2.5 Aggregate/Paste Bond

Concrete may be thought of as a system that derives its strength from three components, the strength of the aggregate, the strength of the paste that fills the spaces between the aggregate, and the strength of the bond between paste and aggregate. The bond between the paste and the aggregate is generally the weakest link, and therefore failure is initiated in the ‘aggregate/paste interfacial zone’, a zone extending approximately 20 to 50 microns from the aggregate into the paste, also referred to as the *‘transition zone’*. The relative inferiority of the paste in this zone stems from a number of defects peculiar to the zone, and given its importance in determining the strength of concrete and abrasion resistance, these are considered in some detail further on.

A second consideration that makes this zone so important is the high proportion of space taken up by it in relation to the total volume of paste. Stamatiou(1993)states that the major differences in this transition zone occur within the first 20 microns from the physical interface, and that the average spacing between adjacent particles is about 80 microns. This suggests that a relatively large proportion of the hydrated cement paste (hcp) lies within the interfacial zone.

2.5.1 Failure Mechanisms

In this section an attempt is made to show that the transition zone contributes more to compression failure than to abrasion wear. The mechanism of compression failure is described, followed by a proposed mechanism for abrasion failure. The relative contribution of the transition zone in the two loading systems is discussed. These considerations are tied in with the findings of some researchers.

(a) Failure mechanism in compression

Newman(1997a) recognises the important role of the transition zone in compressive strength. Axial cracks (cracks that are orientated and grow in the direction of the applied stress) initiate in the transition zone along the vertical interfaces between aggregate and paste. Cracks are initiated in this zone, and propagate as compressive stresses increase. These cracks continue to grow in the direction of the applied stress, eventually extending over the full face of the vertically aligned aggregate/paste interfaces. With further increases in stress the axial cracks breach the surrounding paste, above and below, to join up with other vertically aligned cracks from other aggregate above and below, until failure is complete. He supports his theory of axially orientated crack growth by stating that an examination of a coarse aggregate particle in a crushed cylinder generally shows that it is free of paste along its vertical sides, but has concrete adhering to its upper and lower faces.

Gjorv(1990)’s experience is that for compressive strengths up to a certain level of about 90MPa, ‘the *fracture* of concrete is controlled largely by failure of the bond between the aggregate particles and the cement paste’. (Above this level ‘concrete fracture is controlled largely by the strength of the rock aggregate’).

(b) Proposed failure mechanism in abrasion wear

Stamatiou(1993) investigated the interfaces between the hydrated cement paste and aggregate particles, explaining how they constituted a relatively weak *‘soft’ and ‘compressible’ zone*. (The contributing factors will be considered later on). From this it may be postulated that abrasive forces acting on exposed aggregate particles will have the effect of momentarily displacing them in the relatively ‘soft/compressible’ transition zone. This displacement may take the form of a vertical movement, a lateral movement, or a rotation. As the load is removed, the aggregate particle returns to its original position. For example a rocking effect may take place as a small hard wheel progressively moves over the aggregate particle. A grain of sand rolling or sliding over an aggregate particle will have the same effect, and the extent that the aggregate is displaced will depend on the magnitude of the load on the sand grain. This is illustrated in Figure 9.A of volume 1. In effect the sand grain focuses a relatively large force on a relatively small particle resulting in comparatively relatively large displacements within the transition zone. Eventually these movements can lead to fatiguing and the formation of *micro-cracks* in the transition zone. Ultimately this results in the *loosening* of the particle relative to the paste matrix, and finally to its removal.

(c) Relative contribution of the transition zone

Ramezanianpour(1999) did compressive strength tests to evaluate three different aggregate types, ‘conventional’ referred to as the control hereafter, ‘granite’, and ‘siliceous’ aggregates. The results of *compressive strength tests* showed remarkably good strengths for the ‘granite’ aggregate 93,4 MPa, but poor results for the ‘control’ (40,8MPa) and ‘siliceous’ aggregates (39,7MPa). Given the low w/c ratio of 0,3, and the cement content of 400kg/m^3 used in all the mixes, 93,4MPa is a good but believable result for 28-day compression testing. This is an increase of 129% relative to the control. Since the paste quality was the same for all mixes, and assuming that aggregate does not break down at 40MPa, the 40MPa results for the non-granite mixes points paste/aggregate bond as the weak link. The magnitude of the difference between the granite aggregate relative to the other two underlines the importance of aggregate/paste bond.

