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The ITZ in concrete – a different view based on image analysis and SEM observations

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Abstract

The traditional picture of the ITZ in concrete involves an approximately 30 μm zone around each aggregate, within which the porosity increases as the aggregate interface is approached. The results of the writers' extensive image analysis investigations, and examinations of SEM specimens from various concretes provide a very different picture. While the "wall effect" excluded much of the ground cement from the vicinity of the aggregates, the great increases in pore content within a few μm of the aggregate, up to approximately 30% porosity, that have been reported by others, are not found. On average, only modestly higher porosities are observed within the ITZ than in the bulk paste. This is true even in the innermost areas immediately adjacent to the aggregates. In part, the extra space produced by the wall effect is filled in by CH deposits, many of which are anchored directly on the surfaces of aggregates and are essentially non-porous. Strong indications exist that the ITZ contains as high a proportion of C–S–H per unit volume as the bulk paste; thus some of the extra space that was created by the wall effect is filled in by through-solution deposits of C–S–H derived from elsewhere in the system. It is considered that the structure of the ITZ in ordinary concretes is not different enough from that of the bulk cement paste to provide any basis for significant effects on permeance or mechanical properties.

Keywords: Interfacial transition zone (ITZ); Auréole de transition; Image analysis; Microstructure; Backscatter images; Scanning electron microscopy (SEM)

1. Introduction

The concept that an interfacial transition zone (ITZ) or "aureole de transition" exists around sand and coarse aggregate particles in concrete has been one of the accepted tenets of concrete technology for many years. This generally accepted picture was based originally on optical microscopy and on experimental observations made on "model systems" rather than on concrete per se. In essence, the argument is that a region, extending about 30 μm or more from the aggregate, is deficient in content of cement particles due to the so-called wall effect, and therefore has a substantially higher porosity than the bulk paste [1].

A number of models of the ITZ exist. While some models assume that the ITZ can be viewed as a uniform shell, it is generally assumed that there is a gradation of porosity and other characteristics within the aureole. A very high porosity, ca. 30% or more, is assumed to be characteristic of the innermost portion of the aureole, i.e., within 5 μm of the aggregate surface [2]. At the outer boundary, porosity and other characteristics are considered to merge into those of "bulk" cement paste. The contents of CH and of ettringite are usually considered to be higher within the aureole than in the bulk cement paste, and much of the CH is considered to be preferentially oriented [3].

For concretes in which adjacent aureoles overlap significantly, it has been claimed that a continuous easy percolation path can be established across the section, leading to high permeability and rapid diffusion of ions and other dissolved species [4].

The aureoles are perceived as weakening zones which limit the strength that would otherwise be developed in concrete. High performance concretes are thought to derive augmented strength levels partly from a filler effect, in which fine particles such as silica fume fill up the extra ITZ porosity that would otherwise exist, and by doing so eliminate the weakening effect [5].

We have carried out an extensive series of investigations to attempt to establish the degree to which this

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generally accepted picture of the ITZ is valid for ordinary concrete. Some of the results have been previously published in the Proceedings of a recent RILEM conference [6]. These investigations employ certain specialized image analysis procedures described in some detail in a companion paper [7].

Briefly, our procedures involved preparation of the epoxy-filled, polished flat-surface specimens as ordinarily done for backscatter SEM investigations. In such procedures thin slices are cut from bulk concrete by low-speed precise diamond-bladed saws using non-aqueous lubricants. The slices are dried at low temperature, impregnated with ultra-low viscosity epoxy resin, and the resin is hardened. A new epoxy-impregnated surface is then cut, ground, and polished, all without exposure to water.

Sand grains exposed on the resulting specimen surface are selected for study by a formal random selection process. The area surrounding each selected grain is imaged by backscatter SEM. The normal magnification used for evaluation is 500×, and the pixel resolution is 512 × 512 ; accordingly each pixel is approximately 0.5 μm in each direction, covering an area of 0.25 μm^2.

Sampling units, each ca. 250 μm long and 10 μm wide are cut from the overall image, and subjected to gray-level binary segmentation to determine content (and distribution over the sampling unit) of each of three segmentable phases: unhydrated cement grains, calcium hydroxide, and detectable pores. C–S–H, not independently determinable by segmentation because of variations in gray level, is estimated by difference. In our procedure, each sand grain selected is entirely surrounded by sampling units, which are placed in successive layers up to half-way to the next neighboring sand or coarse aggregate grain.

In the present paper, we illustrate results indicating that the wall effect *does* in fact function to limit the population of portland cement particles near the aggregates, but that despite this, the aureole generally shows only modestly higher porosity than the bulk cement paste. Importantly, there is no indication of especially high local porosity adjacent to the aggregate. If these findings are generally applicable to ordinary field concrete, most of the basis for considering that the ITZ has significant effects on mechanical properties and permeance disappears.

2. The ITZ – general appearance

Fig. 1 shows a more or less typical backscatter SEM view taken at 500× adjacent to a sand grain in a well-mixed three-day-old laboratory concrete of w:c 0.50. The sand and coarse aggregate used in this concrete are both dolomite. A boundary approximately 30 μm from the sand grain surface has been superimposed on the picture. The bright areas within the cement paste are residual unhydrated portland cement particles. It is seen that their content within the 30 μm zone delineated by the boundary is minimal. Gray level segmentation of unhydrated cement particles from the rest of the cement paste is facilitated by their brightness in backscatter-mode SEM; the determination of area percent of such particles in any designated area is thus perfectly straightforward.

