Analytical Solution for ITZ Thickness around Convex Shaped Aggregate Grains based on Section Analysis Method

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Summary

Some related theorems obtained by application of geometrical probability theory to plain sections of aggregate grains in concrete have been introduced. They allow relating mean apparent ITZ thickness and the actual ITZ thickness. The analytical solutions are derived for 2D and 3D convex shaped aggregate grains.

Introduction

The section analysis method has been used commonly for studying the microstructure of concrete, and more recently also for that of the ITZ. Generally, the ITZ thickness is used as a suitable parameter to characterize the influence of factors on the ITZ microstructure. The actual thickness (t) is not revealed however, since the chance of having a section plane perpendicular to the surface of a spherically-shaped grain is negligibly small. Further, the additional complication of arbitrary shaped of grains should be faced in designing a strategy for solving the analytical problem. The apparent ITZ thickness (t') in a random section will exceed therefore the actual one. To what degree the apparent ITZ thickness overestimates the actual one can be assessed by geometrical statistical (*i.e.*, stereological) methods. Stroeven [1] was probably the first to derive by stereological means the ratio of the statistical mean apparent ITZ thickness over the actual one for spherical-shaped aggregate particles. The authors generalized this relationship for the 2D case to also encompass rectangular and ellipse-shaped particle sections [2]. The results revealed the grain shape to exert effects on the afore-mentioned ratio of ITZ extensions. No further information on this topic is provided by the literature. In this contribution, analytical expressions are developed for this size ratio for 2D and 3D cases of convex aggregate grains of arbitrary shape.

Mathematic Description of Physical Problem

For convenience, the following assumptions are made:

(1) Concrete is composed of three phases: aggregate grains, ITZ and bulk paste. Each aggregate grain is surrounded by an ITZ with a *fixed* thickness t. So, aggregate grain surface and outer boundary of ITZ are concentric. The remaining space of concrete is filled with bulk paste;

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(2) Surface spacing between adjacent aggregate particles is large enough to exclude interference of ITZ microstructures around particles;
(3) The region occupied by an aggregate particle constitutes a bounded closed convex region;
(4) Section planes occur with equal probability as to position and direction.

An arbitrary section plane through the concrete body is equivalent to a family of parallel lines in this plane. Hence, the mean apparent ITZ thickness obtained by the randomized section plane method is the same as the statistical mean intercept length obtained by a random line intersecting with the convex ring (for 2D case) or convex shell (for 3D case) of the ITZ. The problem of assessing the mean apparent ITZ thickness can be converted, as a consequence, into the following analytical problem.

$\mathbf{K_1}$ represents the bounded closed convex region occupied by aggregate particle. $\mathbf{K_2}$ does so for the bounded closed convex region occupied by both aggregate grain and ITZ. Then, the region occupied by the ITZ is represented by ($\mathbf{K_2}$-$\mathbf{K_1}$). So, the assessment of the statistical mean apparent ITZ thickness can be conducted along the following steps: (1) calculate mean intercept length ($\overline{l_1}$) between a random set of straight lines and $\mathbf{K_1}$; (2) calculate the mean intercept length ($\overline{l_2}$) between a random set of infinitely long straight lines and $\mathbf{K_2}$; (3) replace $\overline{l_2}$ by $\overline{l_2}'$ on the basis of arguments given below; (4) determine the analytical formula for t'; (5) assess the analytical expression for the ratio of t'/t.

2D Aggregate Particles

Although real objects in the real world are 3 dimensional, it is still attractive to study the 2D case. This allows building up the stereological framework, but it also appeals to common experimental practice in materials technologies. So, in this section, both aggregate grain and the ITZ are regarded as 2D objects, while randomization procedures are also performed in the 2D plane.

According to the afore-mentioned assumptions, $\mathbf{K_1}$ represents the region occupied by the convex shaped 2D aggregate particles. A_1 and C_1 is the area and perimeter length of boundary curve of $\mathbf{K_1}$, respectively. $\mathbf{K_2}$ is obtained by expanding $\mathbf{K_1}$ in all directions by a given value equal to t. So, $\mathbf{K_1}$ and $\mathbf{K_2}$ constitute a pair of concentric convex sets. ($\mathbf{K_2}$-$\mathbf{K_1}$) stands for the ITZ. Finally, A_2 and C_2 define the area and perimeter length of boundary curve of $\mathbf{K_2}$, respectively. $\mathbf{K_1}$ is intersected by a family of straight lines parallel to one axis (say, y-axis) of an orthogonal 2D Cartesian coordinates system. Assume the projected (shadow) length of C_1 onto x-axis is $\overline{l}'_{s1,\perp X}$. Integration of successive intercept lengths yields the area of $\mathbf{K_1}$. According to Cauchy's[3], the average intercept length along the y-axis ($\bar{l}^{Y}$) is $\bar{l}^{Y} = A_1/\overline{l}'_{s1,\perp X}$, whereby the denominator is the average projected

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length of C_1 onto an axis for randomized orientation. According to Crofton's theorem [4], this average projected length ($\bar{l}_1$) is $\bar{l}_1 = \pi A_1/C_1$. Similarly, the mean intercept length ($\bar{l}_2$) between randomly oriented infinitely long straight lines and C_2 is $\bar{l}_2 = \pi A_2/C_2$.

