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Effect of Particle Characteristics on Packing Structure

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The effects of the particle-shape (which is irregular, rod-like or spherical) and the ratio of interparticle-cohesion/particle-weight on the initial fractional voidage before tapping, the rate of tapping-compaction and the final fractional voidage attained by tapping were studied. The samples were monodispersed powders with their sizes ranging from 12 to 127 μ.

A good correlation was found between the initial fractional voidage and $Fo/\rho_p dp^3$ except for the rod-like particles. The initial fractional voidage of the rod-like particles is much greater than that of the irregular or spherical ones, which is explained in terms of the strong mechanical resistance caused by interlocking of particles due to the high elongation of the rod-like particles. In the dense packing the structural resistance is more pronounced than in the loose packing and, moreover, in the dense packing the effect of the compacting-force must be taken into account in the calculation of the interparticle cohesion. The rate of tapping-compaction suffers from little influence of the particle-shape.

The physical meanings of the correlations which have been found empirically between the angle of repose and ε_0 or ΔV could be elucidated to some extent through the relations between each of them and $Fo/\rho_p dp^3$.

Introduction

The packing state is one of the most simple and characteristic properties of a powdery material. Many trials have been made to determine the relations between the factors related to the packing state, such as the apparent density or the fractional voidage, and other properties of powder, the flowability[2] or the filling-weight of a capsule[3] for instances.

The primary characteristics governing the apparent density or the fractional voidage are the particle-shape, the particle-density, the particle-size, the size-distribution and the interparticle cohesion. There are some reports published on the relations between apparent densities and primary characteristics of powder.[4] No direct correlation, however, between an apparent density and an interparticle cohesion has been reported.

When powder is fallen from a constant height and filled into a vessel, or when powder-mass is tapped under constant conditions, the energy given to a particle in the powder-bed is proportional to the particle weight. Therefore, in these cases the driving force for the compaction of the powder-bed is proportional to $\rho_p dp^3$, where ρ_p and dp are the particle-density and the particle-diameter, respectively. On the other hand, the forces to resist against the compaction are mainly due to the interparticle cohesion and a structural factor such as interlocking of particles. Hence, the ratio of interparticle-cohesion/$\rho_p dp^3$ is considered to be one factor governing the apparent density or the fractional voidage. On the basis of these considerations, we studied on the influence of the particle-shape (which is irregular, rod-like or spherical) and the ratio of interparticle-cohesion/$\rho_p dp^3$ on the initial fractional voidage before tapping,

1) Location: *Hiromachi, Shinagawa-ku, Tokyo*.

2) *a*) M. Arakawa, T. Okada, and E. Suito, *Journal of the Society of Materials Science, Japan*, **14**, 764 (1965); *b*) E. Nakajima, *Yakugaku Zasshi*, **81**, 717 (1962); *c*) M. Aoki, S. Ogawa, S. Hayashi, and M. Hirayama, *Yakuzaigaku*, **27**, 18 (1968).
3) G. Reier, R. Cohn, S. Rock, and F. Wagenblast, *J. Pharm. Sci.*, **57**, 660 (1968).
4) H.D. Lewis and A. Goldman, *J. Am. Ceram. Soc.*, **40**, 323 (1966).

the rate of tapping-compaction, the final fractional voidage attained by tapping and the angle of repose. About ten kinds of powders of different sizes and shapes were used and magnesium stearate was used as a lubricant in some samples in order to change the interparticle cohesion.

Experimental

Materials——The preparation of the samples was the same as described in the previous paper.[5] The particle-diameter, -shape and -density for all the samples are shown in Table I. The average diameter, *dp*, is the Green diameter and was determined from the measurements for 500 particles with a microscope. The particle-density, ρ_p, was measured with a Beckman Air-Comparison Picnometer. The particle-shape which is irregular, rod-like or spherical was determined on the basis of Heywood's elongation (=Breadth/Length) and of the roundness, and defined as follows:

irregular particles: sharpcornered particles with the elongation less than 2.0
rod-like particles: particles with the elongation greater than 3.0
spherical (and nearly spherical) particles: round particles with the elongation less than 1.5

The elongations for the representative samples were, 1.6 for No. 6–sample, 1.7 for No. 8–sample, 4.0 for No. 10–sample, and 1.4 for No. 13–sample.

