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Overestimation of ITZ thickness around polygon-shaped aggregate

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ABSTRACT

The interfacial transition zone (ITZ) between cement paste and aggregate particle is the weakest part of composites. In order to quantify the influence of the ITZ on the macro-properties of composites, the volume fraction of the ITZ need to be considered. For circular particle, it is easy to construct ITZ layer by expanding the circle by a given actual thickness t. However, little work was available in the literature on how to numerically construct the ITZ around non-circular aggregate. This paper proposed the construction of ITZ around regular polygons. Then, statistical average values of the apparent ITZ thickness t' in all potential directions are computed by using the systematic line sampling algorithm. Finally, the volume of ITZ can be calculated, this contribution is helpful to further understand the influence of the aggregate shape to the macro-properties of composites.

1. INTRODUCTION

The interfacial transition zone (ITZ) between cement paste and aggregate particle is commonly regarded as a weak link in determining both the mechanical and transport properties of cement-based composites (Chen 2007, Garboczi 1997, Liu 2014, Scrivener 1988, Xu 2013, Xu 2014). In order to quantify the influence of the ITZ on the macro-properties of concrete, the volume fraction of the ITZ should be derived. However, the constituents of concrete are opaque, sectional plane analysis (Zhu 2014) is used to study the ITZ microstructure. In practice, ITZ thickness is obtained by quantitative backscattered electron image analysis from a polished sectioning plane of concrete sample (Gao 2013, Scrivener 1988), as shown in Fig. 1(a). Normally, the

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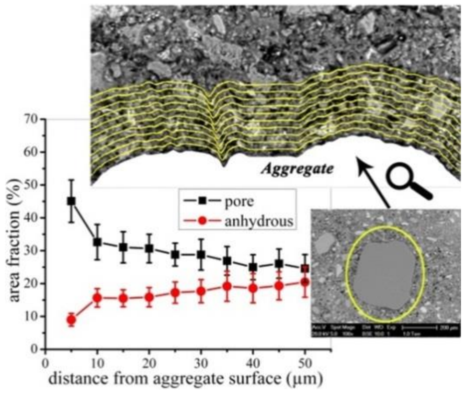
apparent ITZ thickness *t'* is bigger than the actual ITZ thickness *t*, because not all of the sampling line is perpendicular to the boundary of aggregate, as shown in Fig. 1(b). Therefore, it is necessary to calculate the degree of overestimation between apparent and actual ITZ thickness. Chen (2007) has developed a formula in Eq.(1) to calculate the overestimation of ITZ thickness around arbitrary convex-shape aggregate particles.

$$t' = \frac{\pi}{2}(1 - k_{invalid})\left(t + \frac{\pi}{C}t^2\right) \tag{1}$$

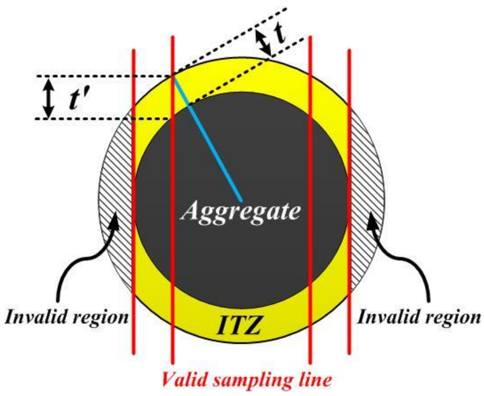
where *t'* and *t* are the apparent and actual ITZ thickness, respectively. *C* is the perimeter of the aggregate. $k_{invalid}$ means the area ratio of invalid region to the ITZ layer, as shown in Fig. 1(b). For 2D circular aggregate with $x = t/r$, r is the radius of circle, the invalid coefficient is given by Eq.(2).

$$k_{invalid} = \frac{2\left[(1+x)^2 \arccos\left(\frac{1}{1+x}\right) - \sqrt{2x + x^2}\right]}{\pi(2x + x^2)} \tag{2}$$

However, the formula only is verified reliability for circle and sphere due to the difficulty of constructing the ITZ layer around non-circular/non-spherical particle. The reliability of the generalized formula on non-spherical particle system has been described in another paper (Zhu et al. 2014). This paper aims to extend the work to the non-circular (i.e. regular polygons) particle system.