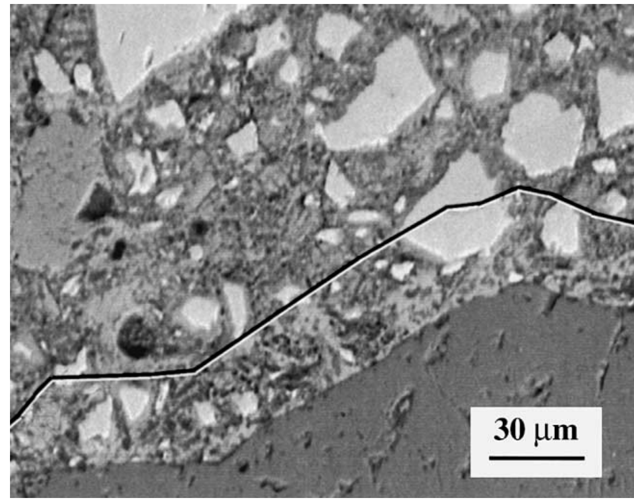

Fig. 1. Backscatter SEM image of an area around a sand grain in a well-mixed three-day-old w:c 0.50 concrete. An approximate boundary at 30 μm from the sand grain is superimposed on the figure.

3. Fundamental basis for the existence of the ITZ – the wall effect

Fig. 2 is a plot of the average area percent of residual cement grains tallied in successive 10 μm wide strips outward from dolomite sand grain boundaries in a three-day-old dolomite-aggregate concrete. Analyses in narrow strips at progressive distance from the aggregate perimeter is the usual method of investigating the structure of the ITZ. In our analyses, each strip entirely surrounds the sand grain under study, but as indicated in the companion paper [7], for technical reasons each strip is actually made up of separate adjacent individual sampling units each about 250 μm long. The plot of Fig. 2 represents averages of many sampling units for each distance.

The pattern observed in Fig. 2 is quite representative of the pattern found in the various concretes we have examined. We have systematically examined four different series of concretes, each at two ages (3 and 100 days); the overall study encompassed well over 6000 individual binary segmentations and evaluations. In all of our results there is always a strong gradient of the area percent of unhydrated cement, starting from the

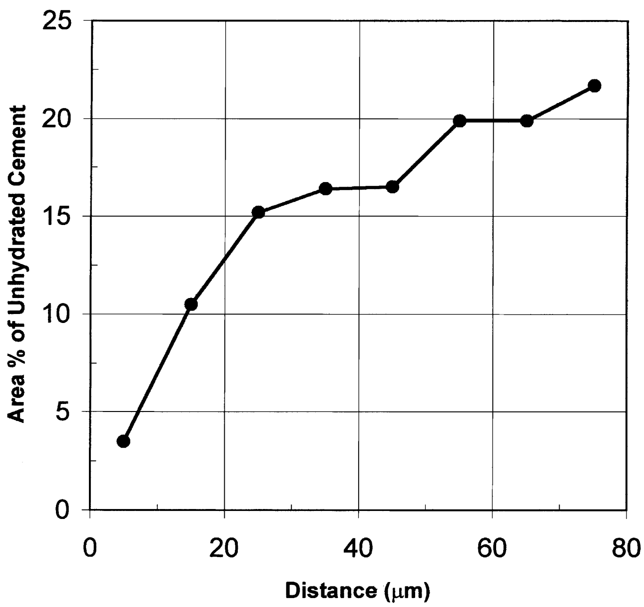

Fig. 2. Average area percent of unhydrated cement vs distance from the aggregate for a three-day-old well-mixed concrete made with dolomite sand and coarse aggregate.

bulk cement paste, with progressively less residual cement as the aggregate is approached. This general pattern confirms that the wall effect deprives the areas near the aggregate grains of their due proportion of portland cement particles, even in thoroughly mixed concrete. This produces a local shell around the aggregate grains that is somewhat different from the bulk cement paste at the start of hydration.

The extent of the affected zone seen in Fig. 2, as in most concretes, is not limited to 30 μm; we typically have found that measurable gradients extend out to 50 μm or more.

The gradient displayed in Fig. 2 is statistical in character. Large variations in unhydrated cement content occur within any given strip as one proceeds laterally around a given aggregate grain. Similar variations have been documented in [7] for the pore content within a given strip. Local variation is a normal and inescapable characteristic of cement paste microstructure in concrete. Thus, despite the fact that data such as that of Fig. 2 indicate that portland cement grains are statistically excluded from the vicinity of aggregates, the writers have seen (and recorded) many instances of individual residual cement particles touching or almost touching aggregates, or occurring in very restricted spaces between two closely-adjacent sand grains.

4. Detectable porosity within the ITZ

The primary basis for the concern of most concrete technologists with the ITZ is the idea that the aureoles continue to be regions of high porosity even after concrete has hydrated. This seemed to be the case in examining various laboratory model systems set up to simulate concrete. It appeared to be substantiated by quantitative image analysis results by Scrivener, Crumbie, and Pratt [8], Scrivener and Gartner [9], and Scrivener, Bentur and Pratt [10], and has recently been reiterated by Scrivener [11]. These workers published several profiles of average % of detectable pore area vs distance from the aggregate surface. Typically the pore area % in the cement paste outside of the aureole, i.e., the bulk cement paste, was of the order of 5% to 10%. Progressively increasing porosity was tallied through the aureole, with reported values reaching 30% or more of pores in the innermost 5 μm. Clearly, aureoles of such high porosity might be expected to lead to ITZ effects of major significance.

