Particle packing: An important concept for mix design of better compacted interfacial transition zone in cementitious composite systems

Y. Gao (1), G. D. Schutter (1), G. Ye (1,2), Z. J. Tan (1)

(1) Magnel Laboratory for Concrete Research, Ghent University, Belgium
(2) Microlab, Delft University, The Netherlands

Abstract

It is well known that the interfacial transition zone (ITZ) of concrete between aggregate and bulk paste often acts as a negative effect with respect to overall properties like strength and durability. To reduce the negative influence of ITZ, it was usually considered to add various pozzolan or mineral additions into concrete, such as granulated blast furnace slag (GBFS). These additions could incur both physical packing and affect the chemical hydration reaction. In this paper, the physical packing effect in ternary blended cementitious system was studied in more detail. The maximum packing density can be calculated by employing linear packing model (LPM), based on the discretization of particle size distribution (PSD). With various mix proportions of raw materials, the overall distribution of packing density was displayed. In order to show how the initial ITZ property was influenced by packing density, numerical modeling by means of random sequential addition (RSA) has also been conducted.

1. INTRODUCTION

As well known, Portland cement based concrete has been one of the most important construction materials in present industrial realities. Vast usage of concrete has led to the occurrence of many extraordinary buildings all over the world. However, the production of raw materials making concrete could bring many resource and environment problems, for instance, CO₂ emission during production of Portland cement [1]. Fortunately, after decades of research, various kinds of pozzolanic materials as partial replacement of cement have been successfully employed in practice, such as granulated blast furnace slag [2, 3], fly ash [4], and silica fume [5]. On the other hand, with the help of superplasticizer, it became possible to design mixtures containing high volumes of pozzolans. Those were the reasons for numerous applications of blended cements in engineering projects.

Many researchers believed that a special interfacial transition zone characterized by higher porosity existed around aggregates, and it played an important role in the overall property [6].
More specifically, the bonding strength of ITZ can influence concrete strength. Furthermore, the existence of higher porosity within ITZ and increased overall pore connectivity would decrease durability of concrete when exposed to aggressive environments [7]. Therefore, many efforts have been dedicated to related research, and results showed that ITZ could be influenced by many factors, such as water binder ratio, curing age, aggregate property and so on [8-10]. Though some research work had been conducted on the influence of particle size distribution of Portland cement [11], the packing density was not often considered as a relevant parameter. Therefore, this paper is trying to make some contribution concerning this topic, starting from analytical prediction of maximum packing density of blended cement systems based on linear packing model. In order to differentiate from other hydration factors, the ITZ is studied in initial state without any hydration. Hereafter, ITZ is referring to the initial ITZ.

The paper outline is arranged like this: the linear packing model enabling an analytical prediction of packing density is introduced in section 2; calculation results are given in section 3; ITZ structure modeling is processed in section 4, including a discussion of the influence of packing density; finally, some concluding remarks are given in section 5.

2. METHODOLOGY

In this section, a brief introduction of LPM and PSD of cementitious materials including Portland cement, GBFS, and limestone filler are presented.

Due to the fact that extremely complicated interactions exist in a multisized mixture consisting of very small particles (for instance in cementitious systems: electrostatic, Van der Waals and all other cohesive or repulsive forces), a comprehensive and inclusive theory of packing of multisized particles does not exist [12]. Therefore, it would be helpful to find some approximation methods to estimate packing density. Among them, the linear packing model had received much attention for the sake of its simplicity and effectiveness [12-14]. The LPM was based on two simplified assumptions: nondeformable particles and linear interactions. The first assumption required no overlap between two particles, and the second assumption implied that complex interaction among particles can be approximated linearly. Consider a mixture composed of \( n \) sizes \( d_i \) \((i=1…n)\) of similarly shaped grains ordered from largest to smallest or vice versa, with a known independent monosized packing density for each component. Here, for the purpose of simplicity, only the final equation for calculating packing density is given while the deduction process is omitted. More details can be found in literature [12-14].

\[
\gamma = \min\left\{ \frac{\theta_i}{1-(1-\theta_i)\sum_{j=1}^{n} g(i,j)\alpha_j - \sum_{j=1}^{n} f(i,j)\alpha_j} \right\} 
\]

(1)

Where, \( \theta_i, \alpha_i \) are monosized packing density and volume fraction of \( i \)th component in mixture, respectively; functions \( g(i,j), f(i,j) \) represent the linear interaction contributed by smaller and larger particles, originating from the filling and loosening effects in particle packing; the symbol ‘\( \min \)’ means the minimum value. Compared with the container, particle sizes of cementitious material are very small, so the boundary effect of packing can be ignored, which means that the monosized packing densities are the same. Interaction functions can be measured by experiment. In current research, \( g(i,j), f(i,j) \) are following expressions as suggested in reference [14].
In order to estimate the packing density of multisized components, it is necessary to determine the particle size distribution of the system. In the present application, the cementitious composite system was composed of Portland cement, GBFS and limestone filler, and their respective densities were 3120 kg/m$^3$, 2896 kg/m$^3$, 2650 kg/m$^3$. The PSD of each composition was obtained by means of laser diffraction measurement as shown in Figure 1. Besides, a kind of fine limestone filler was employed in calculation and the PSD data can be found in reference [15].