(a) ITZ thickness around aggregate from SEM image, (Gao et al. 2013)



(b) Overestimation of ITZ thickness around a circular aggregate

Fig. 1 Overestimation of ITZ thickness by sectional plane analysis

This paper proposed a method to construct the ITZ layer around regular polygon particle and the overestimation of ITZ thickness are obtained by using the systematic line sampling algorithm. And then, the generalized formula needs to be verified for regular polygon particle. Furthermore, the overestimation degree of ITZ thickness around a single particle and multi-sized particle system should be discussed. At last, the overestimation of ITZ volume fraction and diffusivity of concrete are can be calculated.

2. ITZ CONSTRUCTION AROUND 2D REGULAR POLYGONS

2.1 ITZ layer construction

In two-dimensional (2D) case, the construction process of the ITZ layer around regular convex polygon is very similar to the dilation operation in mathematical morphology (Serra 1988), which is strongly related to the Minkowski sum (addition).

$$X \oplus Y = \{x + y \mid x \in X, y \in Y\} \tag{3}$$

where X and Y are two sets of vectors in Euclidean space. In this paper, X is considered to a regular polygon particle, Y is a circle. Herein, regular polygon is a polygon that is both equilateral and equiangular in geometry. It means that has all edges the same length and all interior angles the same. We define a symbol “PN”: it represents that aggregate shape is regular polygon with N-edges, where N is the number of edges of regular polygon particle. For example, P3 means an equilateral triangle; P4 represents a square as shown in Fig. 2. The area A of a regular polygon with N-edges is given in Eq.(4).

$$A = \frac{1}{4} N a^2 \cot(\frac{\pi}{N}) \tag{4}$$

where *a* is the length of edge, *N* is the number of edges. The perimeter *C* of a regular polygon with N-edges can be calculated from Eq.(5).

$$C = Na \tag{5}$$

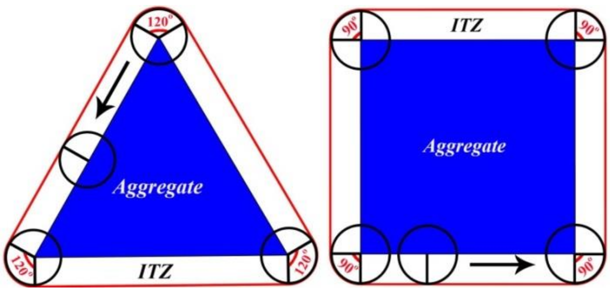


Fig. 2 The construction of ITZ layer around P3 and P4

For aggregate particle with different shape, we need to discuss the influence of particle shape under the same area. Herein, the equivalent circle diameter and circularity are defined. The equivalent circle diameter of a given particle D_{eq} is the diameter of a circle with the same area as the given particle, $D_{eq} = (4A/\pi)^{1/2}$. The relationship between the length of edge and D_{eq} can be written as

$$a = \sqrt{\frac{2\pi}{N\sin(2\pi / N)}} \sin\left(\frac{\pi}{N}\right) D_{eq} \tag{6}$$

Circularity is defined as perimeter ratio of particle and circle with the same area (He 2010), it can be obtained by Eq.(7). Fig. 3 shows that circularity will close to 1 (circularity of circle is 1) with the number of edges of regular polygon, this is consistent with the definition of circle which is composed of an infinite number of edges.

