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Optimization of particle packing by analytical and computer simulation approaches

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Abstract. Optimum packing of aggregate is an important aspect of mixture design, since porosity may be reduced and strength improved. It may also cause a reduction in paste content and is thus of economic relevance too. Several mathematic packing models have been developed in the literature for optimization of mixture design. However in this study, numerical simulation will be used as the main tool for this purpose. A basic, simple theoretical model is used for approximate assessment of mixture optimization. Calculation and simulation will start from a bimodal mixture that is based on the mono-sized packing experiences. Tri-modal and multi-sized particle packing will then be discussed to find the optimum mixture. This study will demonstrate that computer simulation is a good alternative for mixture design and optimization when appropriate particle shapes are selected. Although primarily focusing on aggregate, optimization of blends of Portland cement and mineral admixtures could basically be approached in a similar way.

Keywords: particle packing; analytical approach; optimum mixture; RSA; DEM.

1. Introduction

Mixture design in practice should not just consider the engineering requirements and economic aspects, but also the demands from construction practice, such as rheology, pumpability, finishability, *etc.* (Shilstone and Shilstone 1989). Mixture optimization is aiming to satisfy these requirements (Shilstone 1990). Considering it from engineering and economic viewpoints, concrete containing a larger amount of aggregate that is more densely packed is cheaper, has lower porosity and shrinkage, and could have improved performance. Therefore, optimized aggregate mixture design plays an important role in concrete technology. Moreover, cement blending has become a popular way to generate high performance paste for HPC or UHPC. On this level, particle packing phenomena are also of paramount importance even when inert mineral admixtures are used (Goldman and Bentur 1993). This also holds for concrete with finer fillers such as limestone powder (Ingram and Daugherty 1992, Bonavetti *et al.* 2003, Craeye *et al.* 2010), quartz flour (Lawrence *et al.* 2003, Rahhal and Talero 2005) or other inert fillers (Bentz and Conway 2001). Optimization of blended cement mixture also partly relies on proper mixture design, which is based on optimized particle packing (Bui *et al.* 2005). A number of mathematic packing models were developed in the past century such as by Furnas (1929), Aïm and Goff (1967), Toufar *et al.* (1976), Dewar (1986) and de Larrard

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(1989, 1999). Jones *et al.* (2002) evaluated some of these analytical models on the basis of experimental investigations involving different aggregate mixtures and cement blends. They found the models proposing similar sieve fractions; so, can be considered reliable in case of not too large size differences between mixed grain fractions. Reliability diminished however when the mean size ratio dropped below 0.4. Further research should be done on more fundamental models both considering particle size and particle shape (Jones *et al.* 2002).

In this study, a relatively simply theoretical method is used to predict the maximum packing density and to reveal the influence of particle size. To start with, mixtures are taken bimodal; next, this concept is extended to ternary (tri-modal) and finally to multi-sized particle mixtures. As a result of the progress achieved in computer simulation capabilities, this has basically become a liable alternative in terms of feasibility, reliability and economy (Stroeven *et al.* 2009). This paper approaches the optimization problem first of all by a relatively simple random sequential addition (RSA) model. A series of such models have been developed the past 35 years, and they can therefore be considered quite popular in engineering fields (Wittmann *et al.* 1984, Cooper, 1988, Evans *et al.* 1989, Coelho *et al.* 1997, Sherwood, 1997). Nevertheless, discrete element modeling (DEM) should be considered superior in simulating more realistic particle dispersions also at higher densities (Stroeven *et al.* 2009). The DEM system HADES that is finally applied in this paper for particle packing optimization is a so called dynamic system, which can simulate packing of arbitrarily shaped particles. Shape has been demonstrated earlier exerting significant influences on particle packing (Williams and Philipse 2003, He *et al.* 2009), so that a DEM system takes up a prominent position as to realistically simulating actual packing systems, either being aggregate or cement paste in concrete.

2. Analytical approach to particle packing

2.1 Bimodal particle packing

Two important parameters have to be assessed in bimodal particle packing:
(1) Size ratio of the particles in the two batches used in an optimized packing process.
(2) Volume proportions of the two batches of bimodal particles.