![Figure 1: Particle size distribution of cementitious materials from laser diffraction measurement](image)

As suggested, the Rosin-Rammler function could be used to describe PSD of single cementitious composition [16]. Nevertheless, the mixture was ternary blended in current study, so it would be difficult to fit in unique Rosin-Rammler function including all components. Even if it was possible, the calculation of packing density in terms of mix proportion would still be unavailable. So, the first necessary treatment was to obtain discrete size classes of each composition and sort them in overall mixture. Equation 4 and equation 5 were employed to acquire equivalent discrete sizes $d_j$ and related volume fraction $\alpha_j$.

\[
g(i, j) = (1 - \frac{d_j}{d_i})^{2.0} + 0.4 \frac{d_j}{d_i} (1 - \frac{d_j}{d_i})^{3.7}; \quad d_j \leq d_i
\]  
\[
f(i, j) = (1 - \frac{d_j}{d_i})^{3.3} + 2.8 \frac{d_j}{d_i} (1 - \frac{d_j}{d_i})^{2.7}; \quad d_j \geq d_i
\]

Where, $d_{low}$, $d_{upp}$ are lower and upper limit relating to certain discrete size, respectively.
3. RESULT

In current study, four and eight classes for each composition were used. Specifically, the former was based on the order of general length dimension while the latter took advantage of preset characteristic sizes. Packing densities versus mix proportion of raw materials in terms of mass percentage are shown in Figures 2 to 5. It is found that packing density increases monotonously with increasing limestone filler content (see Figure 2 and Figure 3), however it was different for fine limestone filler (see Figures 4 and 5), now showing some obvious peaks. By comparison, fine limestone filler is able to increase packing density, for instance from 76% to 83% using four classes (see Figure 2 and Figure 4). Besides, the discrete classes also show an influence on the final result. With more size classes, the largest packing density among the whole domain is higher (see Figures 2 and 3, and Figures 4 and 5 for fine limestone filler). For fine limestone filler, the influence is larger, as compared between Figures 4 and 5. The overall gradient of packing density decreases while using eight classes. This indicates that it is necessary to employ more classes to characterize fine limestone filler. After all, no matter how many classes were considered, the mix proportions for obtaining the largest packing density are still the same.

![Figure 2: Packing density using four sizes](image1.png)

![Figure 3: Packing density using eight sizes](image2.png)
4. MODELING

As acknowledged by many authors, the ‘wall effect’ was the main reason to form ITZ around aggregate, and due to difficulties encountered in direct experimental investigation, numerical simulation turned out to be a good option [10, 17]. So, in order to study the influence of packing density, the numerical method by means of random sequential addition was used to model the particle packing process. Based on CEMHYD3D model [10], two rigid boundary walls were set at top and bottom surfaces imposing virtual aggregates, and the remaining faces were assumed to be periodic as shown in Figure 8 (note that the color ID was same as in Figure 6 and Figure 7, specifically, water: 0; cement: 1-5; slag: 10; CaCO$_3$: 26). The model size was 100 µm in each dimension and water binder ratio was 0.4. Details about modeling are specified in Table 1, and it should be noted that such mix proportions are only for numerical implementation.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Fine Limestone filler</th>
<th>Limestone filler</th>
<th>Portland cement</th>
<th>GBFS</th>
<th>Packing density</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mix1</td>
<td>/</td>
<td>10%</td>
<td>85%</td>
<td>5%</td>
<td>0.7760</td>
</tr>
<tr>
<td>Mix2</td>
<td>10%</td>
<td>/</td>
<td>85%</td>
<td>5%</td>
<td>0.7779</td>
</tr>
<tr>
<td>Mix3</td>
<td>60%</td>
<td>/</td>
<td>5%</td>
<td>35%</td>
<td>0.8342</td>
</tr>
</tbody>
</table>

Displaying of modeled initial state shows that closer to aggregates, small particles appear more often than larger ones, which is a direct interpretation of the wall effect (see Figure 6 and Figure 7). Comparison among the three mix proportions is shown in Figure 9. First, the ITZ width does not vary much around 10 microns for Mix1 and Mix2, which is in agreement with the medium size of packing particles. Then, for Mix1 and Mix2 with close packing density, the consequence is that their respective porosity and width of ITZ show very small discrepancy. While for Mix3, it displays obvious lower porosity and narrower transition zone. The results
suggest that there is no direct relationship between filler type and ITZ property. The packing
density however seems to be an important parameter. The most intuitive explanation is that, with
larger packing density, particles tend to pack more compact in a general way, including in the
ITZ.

Figure 6: Image of 2D slice (1 um from top
surface) from Mix2

Figure 7: Image of 2D slice (15 um from top
surface) from Mix2

Figure 8: Modeled particle packing (Mix2)

Figure 9: Porosity variation with distance

5. CONCLUSION
(1) The linear packing model is a good candidate to estimate packing density of cementitious composite systems due to its easy implementation and flexibility.

(2) Packing density varies with varying mix proportions. The discretization methods have an influence on the absolute packing density values and its distribution, however, the mix proportion achieving the largest value does not change as the number of discrete classes change.

(3) Random sequential addition method exposed to rigid boundary can simulate the wall effect very well. A direct relation between packing density and ITZ property was found from the modeling results.

6. REFERENCES


