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Packing structure of cohesive spheres

J. Q. Xu, R. P. Zou, and A. B. Yu*
School of Materials Science and Engineering, The University of New South Wales, Sydney, NSW 2052, Australia

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This paper presents the first set of measured data to probe into the loose packing structure of wet particles featured with large pores, aggregated and chain-connected particles. The structure is also analyzed in terms of radial distribution function and coordination number, and compared with that of the random dense packing.

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Granular matter is widely encountered in nature and in engineering practice but poorly understood [1–3]. One of its key fundamental areas is the quantification of the structure of particle packing. For cohesionless uniform spheres, mainly driven by the need to model liquids [4], such information has been established [5–7] and widely used in the structural modeling of simple liquids, glasses, and amorphous solids, and transport properties of porous media (see Refs. [7–12] for example). Forming a packing is a dynamic process involving various interparticle forces, including the capillary force associated with wet particles and the van der Waals force with fine particles [13]. The presence of these cohesive forces will significantly change the static and dynamic behavior of particles, which has become a research focus in granular research in recent years (see Refs. [14–24], for example). While steady progress is being made in developing a macroscopic understanding of the behavior of cohesive particles, to date no measured structural information is available resulting in a difficulty to generate a microscopic picture for a better description. To overcome this gap, here we report an experimental study of the packing structure of wet spheres.

The packing experiment performed mainly involves three steps: mixing, packing, and measuring, described as follows. A preset amount of liquid and particles was first mixed manually to ensure a uniform distribution of liquid among particles. The resultant particle-liquid mixture was then slowly poured into a calibrated container through a funnel to form a packing. After the excess particles were levelled off, the packing was weighed, and reweighed after drying. The measured weights, together with the particle and liquid densities, allow the calculation of liquid content, expressed as the percentage of volume ratio between liquid and particles. The ratio of the volume of particles to the volume of the container was used as a measure of the packing behavior, and referred to as dry-based packing density. By definition, the dry-based porosity ε is equal to one minus this packing density. This porosity depends on the way to form a packing. For example, the application of tapping or compaction will overcome, either partially or completely, the effect of cohesive force yielding denser packing [15,19,22]. To highlight the effect of liquid-induced force, we did not introduce tapping or compaction in this work. These concepts, together with the experimental details, have been used in our previous studies of the packing of wet particles [15,20,23].

To highlight the effect of capillary force and increase the precision of measurement, expanded polystyrene beads of particle density 136 kg/m^3 are used in this work. Their sizes are uniformly distributed in a narrow range of 5.3–6.2 mm, with the mean size being 5.75 mm. Optical observation demonstrates the selected beads are reasonably spherical since sphericity, defined as the ratio of the surface area of a sphere of the same volume as a particle considered to the actual surface area of the particle [25], is greater than 0.97. There is no observable deformation of particles before and after packing experiment. So the error from the particle shape and deformation should be negligible, particularly compared to the error in measuring particle diameter and center coordinates. The container used is 255 mm in diameter and 210 mm in height. Liquid properties such as density, surface tension and viscosity affect particle packing [15]. For this work, the liquid used is a mixture of 21% commercial glue (DATS product) and 79% water, of density 995 kg/m^3, viscosity 6.01×10^{-3} Pa s, and surface tension 47.6×10^{-3} Nm^{-1}. It can produce a packing with enough adhesive strength allowing a packing sample to be disassembled one particle at a time without changing the remains. The technique employed to measure the coordinates and diameters of particles is similar to that developed by Bernal *et al.* [26]. Here it involves the use of a TM-500 digital microscope with a three-dimensional mechanical stage manufactured by Mitutoyo Corporation. The precision is within ±0.05 mm in the vertical direction and ±0.01 mm in the horizontal direction, producing a standard error of 0.8% of particle diameter in the coordinates of particle centers. To avoid the wall effect, the packing sample is taken from the central part of a packing, about five particle diameters away from the wall, and the top and bottom surfaces of the packing. The coordinates and diameters of particles in the sample are measured one by one and stored in a computer to reproduce the packing structure for analysis. The size distribution of particles is actually obtained from the resulting measured diameters.