$$c = \frac{2\pi\sqrt{A/\pi}}{C} \tag{7}$$

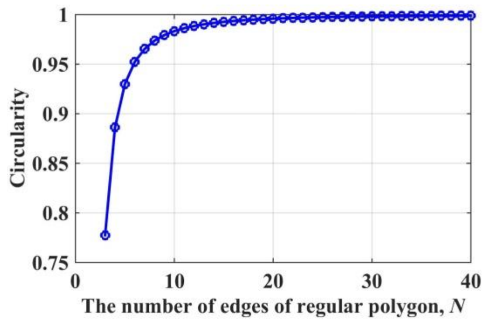


Fig. 1 Circularity of regular polygon

In geometry, the ITZ layer around a regular polygon can be considered as an extra increased region by rolling through a circle with radius *t* (*t* is the ITZ thickness) and its center is exactly located on the boundary of polygon. Obviously, the ITZ layer of regular polygon can be decomposed into two parts, i.e., edge part and vertex part, as shown in Fig. 2 and Fig. 4. The edge part of ITZ layer is parallel to the original edge of regular polygon, and the translational displacement along the normal line of edge is t. Vertex part of ITZ layer is a circular arc, which is a set of all points at a given distance t (radius) away from the vertex, and the angle of circular arc is $2\pi/N$.

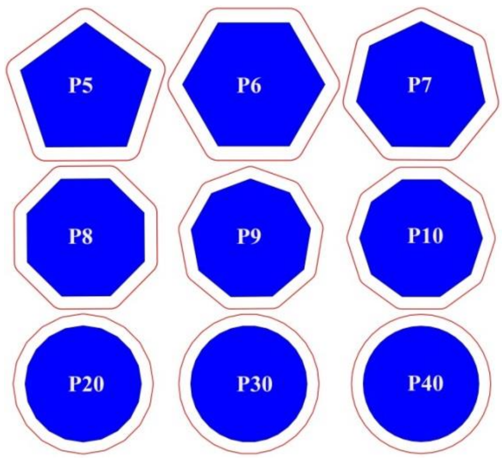


Fig. 4 The construction of ITZ layer around P5~P10, P20, P30 and P40

2.2 Systematic line sampling (SLS) algorithm

For a sectional plane of ITZ layer around regular polygon, the apparent ITZ thickness can be obtained by a series of valid sampling lines which is defined as a sampling line which intersects with both the ITZ and aggregate, as shown in Fig. 1(b). Due to the anisotropy of regular polygon particle, the apparent ITZ thickness should be calculated by average over all planar angles. Now, the question is to determine that how many sampling lines are appropriate for regular polygon with the same area. This can be solved by systematic line sampling algorithm. The SLS algorithm can be described as two aspects: the number of planar angles of sampling line $N_\alpha = \pi/\Delta\alpha$, and the spacing ΔL between two adjacent sampling lines, where $\Delta\alpha$ is the planar angle step between two adjacent sampling lines direction, as shown in Fig. 5.

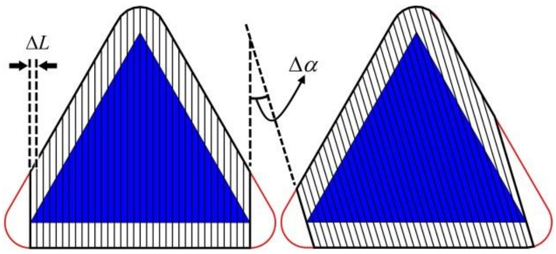


Fig. 5 The planar angle step Δα and spacing ΔL between sampling lines

Firstly, we need to determine the number of planar angles N_α under the assumption of ΔL:D_{eq}=0.001:2 and t:D_{eq} = 0.01. The ratio of apparent and actual ITZ thickness t'/t can be calculated by using the SLS algorithm, Fig. 6 shows that the curves of t'/t becomes stable with the increasing of N_α, i.e., the decreasing of planar angles step Δα. When N_α reach to 120, the results of t'/t can approximately stand for their true values. Meanwhile, the degree of overestimation of *t'/t* will increase with the decreasing circularity of regular polygon as shown in Fig. 6. Therefore, the fixed value of N_α=120 is used to determine the suitable value of the spacing.

