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Random packing of digitized particles

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ABSTRACT

The random packing of regularly and irregularly shaped particles has been studied extensively. Within this paper, packing is studied from the perspective of digitized particles. These digitized particles are developed for and used in cellular automata systems, which are employed for the simple mathematical idealizations of complex systems in physics, chemistry and engineering [S. Wolfram, *Rev. Mod. Phys.* 55, 601–644 (1983)]. In the present paper, the random packing of digitized particles is studied using the packing routines available in the cellular automata cement hydration model by D.P. Bentz [*A Three-dimensional Cement Hydration and Microstructure Program. I. Hydration Rate, Heat of Hydration, and Chemical Shrinkage* (National Institute of Standards and Technology, 1995)] and a modified version of the Lubachevsky and Stillinger algorithm [B. D. Lubachevsky and F. H. Stillinger, *Journal of Statistical Physics* 60, 561–583 (1990)]. It is shown that the packing of digitized particles is comparable to spheres, when taking into account the specific properties of digitized particles.

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1. Introduction

Packing of disks and spheres, both uniform and graded sizes, was described in great detail in literature [1–4]. Besides these ideal disks and spheres also spheroids and irregular particles are studied [5–8]. In this paper, the packing of digitized particles is studied. The digitized particles are developed for and used in cellular automata systems, which are employed for the simple mathematical idealizations of complex systems in physics, chemistry and engineering [9]. ‘Cellular automata consist of lattice of discrete identical sites, each site taking on a finite set of integer values. The values of the sites evolve in discrete time steps according to deterministic rules that specify the value of each site in terms of the values of neighbouring sites’ [10]. According to Kier [11], cellular automata are not restricted to only the use of deterministic rules, since probabilistic rules are used extensively for studying real physical and chemical systems nowadays. The digitized particles are representing spherical particles in ‘cubic’ cellular automata system and are composed of voxels (cubic pixels) (Fig. 1). The properties of the digitized particles are presented in Table 1. During the packing experiments these particles are placed in a confined geometry, here a cubic box with rib size L_{box} . Both the size of particle (d) and box (L_{box}) are expressed in the basic voxel size.

Jia and coworkers [6,12,13] have also performed simulation using digitized particles. Their research mainly focused on packing of particles

of arbitrary shapes mostly in 2D, while the current research focus on regularly shaped particles in three dimensions. Furthermore in the current research, the influence of shape on the state of random close packing (RCP) is studied thoroughly.

2. Methods

Bentz [14] investigated the hydration of cement using a cellular automata model and therefore generated an initial microstructure by placing digitized particles in the confined system (3D) box at random locations from largest to smallest particle size, employing periodic boundaries [15]. This model has been developed for the creation of the initial microstructure using graded particles rather than monosized particles. Using this module for uniform particle sizes, the authors obtained a packing fraction up to 0.379 for $d=13$ and $L_{\text{box}}=100$. This is close to RSA (random sequential addition) of 0.385 as found by Williams and Philipse [16]. However, this value is far below the commonly accepted random close packing (RCP) value of 0.634 [17].

Since Bentz’ model was developed to create a starting microstructure for cement hydration, and not aimed to acquire close packings of monosized particles, a ‘new’ packing routine has been developed. The used routine is comparable to the sequential methods as described by Lubachevsky and Stillinger [2], who obtained a packing fraction of 0.634 for monosized spheres.

Within the applied routine, particles are placed randomly in a box, after placing a new particle all other particles are moved. The movement of the particles and speed of addition of particles is described by two parameters. The first parameter governs the number of

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Fig. 1. Digitized particles with a sizes of 3, 5, 7, 11 and 21 voxels (the top-surfaces in gray, which are used for the computation of the digitized roundness) (the voxel that are removed from the 7 and 11 and 21 particles during shape modification are indicated in black in print and blue online).

moves (over a distance of 1 voxel in x, y or z direction) that each particle attempts to make during a movement cycle (n_m). The second parameter is the number of movement cycles (n_t). Furthermore different boundary conditions can be applied, which each has a certain limitation on moves over the boundaries of the box. These boundary conditions range from periodic boundary conditions on all sides to the limitation of particle movement across the boundaries of the box (here the faces of the cube). During the present research, a combination of boundaries types was applied. The movement through the top and bottom surface of the box was limited, whilst applying periodic boundaries on the other sides. The movement in this direction was limited, but however not impossible. Actually, all boundaries are periodic, but only the movement in this one direction has been restricted in order to have convergence during application of (modified) Lubachevsky and Stillinger routine. This proved to result in the best packing results, which were slightly higher for lower size ratios l (box size divided by the particle diameter, i.e. L_{box}/d) compared to a system with limitations on all sides. In other words, true periodic boundaries have been applied. Furthermore, preliminary studies of the packing systems showed that the n_t had no obvious influence on the packing fraction, and this packing fraction is

influenced by the n_m up to value of 20. By a further increase of n_m , the packing fraction levels out, while the computational time keeps increasing. Jia and Williams [6] showed a similar effect on packing fraction by the speed of addition of particles in their models.

