COMPUTER SIMULATION OF THE PACKING OF DIGITIZED REACTIVE MAGNESIA PARTICLES

Mingzhi Wang (1), Abir Al-Tabbaa (2)

(1) Department of Engineering, University of Cambridge, UK
(2) Department of Engineering, University of Cambridge, UK

Abstract

Reactive magnesia cement can gain significant strength through its hydration-carbonation processes, providing a novel and green construction material. Various factors influence the hydration and subsequent carbonation of reactive magnesia within a porous structure, including interaction between the solid, gas and liquid phases, making it more difficult to model than the traditional Portland cement system. The simulation of particle packing can generate realistic microscopic modelling of the porous structure of cement particles, which can be used in further simulation of the hydration-carbonation processes. In this paper, Cellular Automata (CA) technique was applied to construct the initial structure generating program as the first stage of further simulation of reactive magnesia cement. This digitized particle packing simulation program provides well-matched results to the previous experiment and simulation results from the science of particle packing compared with previous models using the method random sequential addition (RSA). Additionally, some special features of the packing of digitized particles were observed through different simulations, providing impetus for further study.

1. INTRODUCTION

Particle packing is a complex mechanical behaviour of numerous particles being mixed with each other, with or without vibration. The study of particle packing involves physics, engineering and material science. For cement science, it has been found that packing can be used to explain the formation of initial fresh cement microstructure, the glass forming ability of alloys [1] and the origin of interfacial transition zone (ITZ) between cement paste and aggregates [2]. Mathematically, it was proven that 0.74 is the highest average density can be achieved by the packing of mono-sized sphere particles [3]. In practice, it was found that the upper and lower limits for mono-sized sphere particle random packing are about 0.64 [1] and 0.44[4, 5], respectively. On the other hand, it was found that the packing fraction for frictionless spheres obeying log-normal distribution is a function of the standard deviation [1,6]. Interestingly, recent experiments [2, 7, 8] have shown that the packing fraction of fresh cement particles was observed to be close to the value of random close packing (RCP).

Bentz[9] developed a 3D cement hydration model using cellular automata algorithm. Digitized particles composed of voxels were used in Bentz’ model to represent cement
particles and a program was developed to simulate the flocculation process of fresh cement particles. However, the initial packing fraction obtained from Bentz’ model was found to be close to the random sequential addition (RSA) value of 0.385 [10], which was lower than the experimentally measured packing fraction value of fresh cement particle. Jia & Williams [11] developed an packing algorithm for 2D digitized particles, which was further developed and optimised into 3D by Byholm et al.[12], separately. However their focus was on the packing of particles with arbitrary shapes. de Korte & Brouwers [10] studied the RCP behaviour of mono-size digitized sphere particles used a modified version of Lubachevshky and Stillinger algorithm[13]. In this paper, a numerical attempt is made inspired by the paper of Jia & Williams [11] to simulate the behaviour the RCP and random loose packing (RLP) behaviour of 3D digitized sphere with a mono-size and log-normal distributed size.

2. METHODOLOGY

The basic algorithm of this work is still cellular automaton (CA). By definition, CA describes the packing system with discrete time and space, where the movement routine of each particle at current iteration is calculated based on the state of its surrounding sites. As a result, discrete particles of different sizes should be described before the application of CA as shown in Figure 1. The properties of digitized particles used in this study are similar to the original properties in Bentz’ [9] model. However the modified particles with radius of 3, 5 and 10 in de Korte & Brouwers’ [10] study are applied here.

![Figure 1: Digitized particles with different sizes](image)

The amount of particles with each size is calculated using the cumulative log-normal distribution function given by

$$D(r) = \frac{1}{2} \left[ 1 + \text{erf} \left( \frac{\ln r - \ln r_0}{\sigma \sqrt{2}} \right) \right] \quad r = 1, 2, 3, 4, ...$$  \hspace{1cm} (1)

where $\ln r_0$ and $\sigma$ are the mean and the standard deviation of $\ln r$. Due to the nature of discreet sphere, the fraction of particles with the smallest size is directly calculated by applying $r=1$. Then the amount fraction of the particles with the rest sizes is calculated from the smallest ones to the biggest ones by the equation

$$F(r) = D(r) - D(r - 1) \quad r = 2, 3, 4, 5, ...$$  \hspace{1cm} (2)

Then the amount of particle with each size is calculated with a pre-defined total particle amount. It needs to be mentioned that the particle amount of each size is still discreetly distributed. Figure 2 illustrates a comparison between the cumulative log-normal distribution function and the discrete particle size distribution in this work. When the standard deviation is set to be small enough, the particles become mono-sized.
Afterwards, the initial positions of these particles are assigned according to the approach of Bentz’ [9] and Jia & Williams [11]. To be more specific, each particle is placed in a cubic domain one by one from the biggest to the smallest. The position of one placement is randomized, although overlapping with already placed particles is not permitted. The difference between the approach in this study and Bentz’ [9] model is that periodic boundary conditions are not applied on the top and bottom boundaries to simulate the effect of gravity here. Figure 3 illustrates the initial position of 300 particles generated in a 150×150×200 domain following the above approach.