Hence, for the convex grain with any shape, the equations of $\bar{l}_1$ and $\bar{l}_2$ offer unbiased and exact estimates. Although it seems that the mean apparent ITZ thickness would be equal to $0.5(\bar{l}_2 - \bar{l}_1)$, this is incorrect. Among the families of parallel lines on which the equation of ($\bar{l}_2 = \pi A_2/C_2$) is based, only those intersecting with $\mathbf{K_1}$ are valid. Some of these families intersect with the ITZ without doing so with the aggregate grain. So, in the denominator of this equation C_1 should replace C_2. Similarly, among the families of parallel lines that intersect with $\mathbf{K_2}$, only the part intersecting with $\mathbf{K_1}$ is valid. All invalid intercepts are located in the region ($\mathbf{K_2}$-$\mathbf{K_1}$), but they occupy only part of this region. So, A_2 in the nominator of equation $\bar{l}_2 = \pi A_2/C_2$ should be replaced by $[A_2 - k(A_2 - A_1)]$, where k is a coefficient with a value less than 1.The exact value of k depends on the shape of the aggregate, as well as on the relative value of the ITZ thickness. Therefore, the corrected value of the mean intercept length $\bar{l}_2'$ between randomly oriented infinitely long straight lines and $\mathbf{K_2}$ is $\bar{l}_2' = \pi[A_2 - k(A_2 - A_1)]/C_1$. The mean apparent ITZ thickness (t') is

$$t' = 0.5(\bar{l}_2' - \bar{l}_1) = 0.5\pi(1-k)(A_2 - A_1)/C_1 \tag{1}$$

Eq. (1) offers the generalized expression for the mean apparent ITZ thickness, with the only limitation of convexity of the 2 D profile of the aggregate grain.

The purpose of estimating the mean apparent thickness of the ITZ is, of course, to assess its actual thickness. So, this relationship should be established next. According to Santaló's description [5], $A_2 = A_1 + C_1 t + \pi t^2$.So, combining this with eq. (1) yields

$$t' = \frac{\pi}{2}(1-k)\left(t + \frac{\pi}{C_1}t^2\right) \underset{\textbf{then } k\to 0_1}{\overset{\textbf{if } t^2 << C_1}{\approx}} \frac{\pi}{2}t \tag{2}$$

Obviously, because $(\pi t/C_1)>0$, t'/t will exceed $\pi(1-k)/2$, and to an increasing degree at declining size of the 2D convex grain. Hence, the shape of the aggregate grain and the actual ITZ thickness will affect the mean apparent ITZ thickness, t'. Also, the value of k is affected by the shape of the grains, although an exact value will be difficult to derive. Now, let's assume that the influence of shape of the aggregate grain on k can be neglected with respect to the influence exerted by ratio of ITZ thickness over grain section size. In that case, t'/t will reach its maximum value when the shape of the 2D convex aggregate grain is a circular disk. For relatively large aggregate grains, where the actual ITZ thickness is negligibly small with respect to grain size, *t'/t* will be approximately equal to $\pi/2$.

3D Aggregate Particles

The region occupied by a 3D aggregate grain still can be emphasized as a 3D bounded closed convex set $\mathbf{K_1}$. Similarly, the 3D bounded closed convex set $\mathbf{K_2}$ is obtained by expanding $\mathbf{K_1}$ in all direction by a fixed amount t. Therefore, $\mathbf{K_1}$ is a subset of $\mathbf{K_2}$. Further, the shell ($\mathbf{K_2}$-$\mathbf{K_1}$) represents the ITZ. Assume, as before, that volume and surface of $\mathbf{K_1}$ and $\mathbf{K_2}$ are V_1 and S_1, and V_2, and S_2, respectively. Let a family of infinitely long straight lines parallel to one axis of a Cartesian coordinate system (say, the z-axis) pass through $\mathbf{K_1}$. Denote the projection (shadow) area of $\mathbf{K_1}$ onto the xy-plane by $S'_{s1,\perp XY}$. Integration of the successive intercept lengths yields $\mathbf{K_1}$. Because the mean intercept length along z-axis is required, the area of this parallel family of lines meet $\mathbf{K_1}$ onto the plane perpendicular to the lines (here it is xy-plane) should be obtained. Actually, the projected area of this parallel family that meet $\mathbf{K_1}$ onto the xy-plane perpendicular to the lines is equal to the shadow area of the project cast by $\mathbf{K_1}$ on the plane perpendicular to these lines. According to the extended Cauchy's theorem [3], the average intercept length along the z-axis ($\bar{l}^Z$) is $\bar{l}^Z = V_1/S'_{s1,\perp XY}$. The mean intercept length requires an additional averaging operation for a random set of parallel families in all directions. So, it is necessary to calculate the mean shadow area $\bar{S}'_{s1}$ of $\mathbf{K_1}$ for this random set of orientations. According to Cauchy's [5,6], $\bar{S}'_{s1} = (S_1)/4$. So, the mean intercept length, $\bar{l}_1$, between randomly oriented infinitely long straight lines and the 3D convex aggregate grain is $\bar{l}_1 = (4V_1)/S_1$. Similarly, the mean intercept length, $\bar{l}_2$, between randomly oriented infinitely long straight lines and $\mathbf{K_2}$ is $\bar{l}_2 = (4V_2)/S_2$.