3. Results

Fig. 2 shows the results of these simulations, particles with $d = 7, 9, \dots, 37$ and 39 were used, and boxes with $L_{\text{box}} = 50, 60, 75, 85, 100, 200, 300, 400$ and 500. One can notice that there is a clear relation between size ratio l (L_{box}/d) and the packing fraction $f(l)$, and a similar trend can be observed as by Desmond and Weeks [18]. One can see that $f(l)$ tends to a limiting value for the higher size ratios l , higher than the RCP value for spheres (0.64), particularly for $d = 7, d = 11$ and $d = 21$ particles. This can be partly explained by the particle shape, which will be done later in this paper. Jia and Williams [6] found a similar result during their research when packing identical circles in their 2D-system DigiPac. They found a value of 0.875, which is in between the value for random (0.82) and crystals (0.91), which according to these authors indicates that the generated structure contains crystal domains. During preliminary tests in 2D during our research we have found a comparable value of 0.877 for $L_{\text{box}} = 350$ and $d = 11$. Caulkin et al. [19] investigated packing of mono-size spheres, binary and ternary mixed beds both experimental and using DigiPac using cylindrical containers and found a maximum packing fraction of 0.599 for mono-sized spheres. The main difference with this research is the absence of periodic boundaries and the shape of the container.

Furthermore from Fig. 2, one can observe a raise in the packing fractions close to $l = 1$, which is caused by the finite size effect as

Table 1

Properties of digitized particles (with * as the modified particles), the particle volume is expressed in single voxel volumes and the surface in the surface area of one voxel face, and $F = d \cdot A_p/V_p$.

Size d	Digitized particle			Sphericity ψ Eq. (1)	Roundness ε Eq. (2)
	Volume V_p	Surface A_p	Shape factor F		
1	1	6	6.00	0.716	0.00
3	19	54	8.53	0.760	0.44
5	81	126	7.78	0.819	0.57
7	179	222	8.68	0.807	0.76
9	389	414	9.58	0.876	0.81
11	739	582	8.66	0.901	0.78
13	1189	822	8.99	0.905	0.85
15	1791	1062	8.89	0.909	0.88
17	2553	1350	8.99	0.921	0.89
19	3695	1758	9.04	0.928	0.90
21	4945	2094	8.89	0.931	0.89
23	6403	2526	9.07	0.938	0.91
25	8217	2934	8.93	0.942	0.92
27	10,395	3462	8.99	0.946	0.92
29	12,893	3990	8.97	0.949	0.93
31	15,515	4494	8.98	0.951	0.94
33	18,853	5166	9.04	0.952	0.94
35	22,575	5838	9.05	0.956	0.94
37	26,745	6510	9.01	0.958	0.94
39	31,103	7206	9.04	0.963	0.95
7*	171	222	9.09	0.841	0.76
11*	691	582	9.26	0.881	0.87
21*	4801	2094	9.16	0.930	0.92

Fig. 2. The relation between size ratio l (L_{box}/d) and random packing fraction ($n_t = 10, n_m = 20$).

described in Desmond and Weeks [18] and also observed experimentally by McGeary [20].

The data presented in Fig. 2 is obtained using cubic boxes with equal dimensions in the three directions. Fig. 3 shows the effect of changing the height of the box for different dimensions of a square bottom and $d = 13$. Fig. 3 shows the data scaled to the maximum packing fraction obtained for a particular bottom size ratio l (L_{box}/d). As one can notice that the height of the box (H_{box}) has an influence on the packing density. Furthermore a raise in the packing fraction can be observed close to $h = 1$ comparable to the effect at $l = 1$ and which can be explained by the work of Desmond and Weeks [18] as well as McGeary [20].