One important step involved both in the generation of initial particle position and the simulation of particle movements during packing is overlapping check or contact check. In a discrete space, the position data of digitized particles can be seen as a three dimensional matrix, making the overlapping check of two particles becomes the analysis of intersect of their matrix. If the intersect is a zero set, overlap does not exist between these two particles. On the other hand, overlap is detected if the intersect of the two particle matrix is not a zero set. With similar approach, surface contact check of two particles can be achieved by expanding the particle matrix with its surface voxels. Figure 4 illustrates the overlap of two digitized particles in 2D.

Then the CA simulation of particle packing is conducted. To be more specific, 26 directions of one single particle are firstly checked to see whether the particle is contacted. During the contact check, the amount of contacted surface voxels in 26 directions is recorded separately. One of the 26 directions is regarded to be possible move the particle into if the contact number in this direction is 0. Then one of the possible movement directions is chosen based on specified rules, which can be defined and verified, accordingly. In this work, factors involved in the choosing rules are normal contact force simulated statistically, friction force and gravity.

The relationship between Hertz repulsive force and contact properties are shown with the following equations [14]:

\[
\alpha = \left( \frac{3PR}{4E^*} \right)^{1/3}
\]
\[ \alpha_{ij} = \frac{\alpha^2_R}{R} = \left( \frac{9p^2}{16RE^2} \right)^{1/3} \]  

Figure 4: Contact check for digitized particles

where \( a \) is the contact area radius; \( E^* \) is a constant relevant to the elastic modulus of the contacted material; \( 1/R \) is equivalent contact curvature; \( \alpha_{ij} \) is the relative displacement of the centres of two contacted spheres, as shown in Figure 5. With equation (3) and (4), the following equations are derived:

\[ P = \frac{4}{3} E^* \sqrt{R} \alpha_{ij}^3 \]  

(5)

\[ P = \frac{4}{3} E^* \frac{a^3}{\sqrt{R}} \]  

(6)

These equations indicate that the normal force appears to be a function of equivalent contact curvature, radius of contact area and relative displacement:

\[ P = P_f(R, a, \alpha_{ij}) \]  

(7)

Assuming that \( R \) is equal to the radius of currently being analysed particle, the above function can be converted to the following function:

\[ P = P_f(V, A, \alpha_{ij}) \]  

(8)

where \( V \) is the volume of currently being analysed particle, \( A \) is the area value of the contact area. Since \( \alpha_{ij} \) is the relative displacement of the contacted particles during unit computation iteration (time), it is treated as the relative speed in the contact direction. In this work, this value is randomly assigned with the Maxwell-Boltzmann distribution function:

\[ f(\alpha_{ij}) = \frac{2 \alpha_{ij}^2 e^{-\alpha_{ij}^2/2\sigma^2}}{\sqrt{\pi} \sigma^3} \]  

(9)

where \( \sigma \) is the only control parameter.

It was found that the following function can provide a relatively satisfying final packing fraction for mono-size particle packing:

\[ P = V \sqrt{A} \alpha_{ij}(\sigma) \]  

(10)

where \( \sigma \) is taken as 1.1.

With the normal force, the friction threshold in the tangential direction is then calculated:
where \( \mu \) is equivalent to the coefficient of friction, which can be used to adjust the final packing fraction.

Gravity proportional to the particle volume is applied on all particles in vertical directions. The movement in one unblocked direction is possible if the normal force in opposite direction is bigger than the friction threshold. When more than one direction is available for movement, certain priority rules are applied. In this work, one of the available movement directions is randomly chosen with uniform distribution function. For computation efficiency, only the forces in 6 principal directions are simulated as shown in Figure 6. The analysis of the rest 20 directions is considered as a combination of the 6 principal direction simulations. Figure 7 illustrates the packing outcome for the example of the 300 particles shown in Figure 3.

![Figure 6: Six principal directions](image)

![Figure 7: Particles after packing](image)

3. RESULTS AND DISCUSSION

The above method was used to conduct the simulation of the packing of mono-sized and log-normal distributed digitized particles. A total particle number of 3000 was applied for each test. Although the simulation was randomized, it was found that the simulation result is stable with 3000 particles. Additionally, a size ratio between the length of the cubic domain and perimeter of particle \((L_{box}/d)\) of 15 was maintained for all the simulation with mono-sized particles.