Figure 1 shows the measured relationship between the porosity ε and liquid content, where the error bar was estimated from repeated experiments. It is clear that porosity increases rapidly to a maximum and then decreases with the increase of liquid content. This relationship can be well explained in terms of interparticle forces induced by liquid [27–29]. The results in Fig. 1 are qualitatively comparable to those reported for glass beads [15], thereby providing a reasonable basis for the structural analysis planned. However, the porosity values are much higher, mainly because of the difference in the particle density. The lower the density ratio

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*Corresponding author. Fax: +61 2 9385 5956. Email address: a.yu@unsw.edu.au

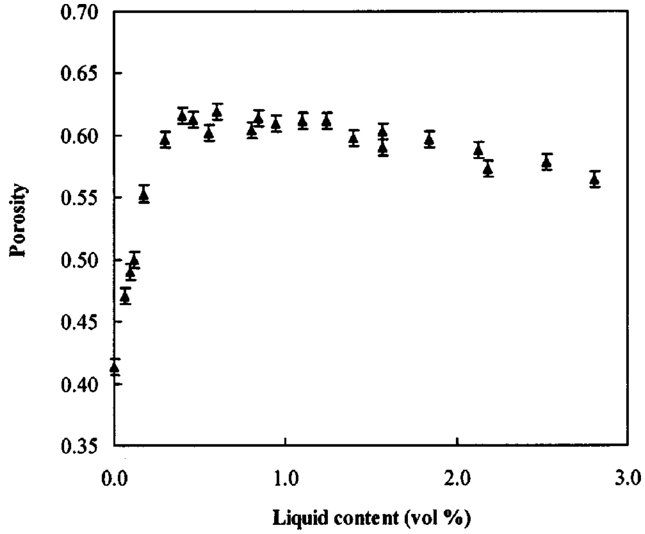

FIG. 1. The relationship between the porosity and liquid content.

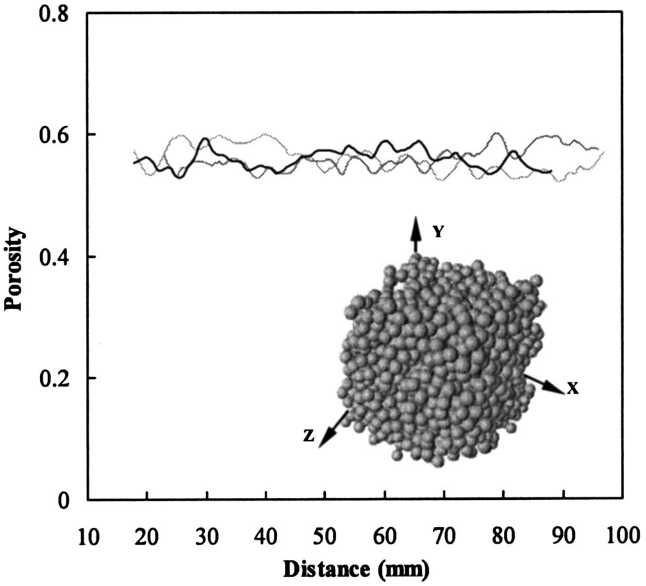

FIG. 2. Variation of porosity along the *x*, *y*, and *z* directions for the packing sample (inset). Dark dotted line, *X* direction; light dotted line. *Y* direction; solid line, *Z* direction.

between particle and liquid, the higher the porosity for a given liquid content. As mentioned, particles of low density have been purposely selected for this work. Note that the low particle density will not affect the porosity of dry particles much. In fact, this porosity is equal to 0.415 under the present experimental conditions, applying tapping and shaking gently can decrease the porosity to 0.388. These values are respectively comparable to the so-called random loose packing (RLP, $\varepsilon = 0.40$) and random dense packing (RDP, $\varepsilon = 0.36$) for coarse spheres [6,8,30]. The higher value observed is mainly due to the relatively high friction between polystyrene particles.