Besides the packing fraction also other parameters are available to characterize random particle assemblies, such as the contact number and translational order metric. Fig. 4 shows the average contact number for several of the performed 3D-simulations. The contact number is linearly related with the packing fraction (Fig. 4). This linear trend is almost identical to the linear trend found by Aste [21], who used a cylindrical container for his simulations, while in this paper a cubic container is used. For some simulations with high packing fraction, the contact number is higher than the caging number, which could indicate some ordering in the system. Furthermore one can notice contact numbers higher than 12, which is higher than one should expect from theory. These extreme values are all caused by particles with $d = 7$, most probably due to their particle shape.

As already noticed in Fig. 2, all particle sizes tend to a random packing fraction larger than 0.64, the RCP value of spheres. The effect was most pronounced for $d = 11$ and $d = 21$. One possible explanation is the angularity of the digitized particle. There are several ways to describe the effect of particle shape on packing [22]. Barrett [23] distinguished three parameters to describe the shape of a particle based on literature review of numerical methods to describe these parameters namely; shape, roundness and surface texture. Davies [22] gave an overview of some of the methods to describe the particle shapes. Most of these methods are developed for 2D-projections and to distinguish regular and irregular natural particles (i.e. sands, gravel), and therefore less suitable to describe digitized particles, which are regularly shaped and have equal dimensions in three perpendicular directions, like spheres. For this paper two methods are applied to account for the shape of digitized particles, namely sphericity and digitized roundness. The sphericity is defined by Wadell [24] as;

$$\psi = \sqrt[3]{\frac{V_p}{V_{cs}}} \quad (1)$$

Fig. 3. The relation between the box height ratio (h) and the scaled random packing fraction for different dimensions of the box bottom ratio (l) ($n_t = 10$, $n_m = 20$, $d = 13$).

Fig. 4. The relation between contact number ($\langle C \rangle$) of the random packing and packing fraction f ($n_t = 10$ and $n_m = 20$).

whereby V_p and V_{cs} are the particle volume and the volume of circumscribing sphere, respectively. Based on the 2D definition of roundness by Lees [25], the following 3D definition for digitized roundness is proposed here

$$\varepsilon = \frac{A_p - 6A_t}{A_p} \quad (2)$$

with A_p as the total surface of the digitized particle and A_t the surface area of one of the top surfaces of digitized particle. The top-surfaces

Fig. 5. a) The relation between size ratio l (L_{box}/d) and the product of packing fraction and digitized roundness $f_\varepsilon(l)$ and b) the relation between size ratio l and the product of the packing fraction and the Wadell sphericity $f_\psi(l)$ (based on the data from Fig. 3 and Table 1).

are defined as the surfaces of the digitized particle, lying perpendicular to the three main directions at either $x=r$, $y=r$ or $z=r$, with x , y , z the distance of axis to the middle of the particle and r the radius of the particle. Fig. 1 shows the top-surfaces of a $d=7$ particle in grey as an example. Both the sphericity and digitized roundness are unity for a perfect sphere and digitized roundness is zero for a cube. Furthermore the particle shape can be described using the shape factor which is described by Chen [26] as

$$F = \frac{d \cdot A_p}{V_p} \quad (3)$$

With d the particle size, A_p the surface area of the particle and V_p the volume of the particle. Table 1 shows the shape factor F , Wadell's sphericity ψ and digitized roundness of the digitized particles used in this research. From this table, one can notice that the shape factor F for $d=7$ and $d=11$ is much lower than for the other particles, which partially explains the differences visible in Fig. 2.

In order to derive a correction factor, the digitized roundness and Wadell sphericity are combined with the computed packing results. Fig. 5a and b show the result of multiplying the packing fractions with the digitized roundness and Wadell sphericity, respectively. Furthermore, both modifications result in a packing fraction for all digitized particles that are below the random close packing value of spheres, and tend to the RCP limit for large size ratio h . One can also notice that the results for $d=7$, $d=11$ and $d=21$ become more in line with the results of the other sizes, but are still higher than those of the other particles. Although the shape corrections bring the packing results in line with the RCP-value of spheres, the packing of particles with $d=7$, $d=11$ and $d=21$ are still behaving differently from the other sizes.