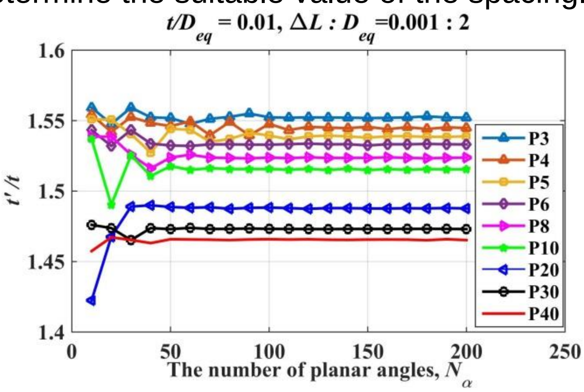


Fig. 6 Determination of planar angles step Δα

Secondly, the determination of the spacing ΔL is obtained by the assumption of t:D_{eq} = 0.01 and N_α=120. The ratio of t'/t in Fig. 7 becomes stable when the spacing reach to ΔL:D_{eq}=0.001:2. So, the rules of N_{α}=120 and ΔL:D_{eq}=0.001:2 is employed as the line sampling in the following sections.

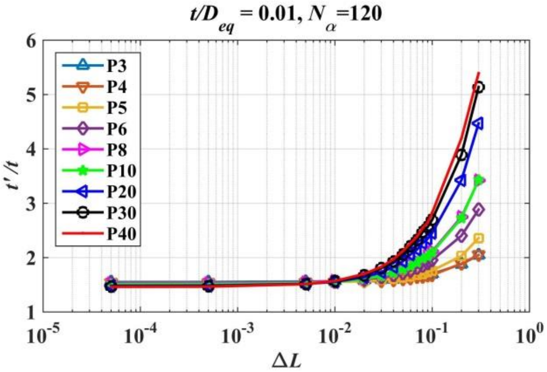
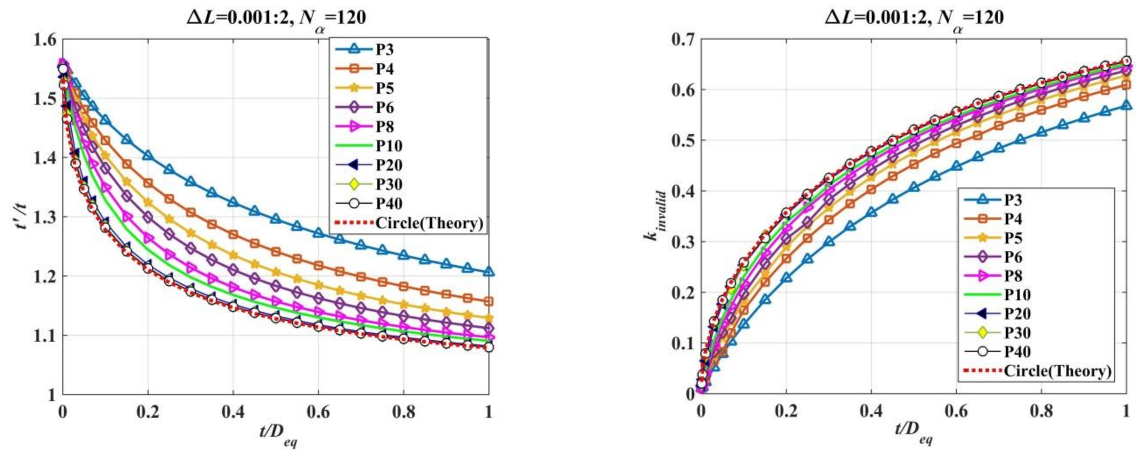


Fig. 7 Determination of the spacing ΔL

2.3 Overestimation of ITZ thickness

Since Chen et al. (2007) only provided a theoretical formulae of *t'/t* and $k_{invalid}$ for a 2D circular particle as shown in Eq.(1) and Eq.(2). No further verification process was conducted on other particle shape. Therefore, this section extends previous work to verify the reliability of the generalized formula for regular polygons by using the above line sampling rule (N_{α}=120 and ΔL:D_{eq}=0.001:2). It can be seen from Fig. 8 that the simulation results of *t'/t* and $k_{invalid}$ for a P40 are consistent with the theoretical values of the circle. This demonstrates the reliability and efficiency of generalized formula Eq.(1) for regular polygon and circle particles.



