Universal Properties and Classification of Randomly-Packed Particles with Power Size Distribution

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Randomly packed particles lose fluidity above a certain packing fraction, namely the jamming point. Although the jamming point and packing structure of monodisperse particles of equal size $r$ have been intensively studied, the general properties of polydisperse particles of various sizes remain unclear owing to their enormous variety of size distributions. Here, we focus on the packing structure of polydisperse particles with a power size distribution $r^{-a}$, where the characteristic length is expected to be ambiguous. We experimentally and numerically constructed two-dimensional packing structures for various polydisperse systems with different exponents $a$. By analyzing the contact number $z$, namely, the number of particles in contact with each particle, we found that universal properties appear in $z$-distribution when the exponent lies within the range $2 < a < 3$. The $z$-distribution follows a power distribution with an exponent of 3.8 independent on $a$. We derive the exponent $\sim 3.8$ from a model with simple assumptions regarding the distribution and average contact number. Furthermore, the jamming point has a peak within this $a$ range, reaching above 0.90, which is higher than the $\sim 0.84$ for a monodisperse system. These results demonstrate the validity of systems with power-size distributions to derive universal properties for polydisperse systems, ranging from granular materials to intracellular biopolymers.

jamming | glass | self-similarity | polydisperse | emulsion

Randomly packed particles flow like a fluid at a low packing fraction $\phi$ and harden like a solid with an increase in $\phi$. This transition occurring at a critical packing fraction $\phi_C$ is called the jamming and glass transitions in athermal and thermal systems, respectively. For monodisperse particles of equal sizes, the universal properties of the packing pattern at the jamming point are well known (1), such as the critical packing fraction $\phi_C \sim 0.84$ (2) and average contact number, which denotes the average number of particles in contact with each particle, $\langle z \rangle = 4$ (3). In contrast to monodisperse particles, the information about polydisperse particles of various sizes in the jammed and glassy states is very limited. However, such polydisperse materials are ubiquitous in our daily life at various size scales, such as granular materials (4) and biopolymers in living cells (5). A universal understanding for such materials is needed in various fields, including soft matter applications, biophysics, and material engineering.

Some studies on polydisperse systems have revealed that such size polydispersity is crucial in the packing structures and physical properties. For example, polydisperse particles are piled to reach a higher density than monodisperse particles $\sim 0.84$ (6). In addition, bidisperse particles exhibit two distinct packing patterns (7). However, to the best of our knowledge, there are no reports on the universal properties of polydisperse systems or the particle size distributions that give polydisperse systems their distinct characteristics.

To derive the universal properties of polydisperse systems, we focus on randomly packed particles whose radii $r$ follow a power size distribution $r^{-a}$. This is because the ambiguous characteristic length and scale-free nature are expected to highlight the universal properties. We experimentally and numerically constructed two-dimensional packing patterns of polydisperse particles with $r^{-a}$ and analyzed the patterns and jamming transitions by varying the exponent $a$.

Universal properties of packing patterns. We analyzed the randomly packed particles with $r^{-a}$ in two dimensions at the jamming point and compared with monodisperse system. Figure 1(b) shows a microscopic image of the packing pattern of monodisperse droplets. The packing fraction was $0.85 \pm 0.01$. To avoid crystallization, we treated a bidisperse system as a monodisperse system by mixing two differently sized particles. The ratio of the radii $r$ was approximately 1:1.5. From these images, we calculated the distribution of the particle radius $N(r)$ and distribution of the contact number $\nu(z)$ as depicted in Figures 1(c) and (d). During the analysis of the contact number, particles with $z \leq 2$ (not included in the contact network) were successively removed as rattlers. The contact number distribution $\nu(z)$ has one peak with an average value of $\langle z \rangle \approx 4.3$, which is similar to the ideal value of monodisperse systems $\langle z \rangle = 4$ (3).