The analytical approach starts from a packed ordered structure of the larger mono-sized particles (type 1) for which obviously holds

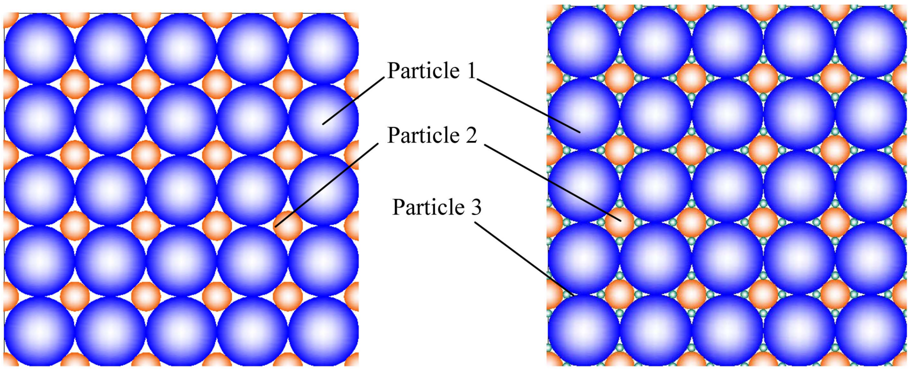
$$V_w = V_{pa1} + V_{p1} \tag{1}$$

where V_w is the total volume of the system; V_{pa1} and V_{p1} are the volume of type 1 particles and of the pores, respectively. The smaller sized particles (type 2) will be used as filler for the pores in the packed structure (see Fig. 1(a)). In the optimum mixture, the filler particle just fits into the pore.

Presumably, volume fraction (or packing density) is an invariable for packing of mono-sized particles 1, i.e. smaller sized particles 2 will only fill in the pore of initial mono-sized packed structure without disturbing the initial structure of particles 1. Therefore, it yields

$$V_{p1} = V_{pa2} + V_{p2} \tag{2}$$

where V_{pa2} and V_{p2} are, respectively, the volume of particles 2 and of the pores left by the combined system of both particle sizes, as shown in Fig. 1(a).



(a) (b)

Fig. 1 (a) Ordered bimodal and (b) tri-modal packed structure of spheres

As a consequence, the following expression is obtained

$$V_w = V_{pa1} + V_{p1} = V_{pa1} + V_{pa2} + V_{p2} \tag{3}$$

2.1.1 Size ratio of bimodal particles and maximum packing density

The final packing density should be highest in the optimized packing structure. As V_{pa1} is well-known for mono-sized random packing structures, the main problem is to obtain the highest value for V_{pa2} or the lowest for V_{p2}.

For instance, random dense mono-sized sphere packing density is approximately 0.64 (Bernal and Mason 1960), somewhat exceeding packing density in the shown ordered state. It is size invariant. Hence, the maximum packing density in bimodal packing would be 0.87, in theory.

In the described model, the small particles 2 fill the pores of the packed ordered structure of mono-sized particles 1. But in practice, where the shape of pores will be irregular, the actual packing structure will hardly reach the maximum value. Since smaller particle have a better capacity to fill in irregular-shaped pores, higher maximum packing density of the mixture can be achieved by using smaller sized particles 2.

2.1.2 Proportions in optimized bimodal packing

Another problem focuses on the proportion of two differently sized particles in the optimum packing structure. As above stated, the proportion can be expressed as

$$P = \frac{V_{pa1}}{V_{pa2}} \tag{4}$$

$V_w = V_{pa1} + V_{p1} = V_{pa1} + V_{pa2} + V_{p2} = 1$ in a normalized system. When the packing density of mono-sized particles 1 is denoted by ψ_{pa1}, Eq. (4) will yield

$$P = \frac{V_{pa1}}{V_{pa2}} = \frac{\psi_{pa1}}{\psi_{pa2} \cdot V_{p1}} = \frac{\psi_{pa1}}{\psi_{pa2} \cdot (1 - \psi_{pa1})} \tag{5}$$

where ψ_{pa2} is the packing density of particles 2 filling the pores of particles 1.

In an ideal situation (without considering influences such as wall effect, particle crystallization, local loose effect, *etc.*), $\psi_{pa1} = \psi_{pa2}$. Hence

$$P = \frac{\psi_{pa1}}{\psi_{pa2} \cdot (1 - \psi_{pa1})} = \frac{1}{1 - \psi_{pa1}} \tag{6}$$

For the case of random dense spherical particle packing, $\psi_{pa1} = 0.64$. Therefore, the optimum proportion of large particles to small particles is 2.778. In other words, the large particles should consume 73.5% of the global volume, when the packing density of the global system reaches its highest value.

2.2 Tri-modal particle packing

Based on the above stated bimodal packing, adding small particles in the mono-sized particle structure is an efficient way to improve packing density. As afore-demonstrated, it is theoretically possible to roughly predict the optimized proportions and maximum packing density. This should be verified by numerical experiments, of course. Next, packing is extended to the tri-modal mixtures. As for the bimodal packing, the main focus is on the optimized proportioning of particles fractions and on estimating maximum packing density. The solution can be visualized by the theoretical set up of Fig. 1(b).