The results of our own image analysis investigations on well-mixed concretes indicate a general level of detectable porosity in the bulk cement paste of the order of 6% to 9 %, i.e., in the same range as those reported in the publications cited above. However, we do *not* find the strong gradient of increasing pore space within the aureole reported by these authors. Profiles of pore area % vs distance found for several well-mixed three-day and 100-day-old concretes are provided in Fig. 3. Our observations indicate that the porosity increase within the aureole is modest. Even the innermost strip (0–10 μm from the aggregate surface) shows porosities no greater than 13% for the young (three-day-old) concretes and less than about 10% for the mature (100-day-old)

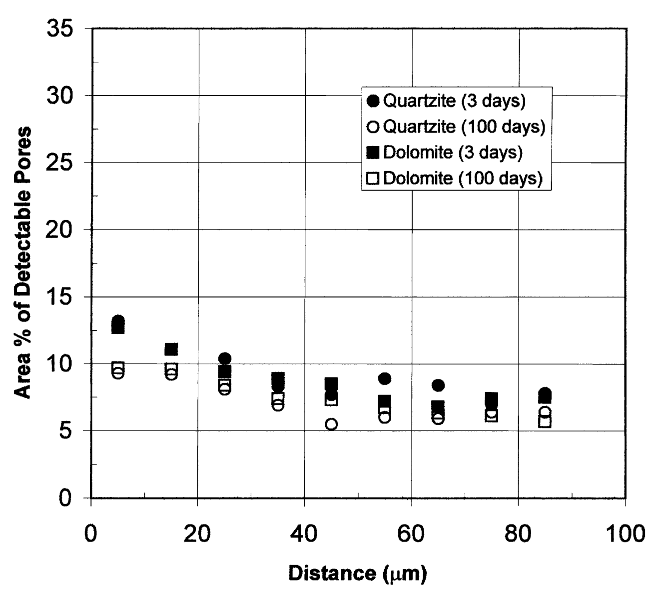

Fig. 3. Average area% of detectable pores vs distance from the aggregate for three-day-old and 100-day-old dolomite aggregate and quartzite aggregate concretes.

concretes, as compared to 30% or more reported in the literature cited above.

Our image analysis results have been challenged in oral discussion, from the perspective that the 10 μm wide sampling units employed by us are not sufficiently narrow to delineate the higher pore content claimed to be present in the very innermost (0–5 μm) layer of the aureole.

The first author has carried out qualitative assessments of several hundreds of individual binary segmented images showing the location of the pores detected in the 0–10 μm sampling units for various concretes. These extensive examinations failed to indicate any particular increase in porosity in the 0–5 μm portions as compared to the 5–10 μm portions of these images.

In Figs. 4 and 5 we provide binary segmented images showing the location of pores within a representative set of 0–10 μm sampling units for each of our concretes whose porosity vs distance profiles are shown in Fig. 3. Images for the three-day-old and 100-day-old dolomite aggregate concretes are shown in Fig. 4; those for the corresponding quartzite aggregate concrete in Fig. 5. Five images are shown for each concrete of each age. The specific images shown were randomly selected, using a table of random numbers, from the ca. 40 to 60 images available for each concrete of each age.

In these figures each strip imaged is 10 μm wide, and closely follows the outline of the perimeter of the aggregate. The black areas represent pore space pixels; pixels for all solid phases are white. The individual pixels making up the images are 0.5 μm in size. Each of the strips has been rotated so that in each case the aggregate is located immediately below the strip as positioned in the figure.

The irregular character of the distribution of pores is apparent in these images. However, one can readily compare the content and distribution of individual pores in the 'lower' part of the strip (0–5 μm from the aggregate contact) with those of the 'upper' part of the strip (5–10 μm from the aggregate contact). Except for one image (Fig. 5(b)) there is no indication of special concentration of porosity within the lower, so-called 'close interfacial' zone immediately adjacent to the aggregate. Thus we find no basis for the assertion that use of 10 μm wide strips in image analysis in some manner 'hides' the porosity of the inner portion of the aureole.

The unusual character of Fig. 5(b) is an exception to this statement. Its origin is readily explained by reference to Fig. 6, which is a gray scale micrograph of the area from which Fig. 5(b) was taken, oriented in the same direction. It is apparent from Fig. 6 that the cement paste adjacent to the aggregate is dense and not