TABLE I. Materials tested

Sample No.	Substance	Particle size (dp) (10^{-4} cm)	Particle density (ρ_p) (g/cm^3)	Particle shape
1	lactose	12	1.53	irreg.
2	No. 1+Mg-St[a)]	12	1.53	irreg.
3	lactose, cryst., −100/+150	127	1.53	irreg.
4	lactose, cryst., −150/+200	90	1.53	irreg.
5	lactose, cryst., −200/+250	65	1.53	irreg.
6	lactose, cryst., −250/+300	55	1.53	irreg.
7	No. 6+Mg-St[a)]	55	1.53	irreg.
8	BTMP[b)]	50	1.44	irreg.
9	cryst. cellulose, −100/+150	127	1.55	rod-like
10	cryst. cellulose, −250/+300	55	1.55	rod-like
11	corn starch	13	1.50	nearly spherical
12	No. 11+Mg-St[a)]	13	1.50	nearly spherical
13	potato starch, −250/+300	55	1.50	nearly spherical
14	No. 13+Mg-St[a)]	55	1.50	nearly spherical
15	glass beads, −250/+300	50	2.42	spherical
16	VB_1HNO_3[c)]	64	1.10	spherical

a) The concentration of Mg-St (Magnesium Stearate) is 0.5%.
b) benzoyl thiamine monophosphate
c) spray-dryed with wax

Interparticle Cohesion, F_0——As described in the previous paper,[5] if interparticle distances were changed irreversibly or plastic deformations occurred with change of the contact-areas by pressing, the interparticle cohesion at point of contact, F_0, corresponding to that under zero pressing force, must be determined by extrapolating the plots of F against p to $p=0$, where F is an interparticle cohesion at point of contact when two particles are pressed by a force, p. F_0 was determined from the data of the shear-tests by use of the same apparatus and procedure as in the previous paper.[5]

Tapping——Powder was filled gently into a 100 ml Glass Cylindrical Graduate by the hollow-cylinder-method[5] and the apparent bulk density was determined. The initial fractional voidage, ε_0, was calculated from this density by using the particle density. The Graduate containing the powder was tapped at a rate of 120 times per minute and a falling-height of 1 cm. Hard rubber was attached on the supporting disk on which the cylinder was tapped. The apparent densities were determined at the following number of tappings; at each of 1, 3, 5, 10, 25, 50, 100, 200, 500, and 1000. The final fractional voidage, εf, was determined from the apparent density where the volume of the powder did not decrease any more by tappings. The measurements were done 5 times on a sample and the deviations of individual densities from the average were within ±5%.

5) K. Kurihara and I. Ichikawa, *Chem. Pharm. Bull.* (Tokyo), **21**, 394 (1973).

Angle of Repose, θ——The angle of repose was determined by a free-pile-method by use of the same apparatus as in the previous paper.[5] The angle was determined from an average of 5 runs on a sample.

Result

The equation proposed by Kuno,[6] eq. 1, was applied to the data of the tapping-compaction and the values of $-\log(\rho_p-\rho_n)$ were plotted against the number of tappings, n. As shown in Fig. 1, the straight line has a bend suggesting a change in the packing-mechanism. From the slope of the initial step, Kt was determined and shown in Table II together with Fo, εo and εf.