It is assumed again that packing starts from the dense ordered packed structure of the largest fraction of mono-sized particles. Next, the median sized particles will be added into the pores of the initial structure up to maximum packing density. Finally, the smallest particles will be added to fill the pores left in the bimodal packed structure. So, the complete system can be expressed by

$$V_w = V_{pa1} + V_{p1} = V_{pa1} + V_{pa2} + V_{p2} = V_{pa1} + V_{pa2} + V_{pa3} + V_{p3} \tag{7}$$

Of course, each fraction of particles can attain the maximum packing density of mono-sized particles in the ideal situation. Therefore, maximum packing density can easily be predicted by

$$\psi_w = V_{pa1} + V_{pa2} + V_{pa3} = \psi_{pa1} + (1 - \psi_{pa1})\,\psi_{pa2} + [1 - \psi_{pa1} - (1 - \psi_{pa1})\,\psi_{pa2}]\,\psi_{pa3} \tag{8}$$

Hence, if density of randomly packed mono-sized spheres is 0.64, the maximum packing density of ternary sized spheres can attain 0.95. Similar as in the bimodal packing situation, smaller particles have a better flexibility to fill in the irregular pores between larger particles. So, this maximum packing density is only possible when there is a wider gap between neighbouring particles.

Another goal is to obtain optimized proportions for each fraction of particles. For this purpose, P_1 and P_2 define the volume proportions of large to median particles, and of the large to small particles, respectively. This leads to

$$P_1 = \frac{V_{pa1}}{V_{pa2}} = \frac{\psi_{pa1}}{\psi_{pa2} \cdot V_{p1}} = \frac{\psi_{pa1}}{\psi_{pa2} \cdot (1 - \psi_{pa1})} \tag{9}$$

$$P_2 = \frac{V_{pa1}}{V_{pa3}} = \frac{\psi_{pa1}}{\psi_{pa3} \cdot [1 - \psi_{pa1} - (1 - \psi_{pa1})\,\psi_{pa2}]} = \frac{\psi_{pa1}}{\psi_{pa3} \cdot (1 - \psi_{pa1} - \psi_{pa2} - \psi_{pa1}\,\psi_{pa2})} \tag{10}$$

In an ideal situation, the packing densities of the three particle fractions will be similar; so, $\psi_{pa1} = \psi_{pa2} = \psi_{pa3}$. Hence, Eq. (9) and Eq. (10) simplify into

$$P_1 = \frac{1}{1 - \psi_{pa1}} \tag{11}$$

$$P_2 = \frac{1}{1-2\,\psi_{pa1} - \psi_{pa1}^2} \tag{12}$$

For the case of random dense packing of spheres, $\psi_{pa1} = 0.64$, the optimized proportions of particles from large to small should therefore be 67.1%:24.2%:8.7%.

3. Computer simulation approach

3.1 RSA approach to bimodal particle packing

Particle packing is partly a geometrical problem without considering the packing process. Any geometrically possible structure of discrete particles can render a packed structure. RSA computer simulation can be employed as a simple way to realize bimodal particle packing, avoiding the mixing problem. Although packing density by traditional RSA systems is low compared with reality or with results by DEM simulation as shown elsewhere (Stroeven *et al.* 2009, He 2010), properties of bimodal packing can still be addressed by this system.

Different bimodal circular (in 2D) and spherical (in 3D) particle packing situations with different size ratios were for this purpose approached by a traditional RSA system; the respective size ratios were 1:1.33, 1:2, 1:4, and 1:8. A total of more than 1000 particles are used in the simulation to fulfill the relevant representative volume element (RVE) requirement (German 1989).

The maximum packing density of mono-sized circular (2D) and spherical (3D) particles achieved by a traditional RSA system were shown to be about 0.524 ($\Psi_{pa1} = 0.53$) and 0.31 ($\Psi_{pa1} = 0.3$),