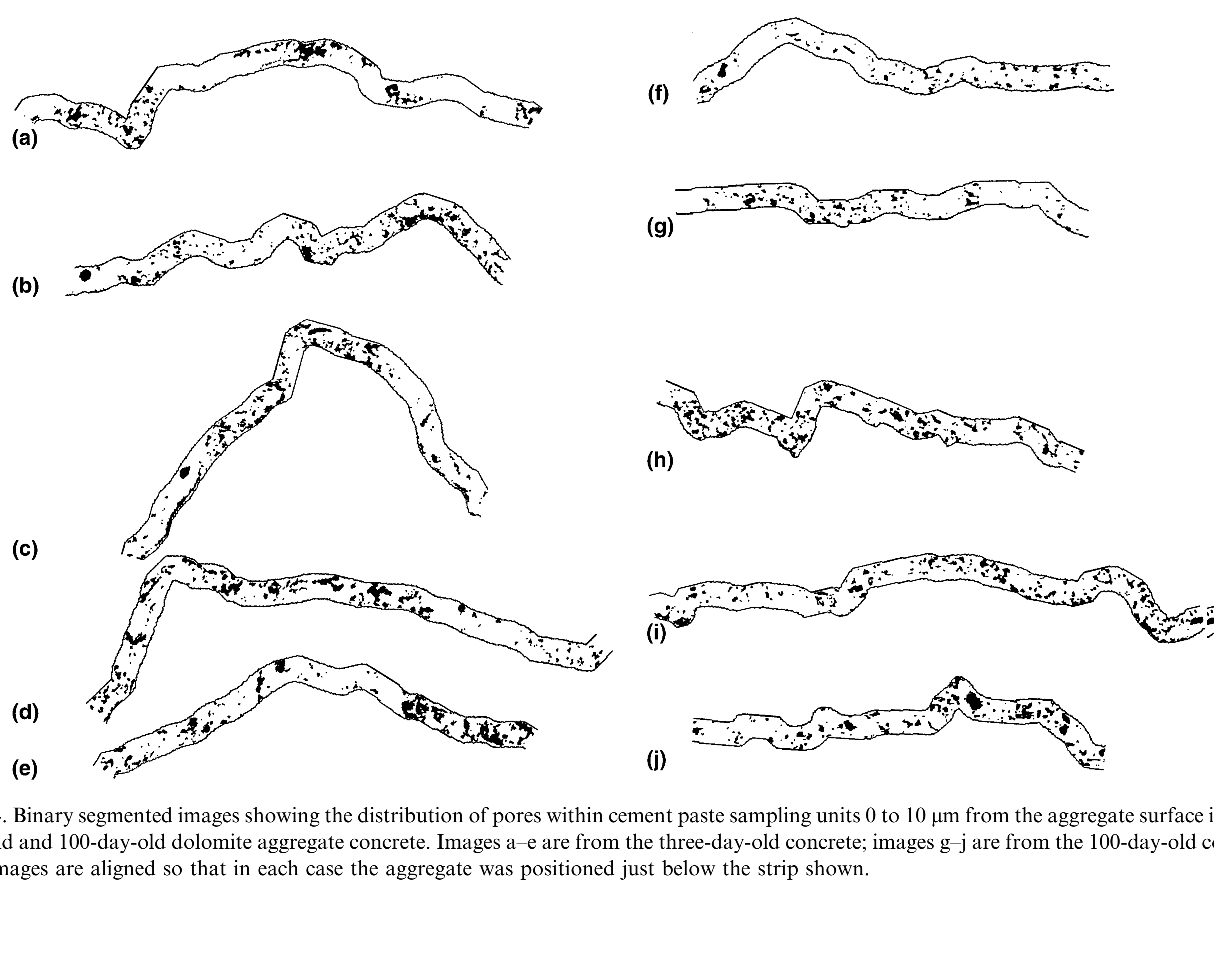

Fig. 4. Binary segmented images showing the distribution of pores within cement paste sampling units 0 to 10 μm from the aggregate surface in three-day-old and 100-day-old dolomite aggregate concrete. Images a–e are from the three-day-old concrete; images g–j are from the 100-day-old concrete. The images are aligned so that in each case the aggregate was positioned just below the strip shown.

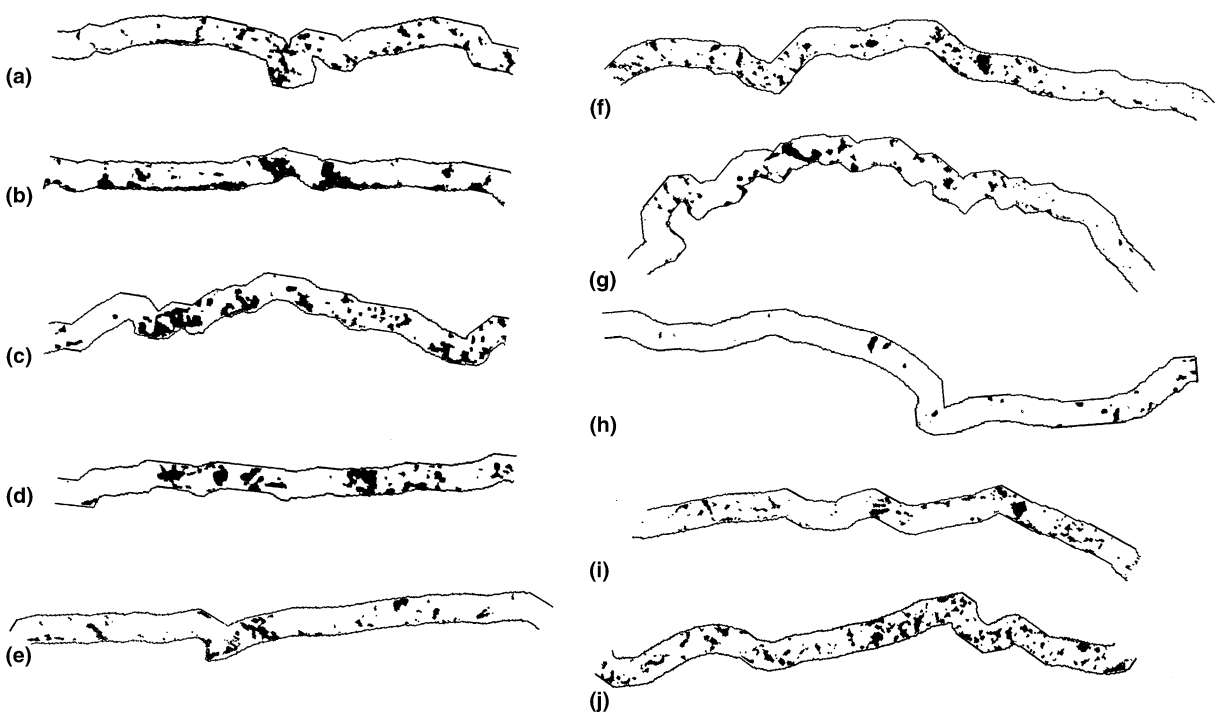

Fig. 5. Binary segmented images showing the distribution of pores within cement paste sampling units 0 to 10 μm from the aggregate surface in three-day-old and 100-day-old quartzite aggregate concrete. Images a–e are from the three-day-old concrete; images g–j are from the 100-day-old concrete. The images are aligned so that in each case the aggregate was positioned just below the strip shown.

