing the paste constituent do not provide clear information on the role of the ITZ in the compressive strength of concrete. One study, however, demonstrates that it is the strength of the mortar constituent, rather than the density of the ITZ, that dominates the strength of concrete and the heterogeneous nature of the composite.

Overall, it may be concluded that the ITZ plays a measurable role in the response of concrete to compressive stress, but that the role is overshadowed by the properties of the cement paste and aggregate constituents of concrete and the heterogeneous nature of the composite.

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