The structure of the packing when the liquid content is 2.5% is then measured using the technique described above. The structure constructed is based on the measurements of 3801 particles, its overall shape is approximately cubic with a length around 105 mm. An analysis of the porosity distribution along the *x*, *y*, and *z* directions shows that its internal structure is reasonably uniform, particularly if the first few layers of particles at the external surface are excluded, as shown in Fig. 2. Note that the *x*, *y*, and *z* coordinates are adjusted values so that the center of the packing is located at about 55 mm in all the three directions. Probably a larger sample can eliminate the fluctuation observed, but this could not be achieved in this work. Nonetheless, the resulting information should be rich enough to construct an overall picture about the packing structure of cohesive particles. The overall porosity of this packing is 0.583 read from Fig. 1 or 0.565 for the structural sample as a result of excluding the wall effect, much higher than 0.36 for the RDP [7,30]. Consequently, the structures differ significantly. As shown in Fig. 3, the structure of this packing is featured with the existence of large pores and aggregated and chain-connected particles. In contrast, the RDP is much denser and has no pore of size greater than one particle diameter. The difference can be quantified in terms of coordination number and radial distribution function, as discussed below.

Figure 4 shows the distribution of coordination numbers for different critical distances. As used by other investigators [8,9,18,29], the so-called critical distance is defined as the distance between particle centers less than which two particles are said to be in contact. Compared to the dry, RDP packing, the distribution does not change much in shape but shifts toward smaller coordination numbers. The mean coordination number is reduced by about 3 when the critical distance is the 1.1 apparent particle diameter, which here is defined as the sum of the radii of two spheres in contact, but the difference is smaller for a smaller critical distance. Notably, there are a proportion of particles whose coordination number is as small as 2, indicating that a chainlike structure exists in this wet packing.

The radial distribution function is another important parameter to characterize the structure of a packing. It is here defined as $g(r) = \Delta N(r)/(4\pi\Delta r \rho_0)$, where $N(r)$ is the average number of particle centers within a spherical space with radii r, nondimensionized by the mean particle diam-

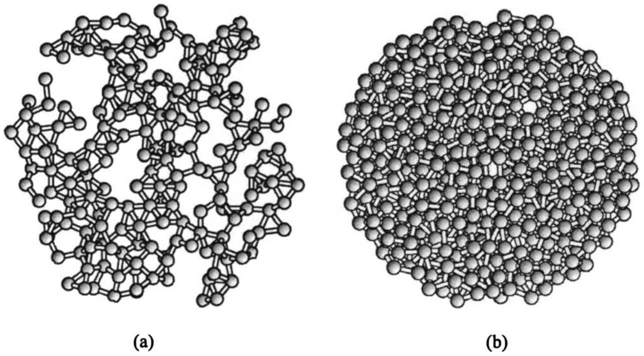
(a) (b)

FIG. 3. Comparison between the measured structures for the present wet packing (a) and the RDP of Finney (b). Here the contact network among particles is established for a spherical sample cut at its center with a thickness of two particle diameters.

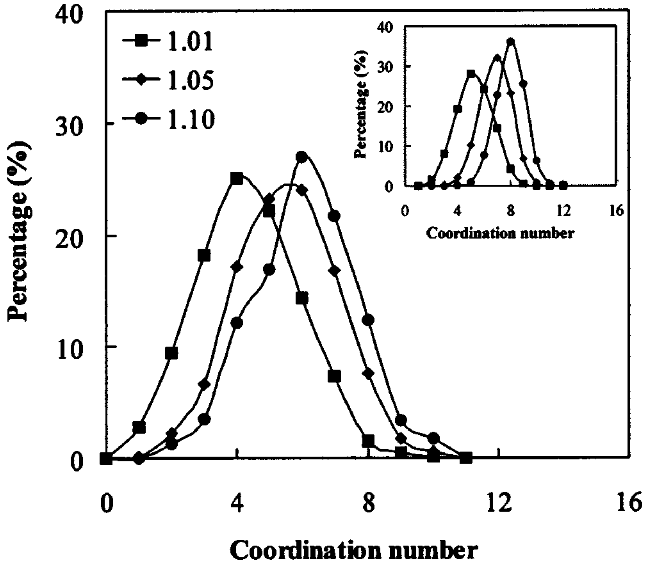

FIG. 4. Distribution of coordination numbers with different critical distance, compared to that for the RDP (inset).