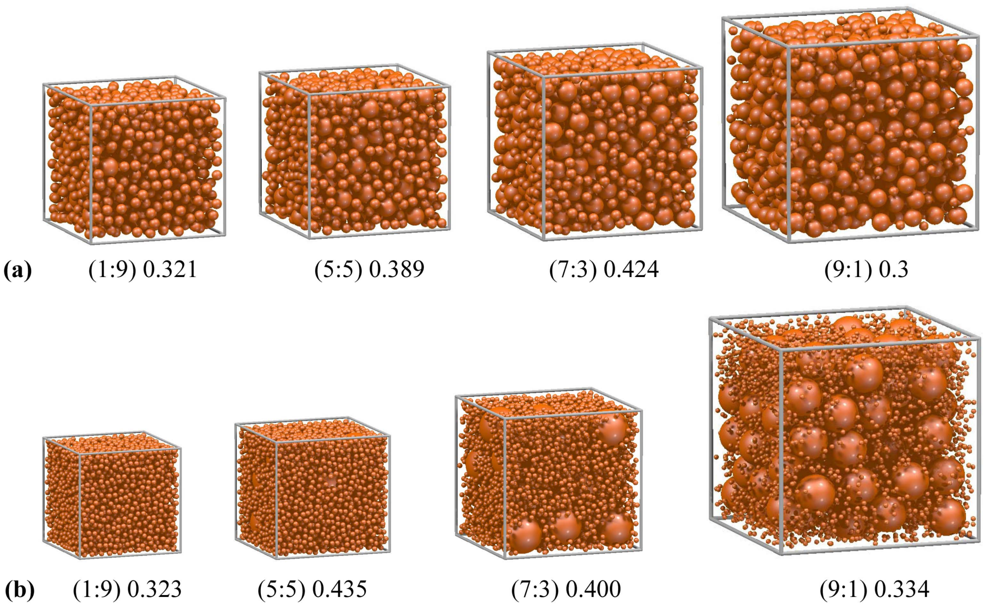


Fig. 2 3D examples obtained by RSA system of bimodal particle mixtures with different size ratios (small: large): (a) 1:2 and (b) 1:8; ratio in bracket is the volume ratio of large to small particles. Also the maximum packing density is given

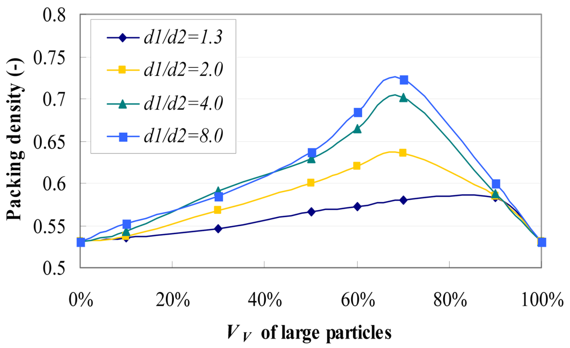


Fig. 3 Maximum packing density versus composition of bimodal mixtures for different 2D circular particle size ratios obtained by RSA packing system (d_1 and d_2: sieve sizes of large and small particles, respectively)

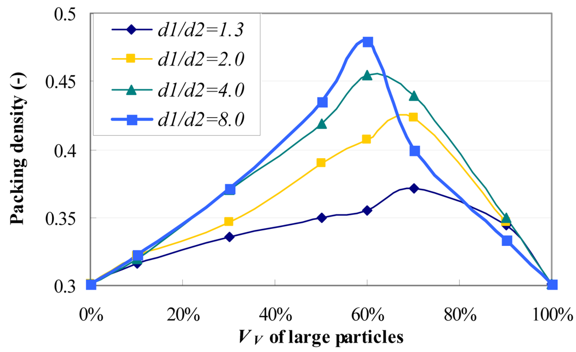


Fig. 4 Maximum packing density versus composition of bimodal mixtures for different 3D spherical particle size ratios obtained by RSA packing system (d_1 and d_2: sieve sizes of large and small particles, respectively)

respectively. Therefore, we can use the aforementioned theoretical set up to roughly predict the optimum proportions of bimodal mixtures and their maximum packing density. The large particles in 2D and in 3D should respectively consume 68% and 59% by volume of the container, assuming $\Psi_{pa1} = \Psi_{pa2}$. Maximum packing densities are 0.78 and 0.51 for mixtures in 2D and in 3D based on Eq. (3), respectively.

The maximum packing densities are determined by an RSA system in models with different volume ratios of bimodal particles (1:9, 3:7, 5:5, 6:4, 7:3, 9:1). Fig. 2 presents but a selection of the 3D ones. The corresponding bimodal particle packing results are presented in Fig. 3 and Fig. 4.

Adding small particles to a mono-sized packing of larger particles can generally improve packing density. Packing density of a mixture reaches a peak value where optimum mixture conditions are achieved. The peak of packing density is also influenced by the size ratio of particles involved. This holds both in 2D and 3D. When the size ratio of large to small particles is below 2 in 3D simulations, the optimum is obtained for 70% large particles in the mixture. The optimum amount of large spherical particles declines to about 60% when the size ratio considerably exceeds the value of 2. Also, the maximum packing density (peak value) is increased at higher size ratios. Obviously, smaller particles are more effective fillers when considerably smaller than the large particles. When compared to the theoretical prediction, we observe a better compliance for bimodal mixtures with larger particle size ratio.

Generalizing from the theoretical approach, we can see that maximum packing density is always obtained when the large particles occupy a major part of the total volume. Although small particles are more flexible in filling irregular pore size between large particles, increase in packing efficiency is slow when size is dramatically reduced. Moreover, segregation will become a problem under such conditions (de Larrard 1999).