A possible solution to bring $d=7$, $d=11$ and $d=21$ in line with the other sizes is to modify the composition of $d=7$, $d=11$ and $d=21$ particles. The particles are namely shaped using a routine that determines if a voxel within the box is part of the digitized particle [14]. A voxel is part of digitized particle in case the distance from the centre of the voxel to the centre of the digitized particle is smaller than half the size of the digitized particle. With the reduction of the diameter by 0.05, the particle shape already alters. The volume of the modified particles decreases compared to the original, while the total particle surface remains equal. In Fig. 1 the removed voxels are colored black/blue. The volume decrease in case of $d=11$ is 48 voxels, which means the removal of 8 voxels in each 6 (top)sides. For $d=21$ besides 8 voxels in each 6 (top)sides, another 96 voxels were removed from the outer layer. For 11 and 21 particles the digitized roundness changes from 0.78 and 0.87 to 0.89 and 0.92, respectively (Table 1), while it remains at 0.76 for $d=7$. While the Wadell sphericity changes from 0.81, 0.90 and 0.93 to 0.84, 0.88 and 0.93, for $d=7$, $d=11$ and $d=21$, respectively (Table 1). The digital roundness of the modified particles are now more in line with those of the other particle sizes. Fig. 6a shows the simulation results for the product of the packing fraction and digitized roundness using the modified particles of $d=7$, $d=11$ and $d=21$, together with the other unmodified particles. When comparing Fig. 6a with Fig. 5a one can notice that the results of the digitized (modified) particles are now closer to the other particle sizes, and only very slightly higher than the other sizes.

To assess the randomness of the generated assemblies, the translational order metric order (T) is computed [20]. Fig. 7 shows the translational order results for $l \geq 5$. The results for $l \leq 5$ are less reliable since the number of particle relations is too limited for a sound statistical analysis, and furthermore finite size and wall effects play an

Fig. 6. The relation between size ratio l and a) the product of packing fraction and digitized roundness $f_\epsilon(h)$ b) the product of packing fraction and Wadell sphericity using the modified $d=7$, $d=11$ and $d=21$ particles $f_\psi(l)$ ($n_t=10$ and $n_m=20$).

Fig. 7. The translational order metric (T) of the simulations concerning $L_{\text{box}}/d \geq 5$, versus a) the product of packing fraction and digitized roundness b) the product of packing fraction and Wadell sphericity.

Fig. 8. Example of the structure of packed bed created using the packing routine for box dimensions $50 \times 50 \times 100$.

important role [18,20,27,28]. Two outliers can be observed in Fig. 7, these are outliers caused by particle of $d = 21$ in a box with $L_{\text{box}} = 300$ and $L_{\text{box}} = 350$.

From Fig. 7, one can notice a negative correlation between packing fraction and translational order metric, which is opposite to the relation shown by Kansal et al. [29] and Torquato [3,30]. Kansal et al. [29] created the lower packing fractions by removing spheres from an face centred cubic lattice leaving the other spheres on existing position, and hence the structure will not change. While in the present research the lower packing fractions are caused by the properties of digitized particles. On the other hand, Torquato [3,30] determined the translational order metric for random packing of spheres also using the Lubachevsky and Stillinger algorithm, and a positive correlation between packing fraction and translational order metric was obtained.

The derived translational order metric from the simulations is in the same range as found by Kansal et al. [29] and Torquato [3,30], which could indicate that the generated (particle) arrangements are random indeed. Especially since Torquato [3,30] showed that the translational order metric of crystalline systems is higher than 0.4 and in the present paper the translational order metric is lower for high values of l (Fig. 7).

Fig. 8 shows an example of a packed structure created using the applied packing routine with bottom dimensions of 50×50 ($L_{\text{box}} \times L_{\text{box}}$) and a height of 100 (H_{box}).

4. Conclusions

Digitized particles are an elementary part of cellular automata models, and the close random packing (RCP) of monosized particles is addressed here. The original packing model of Bentz [14] is not suitable for research of (random close) packing of mono-sized digitized particles, since the found packing fractions are close to the random sequential addition value of spheres. A used model, similar to the

Lubachevsky and Stillinger [2] algorithm, has been introduced in this paper. The found packing fractions during simulations are clearly related to the size ratio (L_{box}/d) for all particle sizes except for $d = 7$ (voxels), $d = 11$ and $d = 21$, which results in higher packing fractions.

The digital roundness was introduced in order to describe the particle shape of the digitized particles. This was not possible by applying the existing methods, since these methods are mostly based on 2D-projection and irregular shaped particles, while the digitized particles are regular shaped and equal dimensions in three main-directions. Using the digitized roundness and Wadell's sphericity, the packing results obtained during simulation could be explained. The product of Wadell sphericity and packing fraction approaches the random close packing fraction of spheres (~ 0.63) for the higher size ratios l (L_{box}/d).

New particle shapes for $d = 7$, $d = 11$ and $d = 21$ have been introduced in order to overcome the aforementioned outliers noticed during packing simulations. The modified particle shapes have a digitized roundness which fits better in the curve together with the other particle sizes and the packing fraction is reduced compared to the packing fraction of the original particle shape used by Bentz [14].

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