Similarly, the distributions of $N(r)$ and $\nu(z)$ for polydisperse particles were calculated from the microscopic images, as shown in Figures 1(e) to (g). The packing fraction was $0.97 \pm 0.05$. For this polydisperse system, $N(r)$ has a power size distribution $a \simeq 3$. The resulting average contact number was $\langle z \rangle \approx 4.8$. Although this value is slightly larger than that of the monodisperse system, the difference is insignificant because of the experimental error near the critical point. On the other hand, the contact number distribution $\nu(z)$ also followed a power distribution $\nu(z) \propto z^{-\gamma}$ but decayed more rapidly than $N(r) \propto r^{-a}$ with $a \simeq 3$ (Figure 1(g)). The exponent of $\nu(z)$ was found to be $\gamma \approx 3.8$.

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To investigate the generality of the contact number distribution for polydisperse systems, $\nu(z) \propto z^\gamma$ with $\gamma \approx 3.8$ suggested by the experiments (Figure 1(g)), we numerically produced randomly packed patterns of various polydisperse droplets with different $a$ values. Figure 2(a) shows examples of numerically produced packing patterns for $a=1.5$, 2.3, 3, and 4. For $1 < a < 4$, the contact number distribution $\nu(z)$ was calculated as plotted in Figures 2(b) to (f). Figures 2(c) to (e) show that $\nu(z)$ for $a=2$-3 follows a power-law distribution with $\gamma \approx 3.8$ independent of $a$. This exponent agrees with the experimentally suggested value of $\gamma \approx 3.8$ (Figure 1(g)). In addition, we have confirmed that $\langle z \rangle$ is approximately 4 regardless of $a$, same as that of the monodisperse system. These results demonstrate a universal property for the contact number distribution of randomly packed polydisperse particles within $a=2$-3. For smaller and larger exponents ($a < 2$ and $a > 3$), deviations from the power law were observed.

To explain the universal property of $\nu(z) \propto z^\gamma$ with constant $\gamma \approx 3.8$ in the range $2 < a < 3$, we modeled the system with two assumptions for dimension $d$: (i) If the particle size follows a power distribution, then the contact number also follows a power distribution $\nu(z) \propto z^{-\gamma}$ for $z \geq d + 1$ and $\nu(z) = 0$ for $z \leq d$ for all particles except rattlers in contact with at least $d+1$ particles. (ii) $\langle z \rangle = 2d$. This assumption is based on the fact that $\langle z \rangle$ is at least $2d$ for a jammed system, and easily derived from Laman’s theorem (8) when $d = 2$.

Under these assumptions, the following equality holds:

$$\langle z \rangle = \frac{\sum_{z=3}^{\infty} z \nu(z)}{\sum_{z=3}^{\infty} \nu(z)} = 4. \quad [1]$$

This can be expressed as follows:

$$\frac{\zeta(\gamma - 1) - 2^{-\gamma + 1} - 1}{\zeta(\gamma) - 2^{-\gamma} - 1} = 4. \quad [2]$$

Solving Eq.2 yields $\gamma = 3.83...$, which explains the property obtained experimentally and numerically (Figures 1, 2). The exponent $\gamma \approx 3.8$ is considered sufficient as long as assumption (i) remains valid. It suggests that the assumptions is not satisfied with $a < 2$ and $a > 3$. (Figure 2(b) and (f))

**Classification of polydisperse systems from jamming point.**

We identified that the universal contact number distribution $\nu(z) \propto z^{-3.8}$ holds in the range $2 < a < 3$. To clarify the physical meaning of the range $2 < a < 3$ and classification of such polydisperse systems, we investigated the jamming transitions for various polydisperse systems. Pressure $P$ was numerically calculated over a wide range of $-5 \leq a \leq 10$ and plotted against the packing fraction $\phi$ (Figure 3(a)). The fraction $\phi$ at which $P$ began to rise was plotted against $a$ as the jamming point $\phi_c$ in Figure 3(b). For extremely small and large values $a = -5$ and $a = 10$, respectively, $\phi_c$ is close to the well-known value for monodisperse systems, that is, $\sim 0.84$ (2). However, $\phi_c$ reaches a maximum value within the range $2 < a < 3$, where a universal value of $\gamma$ exists. This means that the exponent range $2 < a < 3$ exhibits particularly strong characteristics of polydisperse systems.
Characteristics in packing. We have shown that the packing pattern and jamming transition in polydisperse systems with $2 < a < 3$ deviates from those from monodisperse systems. To explain why polydispersity is enhanced in this $a$ range and scale-free nature holds, here we discuss the implications of this range in terms of characteristic length scales. The upper and lower limits of the particle size, namely, $r_{\text{max}}$ and $r_{\text{min}}$ may affect the packing pattern as the characteristic length scales of the packing.