3.2 RSA approach to ternary particle packing

Similar as in the above-described approach, the RSA method is employed for ternary particle packing. Packing simulation will start from a 2D structure, whereupon a 3D structure will be introduced. Basically, the maximum packing density of circles in 2D and spheres in 3D by a traditional RSA system are 0.524 and 0.310, respectively. Therefore, the maximum packing density and proportions of each phase in 2D or 3D can be approximated. The proportions of large particles to median and to small particles in 2D and in 3D are 58.73:27.96:13.31 and 46.17:31.85:21.98, respectively. Maximum packing densities in 2D and in 3D are 0.892 and 0.672, respectively.

A ternary dimension is used to clearly indicate the proportions of each particle phase (see Fig. 5). Each corner of the equilateral triangle represents a mixture with hundred percent of a single particle fraction (large, median or small). Proportions of the three fractions of particles are indicated inside the triangle. Using the traditional RSA system, the maximum packing density can be assessed for each combination of particles, so the triangle can be completely covered by these data.

3.2.1 RSA approach to tri-modal packing in 2D

The RSA system is employed for calculating the maximum packing density of each combination of ternary particles. The size ratio of large particle to median particle and to small particle is set to 8:4:1. The value at each node in the triangle of Fig. 5(a) reflects the maximum packing density achieved by the RSA system. Next, in Fig. 5(b) the contour lines of maximum packing density are shown. The highest packing density is reached inside the red contour line, which includes the theoretical prediction of optimized proportions (58.73:27.96:13.31).

3.2.2 RSA approach to tri-modal packing in 3D

The size ratio of the three particle fractions are selected similar as in the former case of ternary 2D packing, i.e. 8:4:1. 3000 particles are considered in the simulation. Different combination of the three particle fractions are simulated by RSA to find maximum packing density. Quantitative values of maximum packing densities are shown in Fig. 6(a). Fig. 6(b) presents the contour lines for maximum packing density, indicating inside the red contour line the optimized proportion of ternary sized particles leading to the highest densities; this also includes the theoretical solution (46.17:31.85:21.98).

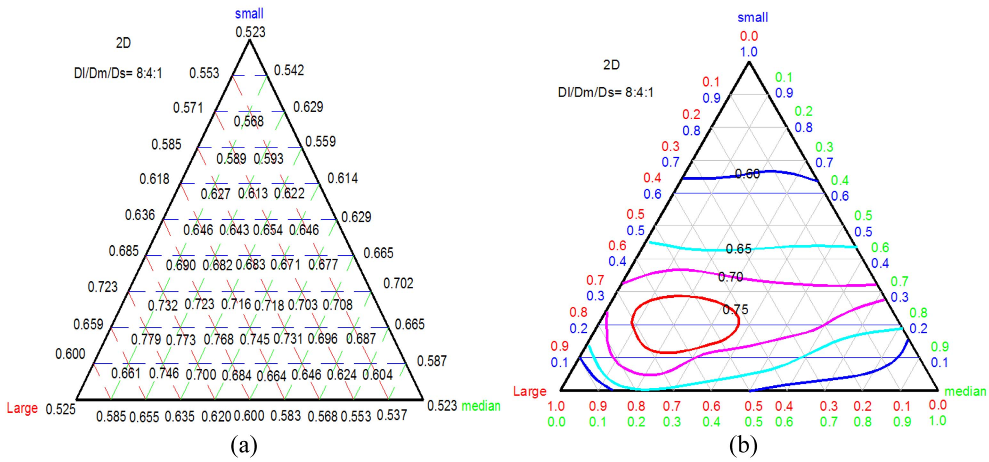


Fig. 5(a) Maximum packing density distribution and (b) the corresponding contour of ternary 2D particle mixtures

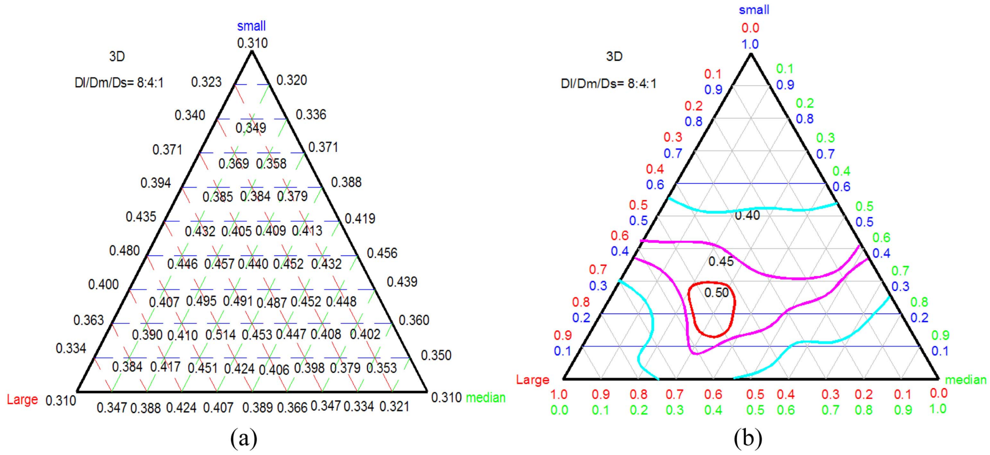


Fig. 6(a) Maximum packing density distribution and (b) the corresponding contour lines of maximum packing density of ternary 3D particle mixtures

3.3 DEM approach to particle packing

Particle packing by a DEM system more realistically simulates the actual situation in practice. The same sequence of operations as accomplished by RSA will be followed.