Ramezanianpour also used an *impacting fine-abrasive* (water blasted) abrasion test (see appendix U.5.22) to evaluate the effect of the three aggregate types on abrasion resistance. Relative to the ‘control’, the ‘granite’ and ‘silicious’ aggregates were respectively improved by 35% and 25%. The higher result for the granite aggregate is expected from the higher aggregate/paste bond, indicated by the compression testing. Clearly a greater bond strength means that impacting sand particles and the stream of water will not dislodge an aggregate particle from the matrix as easily. However, relative to compression testing, the 35% improvement for the granite mixes is relatively small. Surprisingly, the siliceous mix improved almost as much.

Following is an explanation why the granite was 129% better in crushing, but only 35% better in abrasion: In crushing of concrete, aggregate/paste failure may be regarded as the primary mode of failure. (Axial cracks spawn in the interfacial zone and grow in number and length until no further load can be sustained.) However in Ramezanianpour’s abrasion test a substantial quantity of paste must first be eroded before the aggregate particle is sufficiently exposed to the point where the transition zone stresses lead to a breakdown of the aggregate/paste bond. Clearly the erosion process will be similar for all three tests; of similar duration, requiring roughly the same quantity of abrasive, etc, and only once this exposure process has run its course will the improved aggregate/paste bond of granite become apparent. Thus in abrasion, transition zone stresses are only a part of the dislodgement process, whereas they play a major role in compression failure.

(d) Consideration of abrasion forces

The relative insensitively of abrasion resistance to transition zone bond strength may also be explained using another line of logic. In certain abrasion tests it is virtually impossible for the aggregate particles to escape out of their sockets, even though they may have become unbonded. An example would be the commonly used Amsler-Lafon apparatus, where the specimen is pressed down onto a horizontal grinding table. This means that the aggregate particles will continue to contribute to abrasion resistance beyond the point that they would have been dislodged in practice. In effect this test measures the hardness of the aggregate, and is virtually incapable of measuring the bond strength within the transition zone.

On the other hand certain rolling dressing wheels tests, e.g. ASTM C779 Proc B, impart considerable impact on individual aggregate particles. The resulting shock and vibration will tend to break aggregate/paste bond at a relatively early stage, well before the aggregate particle abrades away. The particle is dislodged rather than abraded, and the test is essentially a measure of aggregate/paste bond.

It was explained earlier that the impacting fine-abrasive (water blasted) abrasion test used by Ramezanianpour, like the rolling dressing wheels test, does not abrade primarily by wearing down the aggregate. It tends to erode the paste around an aggregate particle, leaving the aggregate particle relatively exposed, until the bond stresses in the transition zone become greater than the bond strength, at which point the particle is in effect thrust out.

From the foregoing it may be said that abrasive forces, depending on type and magnitude, induce a combination of compression, shear, flexural and tensile stresses upon the aggregate particles, and these stresses are transferred to the surrounding matrix by means of aggregate/paste stresses that are most detrimental in the relatively weak transition zone. It follows that the aggregate/paste bond strength may be defined as the ability of the transition zone to resist abrasive forces. Clearly this is a function of the quality of the paste and certain characteristics of the aggregate. The following two sections are devoted to these aspects.

2.5.2 Quality of Paste

It has been shown by several investigators (including the writer, see volume 1) that the greater the strength of the paste, the greater will be its contribution to abrasion resistance. Firstly, strong pastes are harder and more abrasion resistant; and secondly, they develop a stronger bond to the harder aggregate, enabling the aggregate to resist the abrasive forces for longer.

Given the superior hardness of aggregate, the binding ability of the paste is its primary function relative to abrasion resistance. To be precise, it is the binding ability of the weakest zone of the paste, the transition zone, that determines its bonding ability.

Connell(1985) did abrasion tests on concrete according to BS 812:Part 3:1975 clause 9 [=*sliding fine-abrasive*, see appendix U.5.06]. He found that a low paste strength resulted in ‘plucking’ out of the aggregate particles.

Komonen(1998) did abrasion tests on concrete pavers using *rolling studded tyres* [see appendix U.3.04] and found that the rate of abrasion of concrete pavers was effectively twice as good as that of asphalt ‘concrete’ made with the same aggregate type and equivalent grading. [It is therefore evident that the inherent abrasion resistance and the bonding ability of a high strength paste is superior to that of a bitumen binder].