First, we consider the effect of $r_{\text{min}}$, based on a comparison with an ideal system with $r_{\text{min}} = 0$. For $a < 3$, the total area of particles under $r_{\text{min}}$, $\int_{r_{\text{min}}}^{r_{\text{max}}} \pi r^2 N(r)$, can be made as small as desired by taking $r_{\text{min}}$ sufficiently small, which renders the effect of $r_{\text{min}}$ negligible. However, for $a > 3$, the divergence of $\int_{0}^{r_{\text{max}}} \pi r^2 N(r)$ makes the packing of particles with an ideal distribution undefined. For the actual system, we must set a finite $r_{\text{min}}$ because the limit of $r_{\text{min}} \rightarrow 0$ cannot be taken. It means that $r_{\text{min}}$ behaves the characteristic length for $a > 3$.

Next, we consider the effect of $r_{\text{max}}$ based on a comparison with the size distribution that allows for complete packing. Complete packing is defined as packing without voids constructed by optimal arrangement. Apollonian packing is an example with the smallest number of particles (9, 10), in which the smallest $a$ is described as $a = d_{t} + 1$ where the fractal dimension $d_{t} = 1.30$...

For sufficiently large $a > 3$ and small $a < 2$, a characteristic length scale emerges, and the scale-free nature is broken, which violates assumption (i). In other words, both $r_{\text{min}}$ and $r_{\text{max}}$ have negligible effects on the pattern in the range $2 < a < 3$. This ambiguity in the characteristic length scale enhances the polydispersity and leads to a universal property for polydisperse systems in the range $2 < a < 3$.

We have demonstrated that the packing of polydisperse particles with $N(r) \propto r^{-a}$ has a universal property for the contact number distribution when the exponent is in the range of $2 < a < 3$. This corresponds to the range in which a particularly strong polydispersity appears during the jamming transition. The proposed classification based on $a$ may be applicable to various polydisperse systems with a power size distribution (11, 12) and general probability distributions by generalizing $a$ as follows:

$$a = \frac{\ln N(r)}{\ln r},$$

where $r \ll 1$.

Furthermore, our results suggest that the condition $d_{t} + 1 < a < d + 1$ enhances the characteristics of polydisperse particles for general dimensions. Therefore, our findings should greatly contribute to the understanding and application of polydisperse systems with higher dimensions.

Materials and Methods

Particle packing was prepared by sandwiching an oil droplet in water between two glass plates having a thickness comparable to the diameter of the smallest particle. (Figure 1. See also Supplementary Information (SI) S1. A-D). This confinement deforms the particles into a pancake shape and prevents larger particles overhanging onto smaller ones. With very slow evaporation of water, the area fraction of the particles increases. We analyzed the packing pattern at the jamming point.

For the calculations, the $r$ of the particles were randomly set according to a given probability distribution $N(r) \propto r^{-a}$ in the range $r_{\text{min}} < r < r_{\text{max}}$, where $r_{\text{min}}$ and $r_{\text{max}}$ are the lower and upper bounds of $r$, respectively. Random placements in cells with periodic boundary conditions were used as the initial conditions (See SI S1. E).

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1. Materials and Methods

A. Materials. Distilled water (UltraPure™ DNase/RNase-Free Distilled Water, Invitrogen, Waltham, MA, USA) with a surfactant Tween 20 (Sigma-Aldrich, St. Louis, MO, USA) was used as a continuous phase. Mineral oil (Nacalai Tesque, Kyoto, Japan) with a surfactant Span 80 (Tokyo Chemical Industry Co., Tokyo, Japan) was used as a dispersed phase. To facilitate identification of the oil-in-water (O/W) droplets, a lipophilic dye, capsanthin in vegetable oil (Tokyo Chemical Industry Co.) was added to the mineral oil.

B. Droplet preparation. We used O/W droplets to prepare randomly packed particles. An aqueous solution containing 1 wt% Tween 20 was used for the continuous phase and a mineral oil containing 0.1 wt% Span80 and 3 wt% capsanthin oil for the dispersed phase.