The 2D bi-modal models are shown in Fig. 7 for different mixture proportions. The size ratio is 2. Loose packing and dense packing with compaction are both used for each mixture. Final packing

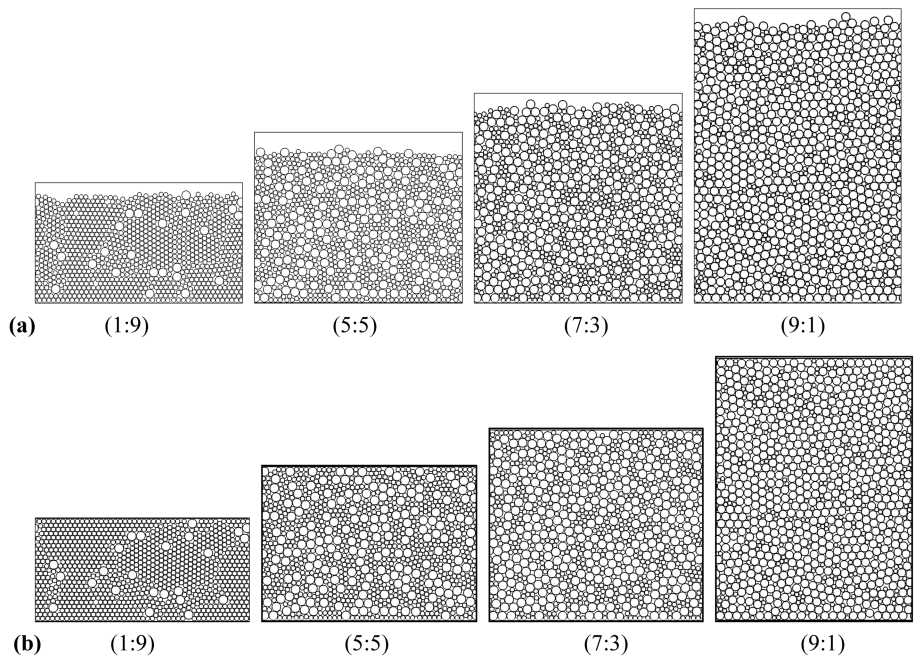


Fig. 7 Display of 2D models of bimodal particle mixtures obtained by a DEM system (size ratios is 2): (a) loose packing; (b) dense packing due to compaction

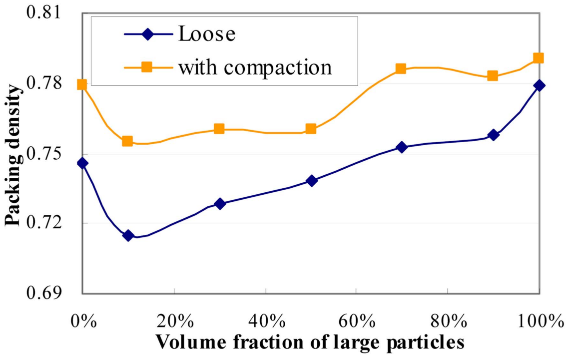


Fig. 8 Packing density obtained by DEM in loose and compacted state of the different bimodal mixtures displayed in Fig. 7

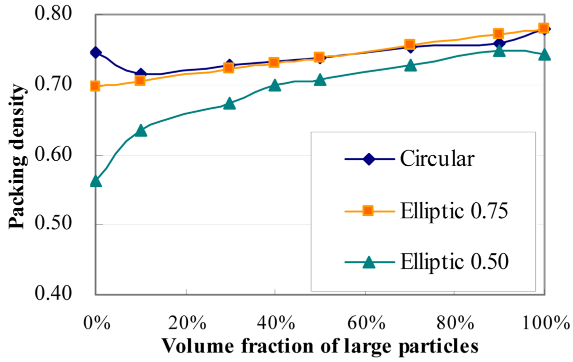


Fig. 9 Bimodal packing of ellipses with different elongation (aspect ratio, size ratio is 2)