3.1 Random packing for mono-size particles

Figure 8 shows the mean packing fraction results of mono-size particle packing simulation with different particle radius and coefficient of friction (cof). It can be clearly observed that the mean packing fraction of the final porous structure decreased when coefficient of friction is increased. With the provided range for the coefficient of friction from 0 to 1.00, the mean fraction ranges from 0.52 to 0.69. This range agrees well with the commonly accepted fraction value of 0.64 for random close packing (RCP) [1, 6, 10, 13, 15], and 0.55 for random lose packing (RLP) [4, 16, 17]. The influence of friction on the mean packing fraction also agrees well with the experimental observation that the packing fraction of sphere appears to be function of coefficient of friction [18].
For packing simulation with a low coefficient of friction or RCP, it can be seen that there is a trend for the packing fraction to approach 0.64 when the particle radius is increased. The factor causing the difference in the fraction result of particles with different radius under random close packing situation is the actual geometry of the digitized sphere. A similar influence of particle size was also observed by de Korte & Brouwers [10], who explained it as a result of particle roundness and sphericity. As shown by Figure 8, digitized particles with big radius tends to appear much more spheres-like than particles with small radius. Comparing with the roundness data of each particle size in de Korte & Brouwers [10], it can be noticed that when the roundness of digitized particles is more close to 1, its RCP behaviour is more sphere-like. On the other hand, a very small roundness will lead to a high packing fraction. As a result, the RCP value of 0.64 for sphere packing is approached when the radius of digitized particle is increased.

For packing simulation with high coefficient of friction or RLP, it was observed that the influence of particle size is not as significant as what was observed in RCP simulation. On the contrary, the mean packing fraction results from simulation with high coefficient of friction is relatively stable for particles with different radius. According to [6], the mean coordination number from RCP is less than what was obtained from RLP, indicating that particles are less contacted in RLP situation. Figure 9 illustrates the change in coordination number distribution resulted from different simulation on particles with radius of 11. It can be seen that, the mean coordination decreases with increasing coefficient of friction, which means particles are less connected in RLP simulation than in RCP situation. Since less contact area is needed to form a stable structure under RLP situation, the influence of actual particle shape on final packing result is not as significant as what it is in RCP simulation.

3.2 Random packing of particles obeying log-normal distribution

Figure 10 shows the packing simulation result of particles obeying log-normal distribution with a mean radius of 7. After the standard deviation reaching 0.1, the packing fraction behaved as a function of the standard deviation of particle radius, which agrees well with previous studies [1, 6, 19]. Similar to the mono-size particle packing simulation, an increase in coefficient of friction leads to a looser porous structure and low packing fraction.
In He’s [6] simulation, the packing density was not changed significantly when the standard deviation was below 0.1. However in this study, a significant drop in packing fraction was observed before this critical point. The reason for this unnatural drop in packing fraction is probably the difference of packing of mono-size discrete particles and polydisperse distributed discrete particles and the standard deviation threshold between is 0.1. However further study is still required to reveal the nature of digitized particle packing. Additionally, the final structure of the log-normal distributed particles at high standard deviation from this work appears to be looser than the result from continuous model done by He et al. [5], indicated by a lower packing fraction. Figure 11 illustrates the corresponding mean coordination number results, the decreasing trend of mean coordination number with increased standard deviation can be clearly observed, which agrees very well with the continuous model.

4. CONCLUSIONS

A cellular automaton based model was developed for discrete particle packing and the packing of both mono-size and log-normal distributed particles was simulated. The effect of the coefficient of friction was included in the model to distinguish between random close packing and random loose packing. With mono-size particles, the results of packing fraction and coordination number agree well with previous study. With log-normal distributed particles, it was observed that the packing fraction behaved as a function of the standard deviation of particles radius, when the standard deviation is above 0.1. Meanwhile, the coordination number results from polydisperse particles are also in good agreement with previous research.

As a discrete model, the relationship between the final porous structure and the geometry properties of particles is not as smoothly developed as that in continuous model. However, very few differences in changing the were observed, indicating that the influence of packing factors such as particle size distribution, and friction can still be simulated to some extend in the discrete model. One significant difference between this discrete model and previous continuous models is the packing behaviour of polydisperse particles with low standard
deviation. When the standard deviation reached 0.1, the behaviour of the packing in the discrete simulation program was switched from mono-size-liked to polydisperse-liked. Although the packing of discrete particles is not completely the same as continuous particles, the more realistic final porous structure generated can still be used in further cement simulations.

REFERENCES