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Kyoto University
Packing Structure of Fine Powder

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Bulkiness of powder, an important characteristic in powder science, was studied to determine the relationship between particle size and cohesiveness. In order to know what determines the bulkiness of powder, a number of experiments were done.

The variation of the void volume of powder beds with particle size, and the change of porosity by adding of fine particles to coarse one were measured. Results suggest that the packing process of powders is determined in relation to particle gravity and cohesive force at a contact point between particles.

The cohesive force at a contact point was also measured by shearing tests of powder beds of various particle size. From the experimental results, it was observed that the fine particles form the scaffold structure in the aggregated state; confirmed by a model experiment with foaming polystyrene particles.

I. INTRODUCTION

A powder is an assembly of fine particles of solid at the state in which the particles can be moved by a small force. Powder has two important faces; (1) the characteristic of the particle itself which has been studied mainly from the view point of physical chemistry of solid or surface chemistry; (2) an assembly of the particles which has been studied in the field of the chemical engineering process. Hitherto, these two standpoints have been independent having very little influence on each other. Under these state, however, the problem of particle size has been taken up as a common basic property.

In this paper, the problems of how the particle size affect the bulk density and the structure of the assembly of powder particles will be discussed.

II. RELATIONSHIP BETWEEN BULKINESS AND PARTICLE SIZE

It is well known that the bulkiness of powder increases with decreasing particle size. Bulkiness is defined as the reciprocal of the apparent density. Roller investigated the relation of bulkiness to particle size in four powders. In this case the pore volume was used instead of the bulkiness. The relationship between pore volume $V_p$ and particle size $D$ was considered as consisting of two parts, one of which was expressed by the equation (1) for particle size $D$ less than a critical diameter $D_c$ and in the other the bulkiness was constant for $D$ greater than $D_c$.

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\[ V_E = k_1 \left( \frac{1}{D} \right)^n \]  \hspace{1cm} \ldots (1)

In the equation (1), \( k_1 \) and \( n \) were constants defined by the characteristics of the particles and experimental methods, but those physical meanings were unknown. In order to confirm the equation (1), we have measured the pore volume of samples of zinc, \( \alpha \)-alumina, calcium carbonate and potassium nitrate.

The samples were sieved into a small flat container until the powder filled it. Bulkiness was determined by the measurement of the weight of the haphaz-

![Fig. 1. Variation of pore volume \( V_E \) for particle size \( D \).](image)

![Fig. 2. Relationship between \( n \) and reciprocal value of density of the materials.](image)
ardly packed powder in the container. The relationship between \( \log D \) and \( \log V_e \) is shown in Fig. 1, and is in agreement with the Roller's equation. The plotting of the value of \( n \) obtained from the slope of these lines against the reciprocal values of the density of the materials give a straight line through the origin as shown in Fig. 2. The value of \( n \) decreases as the density of the particle increases, coming to zero at infinite density.

For the very high density particles, it is expected that the bulkiness has a constant value not related to particle size. On the other hand, it becomes harder to pack the particles in proportion as the particle density decrease. From these experiments, it is concluded that, if the cohesion force at the contact point is smaller than gravity of a particle, the particles take up positions of minimum potential energy related to the packing formed at that stage, as shown in Fig. 3 (a). Therefore, the bulkiness shows the constant value not related to particle size. However, when the particles whose weights are smaller than cohesion forces are deposited, they remain in that position assumed at the point of initial contact with the deposited particles and consequently form a bulky loose structure as shown in Fig. 3 (b).

Fig. 3. Schematic diagram of packing state for powder.

Accordingly, the value of \( D \) means the particle size at which the gravity of a particle is equal to the cohesion force at the contact point, and \( n \) is the compactionness factor that relates to the properties of attractive force of interparticles and particle shape etc.

In this case, we have not considered in regard to the mechanism of cohesion, but the following may be given as the cause of the attraction force the friction caused by surface roughness, surface tension of adsorbed water and electrostatic force etc.

Consequently, it is reasonable to consider that the simple packing phenomena are determined by the proportion between the gravity of a particle and cohesion force of inter particles.
III. VARIATION IN POROSITY UPON ADDITION OF FINE POWDERS

The above experiments were done where the relatively monosized granular particles are allowed to fall on to a growing deposit. Normally, powders consist of the particles of different sizes. There are some cases where the interstices in a packing of large particles are occupied by small particles and then the voids decrease. Packing models proposed by Hudson, Horsfield, Furnas are ordinary used for mixed powders of different sizes, and are not applicable to fine powders in which the attraction force of inter particles cannot be neglected.

The change of bulkiness based upon mixing of fine powders having different particle size, was measured by the same method previously mentioned. The samples used in this experiment were α-alumina and calcium carbonate which had been sized commercially by elutriation or air separation. Particle sizes of the samples are shown in Table 1. Particle size were determined by the air permeability method, and WA #800 was used as standard coarse powder.