densities are plotted in Fig. 8 for the various mixture proportions and each packing method. Besides the mono-sized particle packing, the packing density is initially enhanced with increased usage of large particles. A peak value was roughly obtained when the mixture contained about 70% of large particles (especially in models with compaction). The theoretical estimate amounts to 80%; further a maximum packing density of 0.94 is predicted. Dense packing is largely influenced by so called crystallization (ordering), as can be seen in Fig. 7. Introducing a sufficient quantity of small particles will disturb this ordering in structure formation and thus will cause the packing density to decline, as demonstrated in Fig. 8. It is expected that this phenomenon is less significant in 3D simulation. Crystallization will also be suppressed in DEM-generated irregularly-shaped particle packing, as demonstrated in Fig. 9 where ellipses with different aspect ratio have been simulated in the loose random packing state; at higher elongation or aspect ratio, the "crystal" phenomenon is less significant and effects of the filler are more obvious.

4. Multimodal particle packing

4.1 Size distribution

Basically, adding smaller particles can increase the packing density of a whole system, which has been illustrated by bimodal packing and tri-modal packing. A theoretical framework for multimodal packing can be developed analogously as for bi-and tri-modal packing. So

$$V_w = V_{pa1} + V_{p1} = V_{pa1} + V_{pa2} \dots V_{pan} + V_{pn} \tag{13}$$

in which V_{pan} is the volume of nth particles and V_{pn} is the volume of pores left by the nth particles. Each size fraction can presumably arrive at a similar maximum density under "ideal" conditions. This renders the possibility of predicting total packing density of a whole system. The only variable is the packing density of the mono-sized fraction of largest particles. The optimized proportions of each size fraction can also be roughly predicted by

$$P_n = \frac{V_{pa1}}{V_{pan}} = \frac{\psi_{pa1}}{\psi_{pan} \cdot \{1 - \psi_{pa1} - \ldots - ([1 - \ldots]\,\psi_{pa(n-2)})\,\psi_{pa(n-1)}\}} \tag{14}$$

in which P_n is the volume ratio of the largest particles to that of the n^{th} size fraction of particles. The adoption of the aforementioned "ideal" conditions leads to further simplification of Eq. (14). It can also be expected that optimum proportion ratio of two sized particles are maintained, which also has been illustrated by bimodal and ternary estimates.

Of course, the actual packing state will be influenced by many factors in reality, e.g. shape of particles, packing methods, physical interaction between particles, wall effects, *etc*. But basically, mono-sized packing should be treated as the most important one, from which multi-modal mixing can be developed. The influence of particle shape on packing is also revealed in mono-sized particle packing. When particle packing is only emphasized as a geometric problem, the influence of shape on multi-sized particle packing can be considered by incorporating information of mono-sized particle packing. For instance, eigen packing density of each constituent should be calibrated and used to calculate the combination of each size class in de Larrard's models (LPM or CPM model) (de Larrard 1999). Loose effects and wall effects are also analytically considered, which constitutes also the link to the rheology of concrete in those models (de Larrard 1999).

One popular way of characterizing particle size fractions in concrete is Fuller's curve developed by Fuller and Thomson (1907). This curve is given by

$$P(D) = \left(\frac{D}{D_{\max}}\right)^q \tag{15}$$

in which $P(D)$ is the cumulative volume percentage of particles passing sieve size D; D_{max} is the maximum grain size in the mixture; q is an exponent, originally specified as 0.5.

Different values of q ranging from 0.3 to 0.5 were later proposed by other researchers. It has been proven that $q = 0.45$ leads to optimized mixtures for asphalt concrete (Roberts *et al*. 1996). Andreasen and Andreasen (1930) suggested $q = 0.37$ for optimized packing. The curve with $q = 0.30$ is close to the grading curve used by Su *et al.* (2001) for self-compacting concrete (SCC) (Brouwers and Radix, 2005). Curves with different values of q are plotted in Fig. 10(a). It shows the curve to shift upwards for lower q-values; this implies that larger quantities of fine particles are involved. The finer particles are relatively scarce by the traditional Fuller curve. Optimum grading in SCC