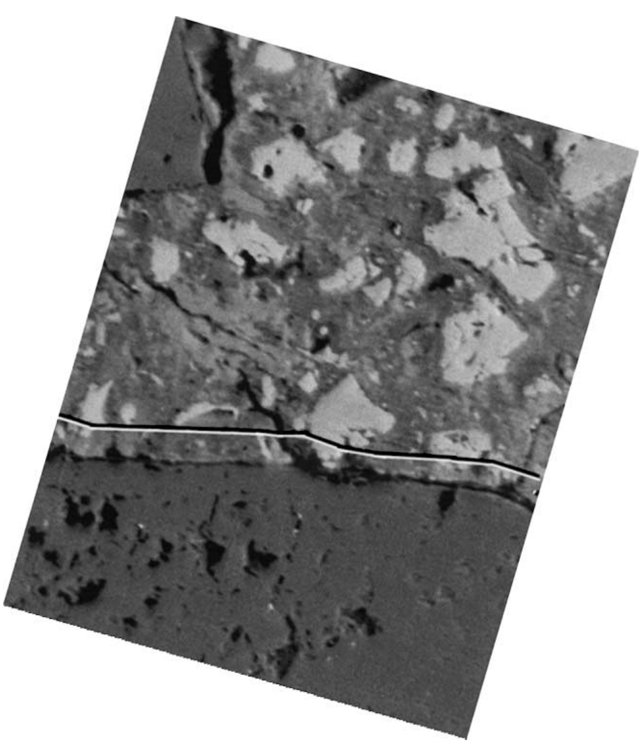

Fig. 6. Gray scale micrograph showing the area from which Fig. 5(b) was secured, aligned for comparison with the segmented binary pore image shown there, with the boundary at 10 μm approximately indicated.

particularly porous. However the bond between the dense paste and the aggregate is imperfect, and a gap exists several pixels wide. Additionally, a microcrack intersects the paste-aggregate contact near the center of the figure. These two local features combine to yield the unusual concentration of pore pixels at the aggregate-cement paste contact shown in Fig. 5(b). In no sense is the microstructure of the cement paste itself highly porous near the contact.

The irregular clustering of pores in some local areas within the various segmented images shown in Figs. 4 and 5, and their virtual absence in other areas, has already been referred to. In part this reflects the patchy and irregular character of the C–S–H within the aureole, which is similar to that typically found in the bulk cement paste as well. However, as is shown in the next section, some areas devoid of pores within the aureole represent local deposits of calcium hydroxide that are anchored on the aggregate. Such deposits are prominent features around the peripheries of many aggregate grains in all ordinary concretes.

5. The architecture of the "inner" ITZ in real concrete

The results of the image analyses investigations described here lead to a considerably different idea of the typical architecture of the ITZ in ordinary concrete than is usually maintained. While partial exclusion of the original cement particles was statistically effective within the aureole, even for well-mixed concrete, the consequences of this exclusion in terms of the porosity that remains after a few days seem to be much less important than has been previously considered. This is true even for the innermost portions of the aureoles. The reasons are apparent under close study of these areas.

Details of the architecture of the innermost zones of the aureole are actually relatively easy to illustrate. Fig. 7 shows a set of images taken from examinations of a three-day-old w:c 0.5 dolomite aggregate concrete, which was deliberately badly mixed. In contrast to our well mixed concretes, which were mixed for 10 min after water was added to the mixed dry components, these badly mixed concretes underwent a mixing period of only 30 s. It has been established that such bad mixing produces concrete in which all of the cement paste has a high content of coarse pores [6]. To show details more clearly, these images were produced at 1000× magnification using 1024 pixel resolution, rather than the 500× magnification and 512 pixel resolution usually employed to cover a wider area in most image analyses.

Fig. 7(a) shows the original micrograph. Fig. 7(b) shows an unsegmented 0–10 μm strip cut from this image, oriented parallel to its position in Fig. 7(a). Figs. 7(c)–(e) are segmented images of Fig. 7(b), showing respectively the spatial distributions of pore space, calcium hydroxide, and residual unhydrated cement.

Examination of the original image in 7(a) indicates that the cement paste in this badly mixed concrete is highly porous, but that the obvious porosity is not concentrated in or confined to the aureole; areas 100 μm distant from the perimeter of the aggregate appear to be equally porous. Certain portions of the paste, identifiable as either unhydrated cement particles or as calcium hydroxide, are not porous. There is a substantial content of unhydrated cement particles within the field, but as indicated in Fig. 7(e), almost none of it is within the inner 0–10 μm strip.

Attention is drawn to the upper right-hand portions of Figs. 7(c) and (d). It is evident from Fig. 7(c) that this section of the innermost strip contains no pores, and from Fig. 7(d) that it is almost entirely solid calcium hydroxide.

A different area around the perimeter of the same aggregate grain is shown in Fig. 8(a). In this area, as seen in Fig. 8(d), a deposit of calcium hydroxide around 70 μm long and about 10 μm wide occupies almost all of the area immediately adjacent to the aggregate. As is evident in Fig. 8(c), the content of pores in the area