2.5.2.1 Porosity

In section 2.3 it was seen that voids of any description are detrimental to the strength of concrete. These include entrapped voids and air entrained voids on one scale, and cavities, capillaries and pores in the paste on a smaller scale, and finally the sub-microscopic gel-pores. Paste that has a high percentage of interconnected capillaries and pores within the paste structure may be referred to as porous, and will have lower strength.

In the discussions that follow it will be useful to consider the strength of the paste in terms of its porosity, and especially the porosity of the transition zone.

The following investigators sketch the structure of the transition zone and outline some factors that lead to a higher porosity in this zone relative to the ‘core’ paste. This increased porosity increases the degree of deposition of orientated $Ca(OH)_2$ crystals in the transition zone.

a) Transition Zone Structure

Subramanian(1999) states that the present view on the aggregate/cement paste transition zone is that there is a 25 to 50 micron thick relatively porous zone surrounding the aggregate having properties that are different from the bulk cement paste.

This transition zone has a number of characteristics:

- In concrete containing excess water, the bleed water tends to collect below the aggregates, thereby creating a weak porous region with an *unfavourable w/c* ratio.
- Near the large surface of an aggregate particle, the cement particles are *not packed in an ideal manner* (wall effect), thereby leading to more porosity than in the bulk paste).
- As the aggregate surface is inert, the *CSH gel* (formed on the cement grain) grows *only in one direction* and not towards the aggregate faces (one sided growth), again leading to a more porous structure closest to the aggregate particle.
- The above factors result in a zone that is considerably more porous than the bulk paste. This encourages the deposition of *orientated crystals of calcium hydroxide,* giving rise to weak planes.

Stamatiou(1993) investigated the interfaces between the hydrated cement paste (hcp) and aggregate particles, explaining how they constituted a relatively weak *‘soft’ and ‘compressible’ zone*.

His analysis of the formation and structure of the transition zone is very similar to Subramanian’s. He explains that during mixing, casting, and consolidation of concrete, a layer of water accumulates around aggregate particles. Prior to initial set, this can be aggravated by additional bleed water gathering under large aggregate particles. Furthermore the so-called wall effect prevents cement particles from packing efficiently around the aggregates. Thus the aggregates are surrounded by a ‘transition’ zone of relatively *high w/c*, in which conditions are markedly different from those in bulk paste. (Note that high w/c leads to high porosity). These various effects result in a transition zone around the aggregate particles that is about 50 microns thick. It is rich in calcium hydroxide (CH) crystals which lie roughly parallel to the aggregate surface. This results in the formation of preferential *fracture planes* due to non-random orientated CH crystals.

Both these investigators have highlighted the negative effect that ‘wall effect’, bleeding and high w/c has on increasing the voids in the transition zone, and also the formation of large and oriented CH crystals. The result is a weak transition zone, exacerbated by preferential fracture planes. Clearly, this will impact negatively on the abrasion resistance.

Consideration will now be given to investigators who found ways of improving the porosity of the paste.

b) Improving Paste Porosity

Subramanian(1999) suggests a number of ways of improving the aggregate/hcp bond:

- Powerful superplasticizers are very effective in reducing w/b and bleeding.

- Aggregates with slightly porous surfaces will reduce the w/b in the transition zone. The excess water in the transition zone is drawn into these pores and cement particles are also drawn closer to the aggregate's surface.
- Limiting the maximum size of the coarse aggregate to 12,5 mm reduces the incidence of bleed-water entrapment.
- Given that the average size of cement grains is about 40 microns, adding silica fume or other fine material will partially fill the interstitial spaces and densify the interfacial zone
- Introducing a physical or chemical reaction between the aggregate particle and paste will also improve the aggregate/paste bond, especially in the immediate vicinity of the transition zone. For example aggregate particles can be pre-coated with a cement/silica-fume slurry. Alternatively, Subramanian suggests that the ideal aggregate would consist of an inert, hard core, coated with a hydraulic layer that is slightly porous, and capable of combining with cement. Tests done on corundum aggregate coated with a mixture of $Ca(CO)_3$ powder, with a small amount of gypsum and water and fired at 1400 degrees for four hours resulted in superior properties relative to untreated granite, corundum of Portland clinker.

ACI Committee 234(1995) states that the microstructure of the cement paste transition zone in concrete, which is about 50 microns thick, is significantly different from that of the bulk paste. Studies of the hydration process in this zone for pastes with fly ash, slag, or silica fume have concluded that all of these materials affect the morphology of the transition zone and particularly *decrease its thickness* and degree of *orientation of calcium hydroxide crystals* that form adjacent to aggregate particles.