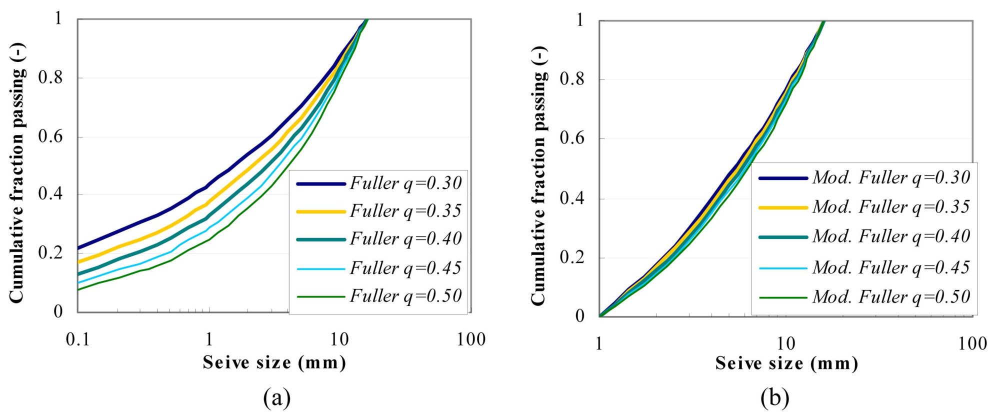
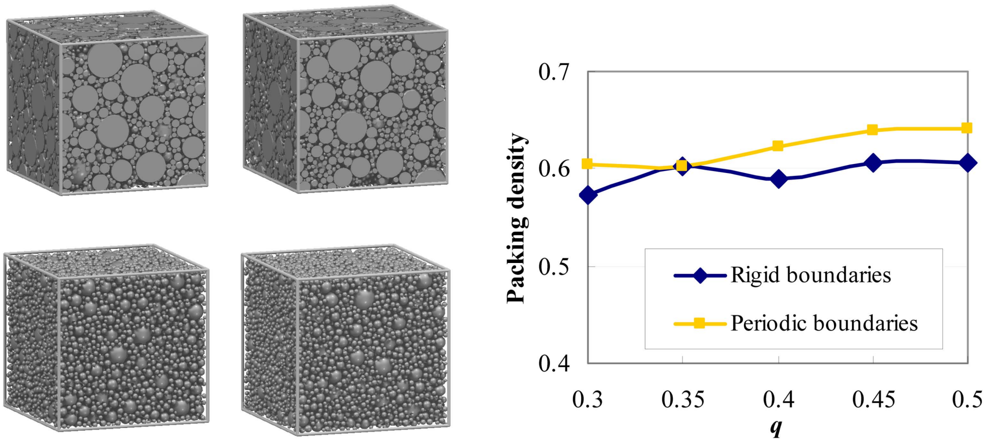


Fig. 10(a) Fuller and (b) modified Fuller particle size distribution curves with different power numbers, q



(a) (b)

Fig. 11(a) Displayed models of RSA-packed particle structures (left column: $q = 0.5$, right column: $q = 0.3$) and (b) packing densities for different values of q in modified Fuller curves

should involve more fine particles compared with traditional Fuller-based concrete.

The Fuller curve describes the whole range of particle sizes downscaled to micro-level. But for practical use or for simulation, the curve can be restricted to a limited range of particle sizes upward from a minimum aggregate size (Brouwers and Radix, 2005)

$$P(D) = \frac{D^q - D_{\min}^q}{D_{\max}^q - D_{\min}^q} \tag{15}$$

where D_{min} is minimum particles size.

Some curves with different value of q and 1 mm for minimum particle size are presented in Fig. 10(b). Differences in the curves are smaller than in Fig. 10(a) for standard Fuller curves.

4.2 Structure simulation

Next, simulations are conducted with the RSA method to check the influence of the PSD governed by the modified Fuller curves on packing density. An example of structural models and the packing density distributions are shown in Fig. 11. The influence of q is found insignificant. Decreasing the power number, packing density is slightly diminished at lower q values due to the larger amount of fine particles involved in the mixture, especially in models with periodic boundaries (representing bulk features). The results still show that modified Fuller particle size distributions lead to optimum situations for $q = 0.5 \sim 0.45$.

5. Conclusions

Influences of particle size on particle packing are studied as the main objective in this paper. A simple theoretical concept is employed for assessment of the influence of particle size on packing and for prediction of the optimum mixture. The numerical approach by the RSA system is then used for packing simulation. The analytical as well as the simulation approach go from bimodal packing to ternary packing.

(1) The results obtained by the RSA system comply well with the theoretical estimates. Although many other parameters are not taken into consideration such as particle shape, boundary conditions or interactions between particles, the analytical approach can still predict the mixture properties reasonably well. Introducing finer particles in mono-sized particle structures is an effective way to improve particle packing density.

(2) Simulations by the DEM approach in 2D space reveal that effects due to the filler can be overtaken by the crystal phenomenon. In other words, particle interaction can be the dominant factor in achieving high packing density in some cases. Basically, an RSA system cannot reproduce this phenomenon. In packed systems of irregular shaped particles this crystallization phenomenon is suppressed and the filler effect becomes more significant.

(3) In multi-sized particle packing, optimum mixture can be calculated based on each mono-sized particle packing as in de Larrard's models (de Larrard 1999). The influence of particle shape can also be derived from mono-sized packing information. The simulation results show that packing density of particles based on modified Fuller curves reaches to a peak value when the power number q is 0.5 or 0.45.

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