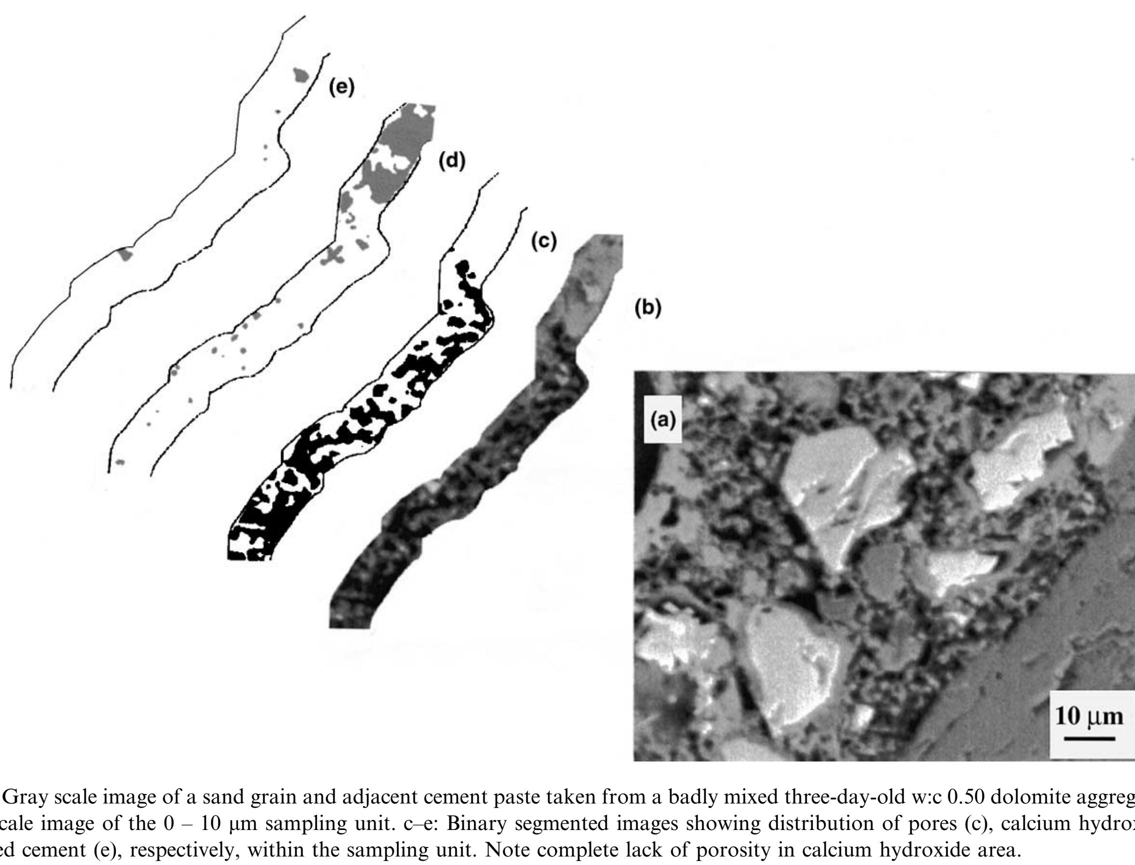

Fig. 7. a: Gray scale image of a sand grain and adjacent cement paste taken from a badly mixed three-day-old w:c 0.50 dolomite aggregate concrete. b: Gray scale image of the 0 – 10 μm sampling unit. c–e: Binary segmented images showing distribution of pores (c), calcium hydroxide (d), and unhydrated cement (e), respectively, within the sampling unit. Note complete lack of porosity in calcium hydroxide area.

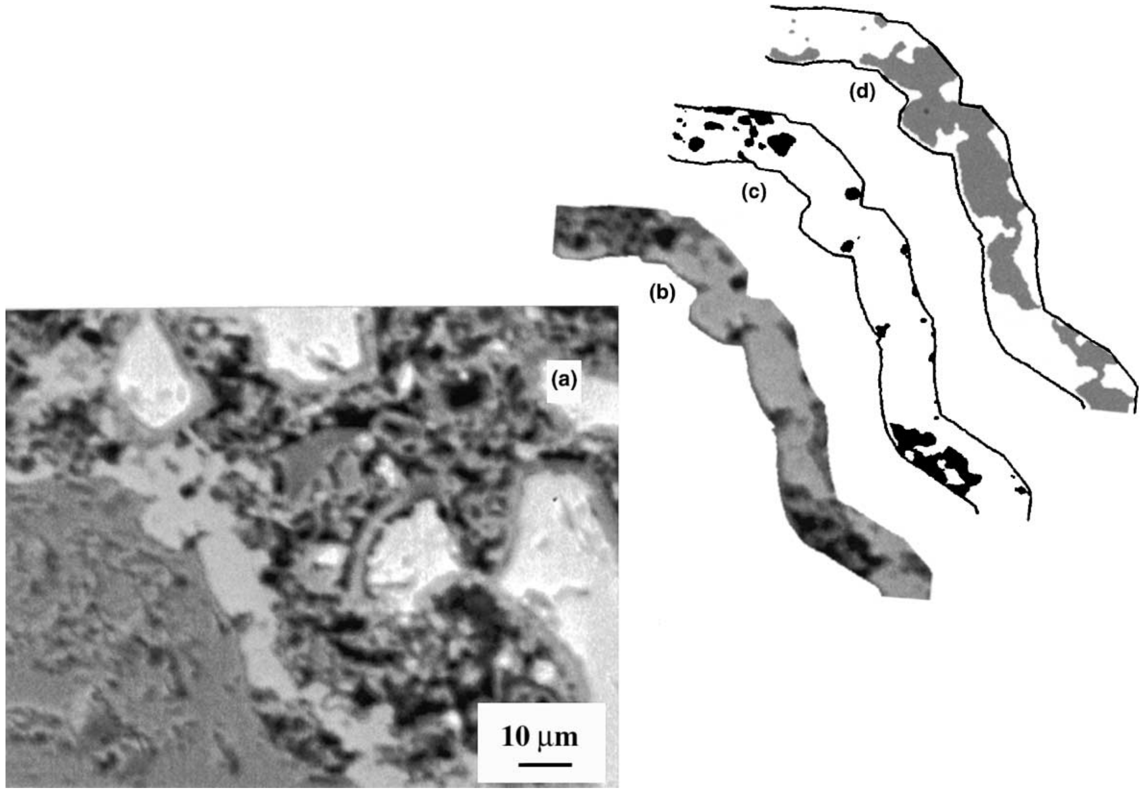

Fig. 8. Images of another area around the sand grain shown in Fig. 7. a: area image. b: Gray scale image of 0–10 μm sampling unit. c–d: Binary segmented images showing distribution of pores (c), and calcium hydroxide (d). In this segment of the perimeter the non-porous calcium hydroxide dominates the "close ITZ" region.