The polydispersible droplets with a power size distribution were produced by impact fracture. To the microtube containing 500 µL aqueous solution, 150 µL of mineral oil was added in 3 portions. Each time oil was added, the microtube was tapped with a finger. The radii of prepared droplets are ranged from 10\(^{-5}\) to 10\(^{-3}\) mm. The principle of polydispersible droplet preparation based on impact fracture will be reported in a forthcoming paper.

For monodisperse droplets, we used a centrifugal microfluidic device, which is a modified version of the previously reported device (1). The device consists of three parts: a glass capillary, a micropipette tip (Labcon, Petaluma, CA, USA), and a microtube. The glass capillary with a thin tip of \(\sim 30 \mu m\) and length of 8 mm from the tip was fabricated from the ready-made capillary (outer diameter: 1 mm, inner diameter: 0.6 mm; G-1, Narishige, Tokyo, Japan) by using a puller (PC-10, Narishige) and microforge (MF-900, Narishige). The capillary was attached to the end of a micropipette tip (200µL standard yellow pipette tip). The micropipette tip was filled with 80 µL of mineral oil and fixed on the microtube containing 500 µL of aqueous solution by passing through the 6 mm diameter hole drilled in the lid of the microtube. The device was centrifuged at 6,000 rpm for 1 minute using a table-top centrifuge (Wako Pure Chemical Industries, Osaka, Japan). The prepared droplets have two different sizes, approximately 50 µm and 80 µm (See Figures 1(c)).

C. Preparation of randomly packed particles in 2D. To prepare the randomly packed particles in quasi-two-dimensional (quasi-2D) space, the O/W droplets were confined between two slide glasses (76 mm \(\times\) 26 mm, thickness \(\sim 0.9\) mm, Matsunami, Osaka, Japan, S1111). These glasses were laminated together with \(\sim 30 \mu m\)-thick double-sided tape to nearly match the diameter of the smallest droplet \(\sim 20\) µm. To cover the glass surface with an aqueous phase, the hydrophilicity of the glass slide surface was improved by using the plasma cleaner (Harrick Plasma, PDC-32G, Ithaca, NY), which reduces the friction with the oil droplets. This quasi-2D confinement deform the droplets into a flat pancake shape and eliminates overlap between droplets. Hence, the center and edges of the droplet can be clearly identified. The water surrounding the droplet evaporates very slowly over time (\(\sim 5\%/h\)). This process increases the total area fraction occupied by the droplets (i.e., the packing fraction \(\phi\)), while maintaining the area of each droplet. Here we determined the moment just before the first avalanche occurred as the jamming point and analyzed the packing pattern at that point.

D. Image Analysis. Images of droplet packing were acquired using a camera (a2A5328-15scPRO, Basler) attached to a microscope (SZX16, Olympus). The images were analyzed using free NIH software, ImageJ (http://rsb.info.nih.gov/ij/). The droplets in the images were detected by binarization after removing noise with a median filter and FFT bandpass filter. When the distance between the droplet surfaces was less than 1 pixel, the droplets were determined to be in contact with each other.

E. Numerical simulation. Initially, 4,000 circular particles are randomly placed in a square space with periodic boundary conditions. The radii of the particles \(r\) are randomly set according to a given power size distribution \(r^{-\alpha}\). The exponent \(\alpha\) is changed in the range \(-5 < \alpha < 10\), keeping the size range \(r_{\text{min}}/r_{\text{max}} = 0.02\), where \(r_{\text{min}}\) and \(r_{\text{max}}\) are the lower and upper bound of the distribution, respectively. Increasing \(r\) of all particles in the same ratio raises the packing fraction. The particles have a pure repulsive potential \(U\):

\[
U = (D_{ij} - r_i - r_j)^2 \left( \frac{1}{r_i} + \frac{1}{r_j} \right),
\]

where \(D_{ij}\) is the distance between \(i\)-th and \(j\)-th particles, and \(r_i\) and \(r_j\) are radii of the \(i\)-th and \(j\)-th particles, respectively. The initial placement takes 3,200,000 steps to relax the randomly-packed pattern by FIRE algorithm (2) and the packing fraction was then increased six points over 3,200,000 steps. The pressure of the system \(P\) was calculated to derive the jamming point.

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