Khayat(1998) In a survey of the literature on silica fume, Khayat refers to the *pore refinement* associated with the pozzolanic reactions, whereby silica fume and $Ca(OH)_2$ combine to form additional *C-S-H products*. This also results in a reduction of the total volume of capillary pores in the cement paste. In particular the volume of large capillary pores are reduced resulting in a finer and less interconnected pore structure, and this refined and improved structure is also extended to the transition zone. There is therefore a reduction in the porosity of the transition zone between cement paste and aggregate which increases the strength and impermeability of the concrete. The conversion of $Ca(OH)_2$ to CSH products generally means that there are fewer and smaller and less orientated $Ca(OH)_2$ crystals in the transition zone. Finally the thickness of the transition zone is substantially reduced by adding silica fume to the concrete, since silica fume cuts down on *bleeding* and the amount of water accumulated under aggregate.

Relevance to abrasion resistance: The greater adhesion of cement-paste to aggregate in mixes incorporating silica fume reduces the incidence of aggregate being 'plucked out of the paste by abrasive action'.

c) Sectional summary

The findings of the foregoing investigators on improving the quality of the paste (and hence transition zone bond) are now summarised in point form:

- Use superplasticizers to lower w/c
- Use silica fume to (a) increase cohesion and so reduce bleeding, (b) densify by filler effect, (c) provide sites for conversion of CH to additional CSHs and thereby also refine pore structure, (d) pre-coat aggregates
- Use aggregates with slightly porous surfaces to lower w/c
- Use 12,5 mm aggregate to limit bleed water entrapment
- Use cement extenders such as silica fume, fly ash, ggbs to reduce the thickness of the transition layer, and reduce the orientation of the CH crystals

Virtually all the effects mentioned above operate on the basis of a reduction in the voids (porosity) in the transition zone.

2.5.3 Aggregate

Many investigators have considered the various properties of aggregate on abrasion resistance – see 2.4.1.and 2.4.2. This section will therefore be confined to a consideration of certain aggregate properties on the transition zone.

Clearly the size, surface texture and porosity of the aggregate, both coarse and fine, will influence the *bond* between aggregate particles and surrounding paste. An aggregate with a slightly *porous surface* will reduce the w/b of the transition zone as explained earlier. A *rough texture* increases the surface area for aggregate/paste contact, resulting in a reduction in the transition zone stress for a given abrasive loading. Finally, the *size* of the particle will influence the force required to rupture the aggregate/paste bond in the first instance, and thereafter dislodge it.

Note that although increased surface *texture* increases the aggregate/paste contact area, thereby potentially reduces the interfacial stresses, it may also increase w/c. Similarly elongated and flaky particles may have increased surface areas for bonding, but again w/c will be higher than for round or cubical shapes. Thus the benefits of increased paste/aggregate contact area must be considered in the light of increasing w/b.

In 2.5.1(c) Earlier is was shown that Ramezanianpour(1999) obtained differences in abrasion resistance depending on aggregate type. While the degree of these differences was not nearly as much as that obtained in compression testing, differences up to 35% for the aggregates he used do indicate the influence of aggregate type on aggregate/paste bond.

Addis(1989) did abrasion tests with sliding fine abrasive (NBRI apparatus) and found that the abrasion resistance of mortars was both a function of aggregate type and compressive strength. For example, considering first 60MPa mortars:

Mortar made from andesite had 50% greater wear than corundum, while at the other end of the scale, at 12 MPa, the converse applied; the corundum mortar had 90% greater wear than that of andesite. It is likely that at the low MPa values, the fine aggregate would have been pulled out by the abrasive, and it therefore appears that andesite has superior aggregate/paste bonding relative to corundum.

2.5.4 Conclusion

The transition zone, at the interface of the aggregate and paste, may be regarded as the weakest link in the concrete system. It is unfortunate that the weakest zone of the paste is also the vital interface with the aggregate, as clearly this compromises aggregate/paste bond.

The morphology of the transition zone has been considered and the mechanisms detrimental to its strength have been discussed.

Evidence is presented to show that the transition zone is not as detrimental to abrasion resistance as it is to compressive strength. Aggregate type influences aggregate/paste bond.

Finally it is shown that the judicious use of certain admixtures and cement extenders improves the morphology of the transition zone, leading to improved compressive strength and abrasion resistance.