covered by the calcium hydroxide deposit is almost nil. Thus, in this area much of the aggregate is virtually sealed off from the surrounding porous cement paste by the inner layer of calcium hydroxide. Where such deposits occur, the innermost portion of the aureole is in fact *non*-porous rather than highly porous.

6. Discussion: resolution of the ITZ paradox

The implications of the data provided above require some discussion. The image analysis results described here indicate that no high concentration of pores is found in the aureole, even in its innermost portion. However, since the aureole was highly deficient in content of cement particles, it follows that the ITZ, and especially its innermost portion, must indeed have had substantial extra porosity at the start of hydration. Why then is so little of this extra porosity retained? This seeming paradox has important implications.

One pore-filling mechanism is evident from Figs. 7 and 8, in the presence of the calcium hydroxide that has precipitated extensively within the aureole, and especially against the aggregate surface itself. As indicated, for example by Nielsen [12] heterogeneous nucleation, i.e., nucleation onto solid surfaces from mildly supersaturated solutions, is a common phenomenon in physical chemistry. In the evolution of the structure of the ITZ, one would expect a number of small surface deposits to be produced by heterogeneous nucleation at various places around the surface of a given aggregate. One would also expect that over time the larger deposits will grow and the smaller ones will re-dissolve, a process analogous to the growth pattern of embryos that nucleate within in a homogeneous solution phase. Thus over time some areas on the surface of aggregates may be expected to be covered with calcium hydroxide, while others are free of it. Indeed, it has been observed that some small aggregate grains, for whatever reason, are entirely free of such deposits.

That such a pattern of calcium hydroxide deposition is common in ordinary concrete has been reported by other observers, including Scrivener, Bentur, and Pratt [10] and recently Kjellsen et al. [13]. These authors stated that their Fig. 1 illustrates that "an irregular layer of CH, apparently some 5 μm thick, is observed in contact with, or very close to the aggregate particle". They further indicated that there were also many aggregates or parts of aggregates that were *not* surrounded by layers of calcium hydroxide, an observation in accord with our own findings. The fraction of total aggregate surface covered by calcium hydroxide in various concretes has never been established to the writers' knowledge, but it certainly would constitute an appropriate topic for investigation.

Unfortunately, computer-based models of concrete that purport to represent the ITZ, such as the NIST models described by Bentz et al. [14] and more recently by Bentz and Garboczi [15] fail entirely to predict or record the existence of calcium hydroxide deposits in contact with aggregate surfaces.

It should be specifically recognized that not all of the calcium hydroxide found in the aureole around aggregate grains is the result of heterogeneous nucleation on their surfaces. Deposits of calcium hydroxide within the aureole, but not connected to the surface of the aggregate grain, are present in many of the areas examined. Presumably these precipitate by the normal processes of homogeneous nucleation in water-filled space within the aureole, as they would do within the bulk cement paste.

It thus appears that part of the extra pore space originally present within the aureole (and especially near the surface of the aggregate) is filled in by deposits of calcium hydroxide. However, since many areas are not so affected, this deposition of calcium hydroxide does not fully account for the degree to which the extra pore space near the aggregates ceases to exist in ordinary concrete.

An important observation can be made by summing the components of the cement paste that are separately capable of being segmented and quantified (pores, calcium hydroxide, and residual unhydrated cement) and subtracting them from 100%. The residue is essentially C–S–H, but contains ettringite and possibly other minor hydrated components, and includes some pores too fine to be detected. Unfortunately, because of locally varying composition and internal porosity, C–S–H cannot itself be segmented and evaluated directly.

Plots of such indirectly determined C–S–H vs distance from the aggregate surfaces are provided in Fig. 9 for three-day-old and for 100-day-old well-mixed dolomite aggregate concretes. It is evident that there is remarkably little change in average value of this C–S–H parameter with distance from the aggregate. One can detect a slight increase as the aureole is entered, and for the older concrete, a slight reduction in the innermost 0–10 μm layer, where calcium hydroxide deposits occupy some of the space that might otherwise be available for C–S–H.

The writers consider that the relative constancy of content of this C–S–H parameter is of great significance. To the extent that the value derived this way actually represents C–S–H, it indicates that, in the concretes examined, there is at least as much C–S–H *per unit volume of cement paste* within the aureole as outside of

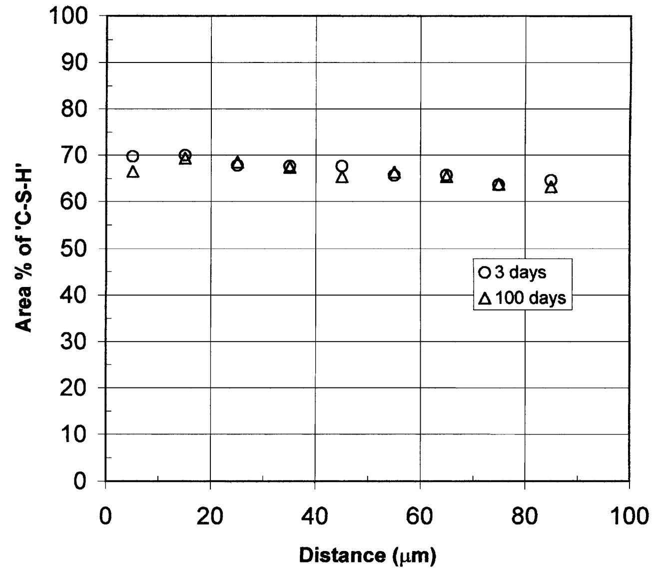

Fig. 9. Area % of 'C–S–H' (by difference) vs distance from the aggregate in three-day-old and 100-day-old well-mixed dolomite aggregate concretes.