The porosity increases rapidly by addition of fine powders as shown in Figs. 4, 5, but remains constant within a certain range of further addition of fine powders. However, porosity increases again upon more addition of fine powders.

<table>
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<tr>
<th>materials</th>
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<th>fine powder</th>
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<tr>
<td>mark</td>
<td>α-alumina</td>
<td>calcium carbonate</td>
</tr>
<tr>
<td></td>
<td>#800</td>
<td>#3000 #4000 #6000 S 5 M</td>
</tr>
<tr>
<td>particle size</td>
<td>11.8 3.5 3.0 1.2</td>
<td>8.4 5.0 2.4 3.4</td>
</tr>
<tr>
<td>symbol</td>
<td>—</td>
<td>83 84 86 8CS 8C5 8CM 8T</td>
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Fig. 4. Change of porosity produced by mixing of small particles.
In Figs. 4 and 5, the change of the porosity with the weight ratio is shown, where $W_s$ is the weight of added powder, $W_a$ is that of total powder mixed.

In Fig. 6, the number ratio is used instead of the weight ratio. In this case, the number ratio until which the porosities initially increased are $0.5 \sim 1$ independently of the size of added particles, but the one from which the porosities increased again are greater as the particle size decreases. The relation between the number of small particles to a large one at these point, $N_n$, and the particle diameter ratio $D_s/D_l$ are shown in Fig. 7.

The mechanism of these phenomena is considered as follows; assuming that
the cohesion force at a contact point of large particles is slightly smaller than the momentum of a particle and each particle cannot be supported at a contact point but can have two or more points, the particles will assume geometrically stable position producing a relatively dense packing as shown in Fig. 8 (a). Fine particles cohere to the surface of larger particles because the cohesion force of fine particles is relatively great. The large particles are supported with more contact points and make a bulky packing as shown in Fig. 8 (b).
are two effective contact points obtained by the cohesion of a small particle, so
the number of small particles to large one are 0.5, being in agreement with the
results in Fig. 6. Addition of more particles increases only the number of small
particles ineffective to the contact between large particles. These small particles
can not change the whole structure, and the porosity remains at almost con-
stant value since the weight of small particles is only 2~3%. But after the
surface of large particles is covered with cohesived small particles, the cohe-
sion between fine particles mainly influence the packing process and the porosity
increase until the powder bed consists of small particles only. Accordingly, $N_n$
depends upon the number of small particles cohering to the ineffective surface
of large particle and increases as the particle size decreases.

The relation between the increase ratios of porosities due to the addition
of the small particles and their particle sizes is shown in Fig. 9. The difference
between $\alpha$-alumina and calcium carbonate is observed. The relation between
the weight fraction of the small particles added and the increase ratio of
porosity of powder bed in every mixed powder gives the straight line as shown
in Fig. 10, and can be expressed in following equation

$$
\frac{W_s}{f(\varepsilon)} \times \frac{W_s}{W_0} = \alpha + \beta \frac{W_s}{W_0}
$$

where $\varepsilon$ is porosity and $f(\varepsilon)$ is the increase ratio of porosity, i.e. $(\varepsilon - \varepsilon_0)\varepsilon_0$.

The physical meaning of the constants $\alpha$ and $\beta$ is not yet known, but they
are considered to be useful as parameters standing for the coherency of powder.

Fig. 9. Relationship between the increase ratio
of porosity and particle size of adding
particles.

Fig. 10. Relationship between $W_s/f(\varepsilon)$
$\times (W_0-W_s)$ vs $W_s/W_0$.  

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IV. MEASUREMENT OF THE INTERPARTICLE FORCE

The shearing tester was used for the powder beds in order to obtain the interparticle force. The relation between the weight added to the powder bed and the shearing stress is linear as shown in Fig. 11, allowing to apply Coulomb's equation (3) to them.

\[ F = \mu W + C \] ............(3)

where \( F \) is the shearing stress, \( W \) is the weight added to the powder bed, \( \mu \) is the frictional coefficient and \( C \) is the cohesion force per unit area which can be obtained from the intersection on the longitudinal axis. In order to calculate the cohesion force per a contact point between two particles, \( C_p \), Rumpf's equation was used as follows,

\[ C_p = \frac{C}{K_0} \]
\[ K_0 = (1 - \varepsilon) k / \pi D_p^2 \] ............(4)

where \( k \) is co-ordination number of a particle and \( \varepsilon \) is the porosity in the powder bed. The co-ordination number is expected to vary with the packing state, but we reckoned that \( \varepsilon \cdot k = 3.1 \) according to Rumpf's assumption.

The cohesion force obtained by the above method about various \( \alpha \)-alumina powders is plotted to the particle size as shown in Fig. 12. The dotted line is the relationship obtained for the samples coated with fatty acid, but this treatment is considered to have no effect upon the interparticle force. According to Rumpf, the cohesive force per a contact point of \( 10^{-5} - 10^{-6} \) g would be classified to the range of van der Waals' force between two particles with the adsorbed layer, and it is considered to be appropriate for such inorganic powder as \( \alpha \)-alumina which is not hygroscopic and chemically stable.
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Fig. 12. Cohesion force per a contact point for various particle size.