(a) *t'/t* (b) $k_{invalid}$

Fig. 8 Effect of actual ITZ thickness t/D_{eq} on *t'/t*

Moreover, Fig. 8(a) also indicates that the value t'/t reduces with the increasing t/D_{eq} and circularity of regular polygon. On the contrary, the invalid coefficient $k_{invalid}$ (as shown in Fig. 8(b)) increases with the increasing t/D_{eq} and circularity of regular polygon.

In practically, the apparent ITZ thickness around aggregate particles always has different sizes in a sectional plane. So, it is necessary to consider the effect of particle size distribution (PSD) on statistical average apparent ITZ thickness. Normally, Fuller distribution with volume-based cumulative probability function $F_V(D_{eq})$ as shown in Eq.(8) is commonly used to characterize aggregate size distribution in concrete.

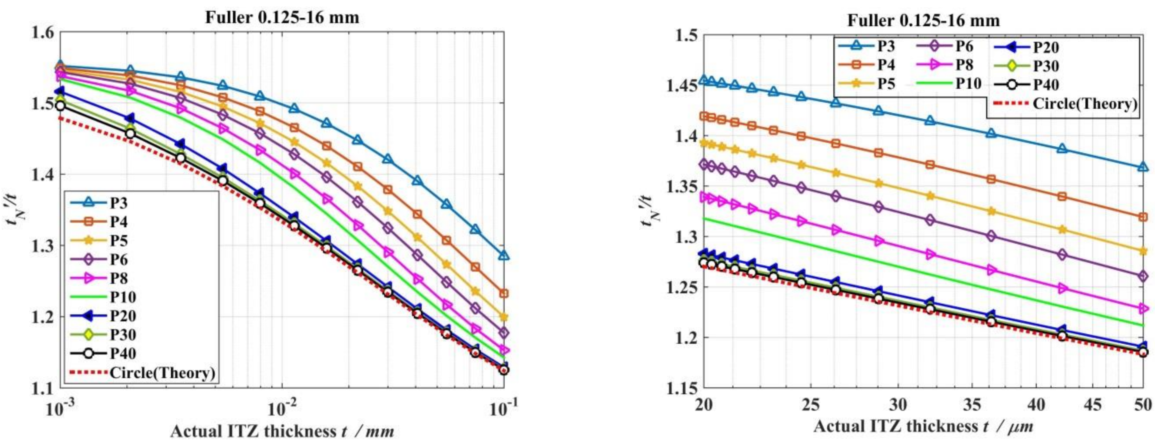
$$F_V(D_{eq}) = \frac{\sqrt{D_{eq}} - \sqrt{D_{eq\_\min}}}{\sqrt{D_{eq\_\max}} - \sqrt{D_{eq\_\min}}} \tag{8}$$

where $F_V(D_{eq})$ is the volume-based cumulative probability function, D_{eq_min} and D_{eq_max} respectively represent the minimum and maximum equivalent circular diameter of a particle. The number-based probability density function $f_N(D_{eq})$ in Eq.(9) is used to generate the number of aggregates with different size.

$$f_N(D_{eq}) = \frac{5}{2D_{eq}^{7/2}(D_{eq\_\min}^{-5/2} - D_{eq\_\max}^{-5/2})} \tag{9}$$