it. Since the aureole starts out with a considerable deficiency of cement and a considerable excess of pore space in order to produce the same amount of C–S–H within it as outside of it, much of the C–S–H found within the aureole must have been derived from the more cement-rich region outside of it. This presumably takes place by the well-established through-solution C–S–H growth mechanism. Thus it appears that space-filling deposition of extra C–S–H within the aureole acts in concert with the deposition of extra calcium hydroxide to reduce what was initially high excess porosity to modest amounts such as seen in Fig. 3.

7. Discussion: to what extent does the ITZ affect concrete properties?

The technical literature over the past ten years or more contains dozens, or perhaps even hundreds of examples of reports ascribing major significance to the properties of the transition zone in controlling the behavior of ordinary concrete. This concept has been echoed in current standard textbooks on concrete; for example, Mehta and Monteiro [16] devoted a significant portion of their description of the structure of concrete to the ITZ and to its purported effects in weakening concrete and rendering it more permeable.

The results reported here on the structure of the ITZ indicate that there is little physical basis to expect such effects on concrete properties.

Some recent efforts to objectively assess the ITZ influence on concrete properties have tended to suggest that the ITZ is indeed not of major significance. For example, in their review of the many published papers of the purported effects of the ITZ on ionic diffusion and leaching in concrete, Marchand and Delagrave [17] concluded that as a practical matter, the major effects of aggregate particles in increasing the tortuosity far outweigh any percolative effect due to any possible overlapping of pores in the ITZ.

With respect to mechanical effects, as the result of an intensive review of many extremely complicated attempts to apply the concept of the ITZ to the mechanical behavior of concrete, Van Mier and Vervuurt [18] concluded that the conventional physical model in which the ITZ is treated as a separate mechanical phase "should perhaps be replaced" by a model in which "the conventional definition of the ITZ loses its meaning".

The present writers suggest that these judgments, arrived at by analyzing a large number of complex experiments and analytical models, tend to bear out the perfectly straightforward results of Rangaraju [19], as summarized previously [6]. Rangaraju varied the size distribution of the sand used in concretes without altering the total volume of sand. The finer sands generate progressively greater proportional volumes of ITZ, and closer grain-to-grain spacing, i.e., resulting in more overlap of aureoles. Nevertheless, rapid chloride permeability tests yielded almost identical results for all of the concretes of a given w:c ratio, regardless of degree of overlap of the aureoles. A similar lack of any effect produced by different proportions of ITZ paste in different concretes was found in measurements of tensile and compressive strengths and of dynamic elastic modulus.

The variant local architecture within the ITZ may have important effects in certain circumstances, however. As an illustration, in the special case of relatively permeable field concretes exposed to external solutions, as pointed out by Bonen [20], dissolution of some of the calcium hydroxide immediately surrounding aggregates may in fact open up channels of relatively easy transport and provide preferred sites for deposition of secondary reaction products. Even when this occurs, however, leaching does not necessarily proceed faster along the interfaces. Delagrave et al. [21] indicated that in well-cured mortars of reasonably low w:c ratios (0.45 and 0.25), a front of dissolution of calcium hydroxide proceeds uniformly down the specimens, with no preferential leaching or decalcification around the aggregates ahead of the front.

8. Conclusions

1. The ITZ or aureole de transition surrounding aggregate grains in concrete can be objectively designated as a volume from which portland cement particles have been preferentially excluded on a statistical basis. The effects of such statistical exclusion are manifested even after most of the cement is hydrated, in a well-defined gradient marking the reduced content of residual unhydrated cement with decreasing distance from the aggregate surface. This gradient often extends out to as much as 50 μm or more.
2. The highly enhanced porosity (up to 30% pore space) reported by others as being present within the boundary of the aureole, and especially within the "close interfacial region" within 5 μm from the aggregate, is not found with the concretes studied. These well-mixed concretes, of normal mix design, show only slightly greater average porosity within the aureole than elsewhere, and show no evident concentration of pores in the cement paste within a few μm of the aggregate.
3. Portions of the perimeters of many aggregates are covered by non-porous deposits of calcium hydroxide, which may extend continuously for many tens of μm along the perimeter. These deposits are taken to represent the effect of heterogeneous nucleation and subsequent crystal growth of calcium hydroxide along the surface of the aggregate. In addition, other crystals of calcium hydroxide nucleate elsewhere within the ITZ, as they do in the bulk cement paste.
4. Almost identical contents of C–S–H are found throughout the concrete, suggesting that within the aureole the content of C–S–H formed from the limited content of cement locally present is supplemented by C–S–H derived from cement-rich areas outside of the aureole.
5. It is thus considered that extra pore space originally associated with the wall effect in ordinary concrete is mostly filled up by deposits of calcium hydroxide and by deposits of C–S–H passing through solution, leading to the retention of only a modest residual extra average pore content within the ITZ.
6. These considerations lead to the judgment that in ordinary concrete the existence of the ITZ around aggregates can have only marginal effects, if any, on both mechanical properties and on permeance.

Acknowledgements

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