The cohesive force between two sphere particles due to van der Waals' force had been expressed as follows by Hamarker,

\[ C_p = AD_p / B a^2 \]

where \( A \) and \( B \) are the constants, and \( a \) is the distance between two particles. According to this equation, the cohesive force \( C_p \) has to be proportional to the particles size \( D_p \). However, Fig. 12 shows that the cohesive force for the sample #6000 is nearly 1/100 of that for the sample #800, while the particle size of the former is nearly 1/10 of that of the latter. It is considered that this inconsistency is due to the fact that we had reckoned \( k=3.1 \) in equation (4). Because, this relationship had been found by Smith for the packing of lead shots where the void ratios were 0.259~0.447, and it would not be acceptable for the case of the fine particles like as the sample #6000 of which void ratios are very large. But the cohesive force \( C_p \) obtained for the sample #800 is considered to be proper, because the void ratio of this sample was 0.47 which was near the range used in Smith's experiment. So, the cohesive force of #6000 would have to be 1/10 of that of #800, \( i.e. \ 4 \times 10^{-8} \) g, for the particle size is 1/10 of that of the
latter. When the co-ordination number is conversely calculated with this value of $C_p$ from the equation (4), it is obtained to be about 2.3. This indicates the state in which the particles are connected in a row and branched in several places as shown in Fig. 13.$^9$

The relation between the void ratio and the co-ordination number obtained by the above method is shown in Fig. 14 together with others, i.e. experimental results of Smith, Ridgway and Tarbuck,$^{10}$ and theoretically calculated results of

$$\epsilon \sim \frac{n}{C_p}$$

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From these results, it is concluded that the relationship $e/k = 3.1$ is proper in the case that the void ratio lower than about 0.5, but in higher range, the co-ordination number becomes less than the value obtained from that relationship. At the same time, it was found that the particles formed the net structures (so-called the scaffold structures) in the packing states of high void ratio.

V. A MODEL EXPERIMENT OF PACKING WITH FOAMY POLYSTYRENE PARTICLES

According to the observations of the stereophotographs, to some extent naturally deposited fine particles form a scaffold structure. In order to test this idea more experiments were done to determine that these structures appear when the interparticle force are greater than the weight of a particle. Large visible particles with a very small density are preferable. Commercial foamy polystyrene particles which density was about 0.02 g/cm$^3$ and diameter was 3~8 mm were used. The cohesive force of these particles, however, is not so large as to keep a particle at a contact point against the weight of the particle. The apparatus which shown in Fig. 15 was used so as to increase the cohesive force between polystyrene particles. In Fig. 15, A is a desicater and B is a vessel which contained benzene. B was slightly heated to allow to saturate benzene vapor in A, and polystyrene particles were fallen from the top of A. The particles surface had been slightly solubilized in benzene vapor during falling adhered to the packing at the touching points and forming a large void ratio. Fig. 16 (a) shows an example of this packing. The scaffold structure, as shown in Fig.
13, is distinctly observed in this figure. Figure 16 (b) shows a packing which formed without treatment. The co-ordination numbers of the particles in the scaffold structure were measured and the results are shown in Fig. 17. The distributions are nearly the same as two samples, one of them is the originals (marked with ○) and the other is the sample made by sieving of the originals (marked with ●). These results agree with the dotted line which was obtained.
from the theoretical calculation by Nakagaki and Sunada. The average coordination number of the particles obtained from the distribution curve in Fig. 17 is about 2.5 and the void ratio at the above state is 0.65\~0.70.

The cohesive force at a contact point between two particles in these loose packing was measured by the following two methods. One of them was the direct method; namely, a cluster consisting of several particles was hung to the hook of the balance and the terminal particle was pulled slowly down. Then the scale was read when that particle was separated from the cluster. The distribution of the cohesive force of about 300 particles obtained by this method is shown in Fig. 18. Average cohesive force of them is 58.5 mg.

The shearing test was also used. In this experiment, the particles were packed in a cylinder of which diameter was 11 cm by the same method as shown in Fig. 15. The results are shown in Fig. 19 which includes the one of ordinary packing. Both of them satisfies the Coulomb's law. According to the equation (4), the cohesive forces per a contact point in the two packing states are estimated at 0.7 mg and 38 mg respectively.

As the particles not treated with benzene vapor cannot be supported by the cohesive force of 0.7 mg due to a weight was 2\~20 mg, these particles are densely packed. The particles accumulated in benzene vapor are loosely packed due to their large cohesive force. This value of cohesive force obtained by the shearing test agreed considerably with the result of the direct method.

Accordingly, it can be concluded that the value C in Coulomb's equation obtained on the adaptation to the shearing test is satisfactory as the cohesive force between particles.
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Fig. 19. Relationship between shearing stress and load in polystyrene particle bed.

REFERENCES