It is incorrect to directly use $0.5(\bar{l}_2 - \bar{l}_1)$ to calculate the mean apparent ITZ thickness. Among the families of parallel lines, only the part intersecting with $\mathbf{K_1}$ is valid. So, in the equation $\bar{l}_2 = (4V_2)/S_2$, the denominator should be S_1 rather than S_2. Similarly, among the intercept lengths families of parallel lines intersecting with $\mathbf{K_2}$, only the part intersecting with $\mathbf{K_1}$ is valid. The invalid intercepts are located in the region ($\mathbf{K_2}$-$\mathbf{K_1}$). Hence, V_2 in the nominator of $\bar{l}_2 = (4V_2)/S_2$, should be replaced by $[V_2 - k(V_2-V_1)]$, where k is a coefficient with a value smaller than 1. The value of k is governed by the shape of the aggregate grain and by the actual ITZ thickness. Therefore, the corrected mean intercept length, $\bar{l}_2'$, is given by $\bar{l}_2' = 4[V_2 - k(V_2-V_1)]/S_1$. Therefore, the mean apparent ITZ thickness, t', is

$$t' = 0.5(\bar{l}_2' - \bar{l}_1) = 2(1-k)(V_2 - V_1)/S_1 \tag{3}$$

Eq. (3) offers a generalized expression for the mean apparent ITZ thickness for bounded closed convex sets (particles). No other assumption as to the particle shape is required.

The next step is the development of a quantitative relationship between the mean apparent ITZ thickness and the actual ITZ thickness. As in section2.2, we assume that the mean width of $\mathbf{K_1}$ is B_1, Michielsen *et al*. [8] provided a quantitative relationship between the volumes of $\mathbf{K_1}$ and $\mathbf{K_2}$ based on Steiner's formula [5,7]

$$V_2 = V_1 + S_1 t + 2\pi B_1 t^2 + \frac{4}{3}\pi t^3 \tag{4}$$

Upon substitution of eq. (4) into eq. (3) the relationship between mean apparent ITZ thickness t', and the actual ITZ thickness t, is obtained

$$t' = 2(1-k)\left(t + \frac{2\pi B_1}{S_1}t^2 + \frac{4\pi}{3S_1}t^3\right) \underset{\text{then } k\to 0}{\overset{\text{if } t<<S_1}{\approx}} 2t \tag{5}$$

Obviously, because $(2\pi B_1 t)/S_1 > 0$ and $(4\pi t^2)/(3S_1) > 0$, t'/t is exceeding $2(1-k)$. However, the apparent mean ITZ thickness will only be influenced by the shape of the aggregate grains and by the actual ITZ thickness when the actual ITZ thickness and the grain size are comparable. Also, the value of k is affected by the shape of a 3D convex aggregate, but determination is complicated. Now, we assume, as we did earlier, that the influence of shape of the aggregate grain on k can be neglected with respect to the influence exerted by ratio of ITZ thickness and grain size. In that case, t'/t will reach its maximum value when the shape of the aggregate grain section is spherical. For relatively large aggregate grains, where the actual ITZ thickness is negligible small with respect to grain size, t'/t will be approximately equal to 2.

Conclusions

In this paper, the authors borrowed the theorems and results from geometric probability to build the quantitative relationship between the mean apparent ITZ thickness (t') and the actual ITZ thickness (t) for the convex 2D and 3D aggregate with any shape. The analytical results showed that:

(1)For any bounded closed convex 2D aggregate grain, $t' = 0.5\pi(1-k)(t+\pi t^2/C_1)$, where C_1 is the perimeter of boundary of aggregate grain, k is the constant coefficient related to the shape of the aggregate and the actual ITZ thickness($k<1$). Since the value of both k and C_1 are related to the shape of the aggregate grain, the ratio of t'/t will be influenced by the shape of aggregate grains. If the actual ITZ thickness is far less than the size of aggregate grain, then the ratio of the mean apparent ITZ thickness (t') to the actual ITZ thickness (t) is approximately equal to $\pi/2$.

(2)For any bounded closed convex 3D aggregate grain, $t' = 2(1-k)[t+2\pi B_1 t^2/S_1+4\pi t^3/(3S_1)]$, where, S_1 and B_1 are surface area of boundary of aggregate grain and mean breath of aggregate grains, k is a constant coefficient related to the shape of the aggregate and the actual ITZ thickness(k<1). Since k, S_1 and B_1 are related to the shape of grain, the ratio of t'/t will be influenced by the shape of aggregate grains. If the actual ITZ thickness is far less than the size of aggregate grain, then the ratio of the mean apparent ITZ thickness (t') to the actual ITZ thickness (t) is approximately equal to 2.

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