For multi-scale aggregate system with Fuller distribution, the statistical average of the apparent ITZ thickness can be expressed as

$$t_N' = \int_{D_{eq\_\min}}^{D_{eq\_\max}} t' f_N(D_{eq}) dD_{eq} \tag{10}$$



(a) 1-100 μm (b) 20-50 μm

Fig. 9 Effect of actual ITZ thickness on t_N'/t

For different regular polygon shape, their statistical average of the apparent ITZ thickness is computed by SLS algorithm. The results of t_N'/t of regular polygon are shown in Fig. 9. Practically, the ITZ thickness was generally regarded within the range of 20~50 μm (Scrivener 2004), the corresponding overestimation of ITZ thickness varies from 1.186 to 1.455 times for regular polygon and from 1.183 to 1.270 times for circle, respectively, as shown in Fig. 9(b).

3. OVERESTIMATION OF ITZ VOLUME FRACTION

When the statistical mean of the overestimation t_N'/t which follows Fuller distribution are known, the analytical formula on ITZ volume fraction V_{ITZ} can be calculated. And then the overestimation degree of V_{ITZ} can be determined. Garboczi (1997) proposed an analytical formula in Eq.(11) to calculate V_{ITZ} for multi-sized spherical aggregate system. Recently, Xu (2013) extended the formula to the multi-sized Platonic particle system.

$$V_{ITZ}(t) = 1 - V_{agg} - e_V(t) \tag{11}$$

where V_{ITZ} and V_{agg} is the volume fraction of ITZ and aggregate, respectively, and $e_V(t)$ is the volume fraction of matrix in the concrete. For two-dimensional polydispersed hard disks systems, Eq.(12) is given by Lu and Torquato (Lu 1992).

$$e_V(t) = (1 - V_{agg})\exp\left\{-\pi N_A\left[\frac{t^2 + \overline{D_{eq}}\,t}{c(1-V_{agg})} + \frac{\pi N_A \overline{D_{eq}}^2 t^2}{4c^2(1-V_{agg})^2}\right]\right\} \tag{12}$$

where c is circularity representing a shape descriptor of particles, N_A is the number density per unit area of hard particle in polydispersed systems, i.e.,

$$N_A = \frac{4V_{agg}}{\pi \overline{D_{eq}^2}} \tag{13}$$

An average over a particle size distribution of D_{eq} is given in Eq.(14).

$$\overline{D_{eq}^n} = \int_{D_{eq\_\min}}^{D_{eq\_\max}} D_{eq}^n f_N\left(D_{eq}\right) dD_{eq} \tag{14}$$

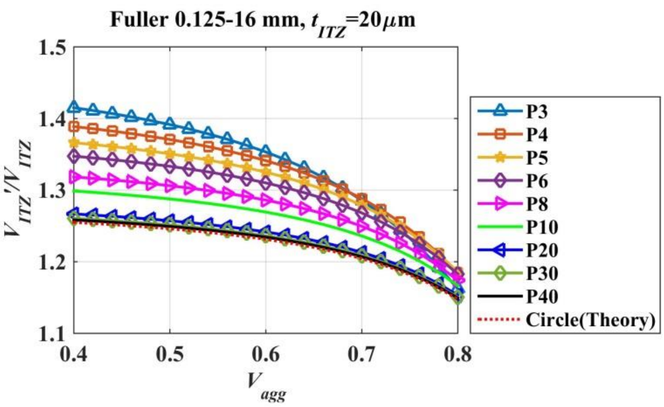


Fig. 10 Effect of polygon shape on the overestimation of ITZ volume fraction

In Fig. 10, V_{ITZ} is the actual ITZ volume fraction, while V_{ITZ}' is the numerical value of ITZ volume fraction based on the above statistical apparent ITZ thickness. It can be seen from Fig. 10 that the overestimation degree of ITZ volume fraction is decreasing with the increasing volume fraction of aggregate and the increasing circularity of regular polygon. If the volume fraction of aggregate is within 60%~80%, the actual ITZ volume fraction is potentially overestimated by 14.9%~35.3%, as shown in Fig. 10. In other words, the sectional analysis approach may significantly overestimate the effect of ITZ on macro-mechanical or transport properties of cementitious composites.