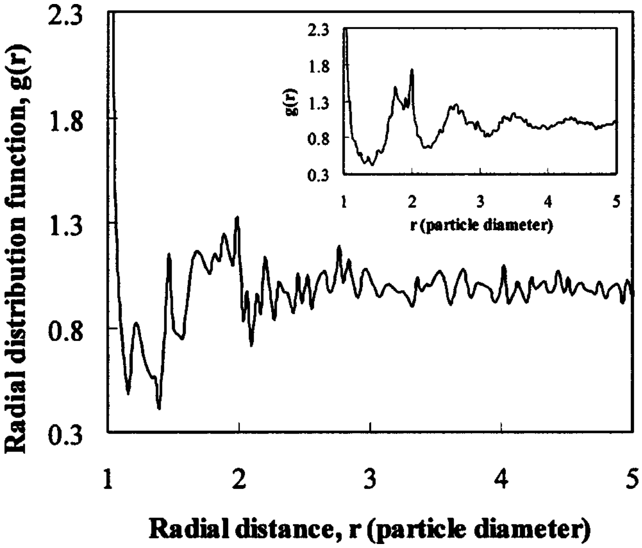

FIG. 5. Radial distribution function compared to that for the RDP (inset), both obtained when $\Delta r = 0.03$ is the particle diameter.

eter, around a reference particle and ρ_0 is the average number of particle centers in unit space of packing, equal to $6(1-\varepsilon)/\pi$. For the RDP, the most striking feature is the split second peak at $r = 1.73$ and 2.0 in the $g(r)$ function [7]. As shown in Fig. 5, this feature is also observed in this wet packing. However, the following key changes can be identified. First, the peaks at $r = 1.74$ and 2.0 are much weaker. Second, the peaks beyond the second one vanish. Third, the variation of radial distribution function is largely limited to a small distance $(r<2)$, and there is a strong peak at $r = 1.47$.

These changes must come from the effect of the capillary force. As illustrated in Fig. 3(a), this cohesive force results in the formation of agglomerates or aggregates in which the structure is relatively dense, so that some RDP structural features are inherited. On the other hand, the large pores among agglomerates and the existence of chain-connected particles in the packing mean a more irregular short-range structure and a more uniform long-range structure. Consequently, $g(r)$ varies significantly when $r<2$, and just fluctuates around its mean and has no strong peaks when $r>2$. For the RDP, as shown in Fig. 3(b), the connections among neighbor spheres are mainly triangular, leading to a high proportion of tetrahedra, 72.98% by number [7]. For cohesive spheres, Fig. 3(a) shows that a large proportion of spheres are connected in a twisted rectangular form corresponding to the peak in $g(r)$ at $r = 1.47$.

Interestingly, most of the packing features revealed here are similar to those for fine or wet particles, recently found by Yang *et al.* [18,29] by means of a granular dynamic simulation. This fact supports that the van der Waals force and the capillary force function similarly, both restricting the relative movement between particles and resulting in loose packing structure. Computer simulation has been recognized as an effective method to generate information for structural analysis. Obviously, a successful simulation method must take into account all dynamic factors related to both geometry and force. Most of the previous methods only considered the former and ignored the latter, and hence failed to generate a realistic packing structure, particularly when forces rather than the gravity are dominant [31]. A dynamic simulation like that used by Yang *et al.* is therefore very attractive in the study of cohesive particles. However, at this stage of development, the equations to calculate the interparticle forces are not so perfect and their application must be carefully examined. Experimental measurement, although arduous, is necessary in order to generate a sound basis for model validation and further development.

Here we show that such experiments can be done with proper experimental materials and techniques. Our results clearly demonstrate that the presence of cohesive forces among particles results in large pores, aggregated and chain-connected particles in a packing. The data should be very useful for structural modelling and analysis of cohesive particles. They are available to interested readers [32].

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[32] Please address to ABY; email address: a.yu@unsw.edu.au