4. OVERESTIMATION OF DIFFUSIVITY

The influence of ITZ volume fraction on macro-properties of cementitious composites are investigated in the literature by using various theoretical models, such as Maxwell approximation, self-consistent approach, generalized self-consistent scheme, different effective medium method, Mori-Tanaka approach, and Hashin-Shtrikman bound, etc.(Christensen 2012, Garboczi 2001, Liu 2014, Torquato 2015, Zheng 2014). In this paper, the classical differential effective medium approximation (as shown in Eq.(15)) is employed to calculate the diffusivity of concrete D_{con} and D_{con}' based on the actual ITZ thickness and the statistical apparent ITZ thickness, respectively.

$$\left(\frac{D_{cp}}{D_{con}}\right)^{1/3}\left(\frac{D_{con}-D_e}{D_{cp}-D_e}\right)=1-V_{agg}-V_{ITZ} \qquad (15)$$

where D_{con}, D_{cp} and D_{ITZ} are the diffusivities of concrete, cement paste matrix and ITZ, respectively. $D_e=\frac{2V_{ITZ}D_{ITZ}}{3V_{agg}+2V_{ITZ}}$, V_{ITZ} and V_{agg} is the volume fraction of ITZ and aggregate, respectively.

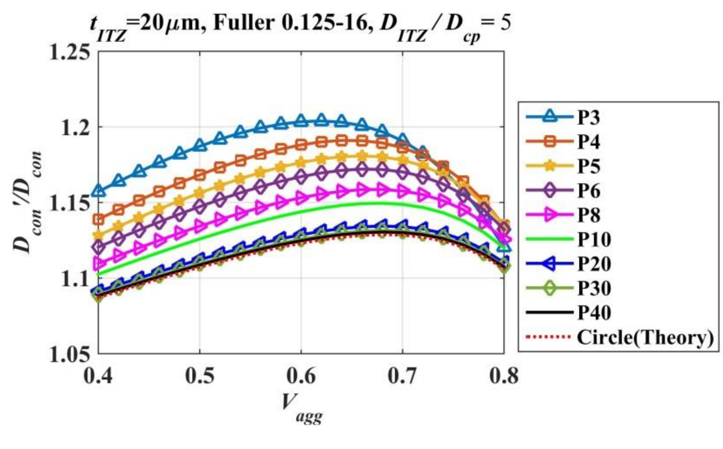


Fig. 11 Influence of polygon shape on the overestimation of concrete diffusivity

The overestimation degree of diffusivity of three-phase composites is sensitive to the volume fraction of aggregate, D_{ITZ}/D_{cp}, actual ITZ thickness and aggregate particle shape, as shown in Fig. 11. The curves of D_{con}'/D_{con} increase first and then drops down with the increasing of V_{agg}. Furthermore, the values of D_{con}'/D_{con} is decreasing with the increasing circularity of regular polygon. If the volume fraction of aggregate is within 60%~80%, the actual diffusivity of concrete is potentially overestimated by 10.6%~20.4%, as shown in Fig. 11.

5. CONCLUSIONS

This paper has proposed a methodology to construct the ITZ layer around regular polygon in 2D space. In order to quantify the influence of aggregate shape to the overestimation of ITZ thickness, SLS algorithm is used to calculate the apparent ITZ thickness and invalid coefficient. Then, the reliability of the generalized formula is verified, and the particle shape on the overestimation of the ITZ thickness is extended to multi-sized circular and regular polygon particle systems which follow Fuller distribution. Furthermore, the overestimation degree ITZ volume fraction is calculated. Finally, the overestimation degree of concrete diffusivity is evaluated based on differential effective medium theory. It is found that the overestimation of concrete diffusivity is sensitive to the volume fraction of aggregate, D_{ITZ}/D_{cp}, actual ITZ thickness and aggregate particle shape. This paper is helpful to understand the influence of ITZ volume fraction on the macro-properties of cementitious composites.

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