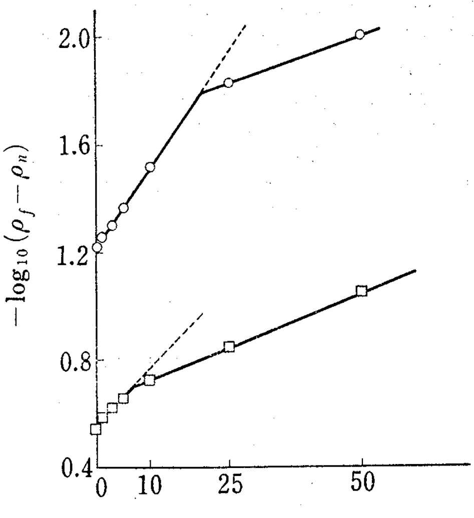

Fig. 1. Relation between $-\log_{10}(\rho_f-\rho_n)$ and Number of Tappings, n

—○—: No. 1 —□—: No. 16

Initial Fractional Voidage, εo

The ratio of interparticle-cohesion/particle-weight is considered to be one factor governing the fractional voidage, and another factor is a structural resistance which is dependent on the particle-shape. It is considered that in a loose packing the ratio of interparticle-cohesion/particle-weight is predominant, whereas in a dense packing the structural resistance is pronounced as well.

A good correlation was found between εo and $Fo/\rho_p dp^3$ for both the irregular particles and the spherical particles as shown in Fig. 2. The initial fractional voidage, εo, increases monotonically with $Fo/\rho_p dp^3$. On the other hand, εo's for the rod-like particles are much greater than those for the irregular or spherical particles. These findings suggest that even in the loose packing, where the structural resistance is considered to be comparatively small, the mechanical resistance caused by interlocking of particles will affect considerably the packing-structure when the particle has a highly elongated shape.

Table II. Interparticle Cohesion at Point of Contact, Fractional Voidage and Rate Constant of Tapping Compaction

Sample No.	Fo (g)	Dependency of F on p	ε_0 (—)	ε_f (—)	$Kt\times 10^2$ (—)
1	21×10^{-6}	dependent	0.655	0.465	3.8
2	17×10^{-6}	dependent	0.600	0.415	3.3
3	11×10^{-4}	independent	0.495	0.455	26
4	63×10^{-5}	independent	0.505	0.460	16
5	35×10^{-5}	independent	0.510	0.460	14
6	21×10^{-5}	dependent	0.555	0.450	11
7	19×10^{-5}	dependent	0.465	0.395	14
8	30×10^{-5}	independent	0.535	0.450	8.6
9	70×10^{-4}	dependent	0.830	0.800	9.3
10	13×10^{-4}	dependent	0.805	0.755	8.6
11	25×10^{-6}	independent	0.660	0.545	2.4
12	18×10^{-6}	independent	0.600	0.460	4.9
13	15×10^{-5}	dependent	0.485	0.385	8.5
14	98×10^{-6}	independent	0.395	0.305	9.7
15	0.0	dependent	0.420	0.395	8.4
16	14×10^{-4}	dependent	0.650	0.595	6.7

6) H. Kuno, *Proc. Faculty Eng. Keio Univ.*, **11**, 1 (1958).

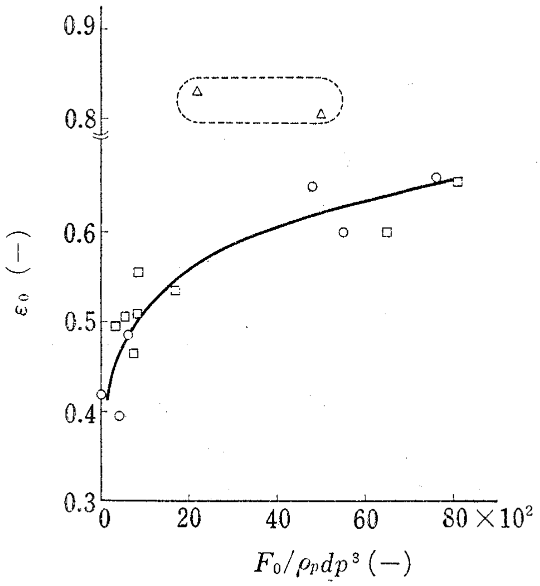

Fig. 2. Relation between Initial Fractional Voidage, ε_0, and $F_o/\rho_p d_p^3$

□: irregular particle
△: rod-like particle
○: spherical particle

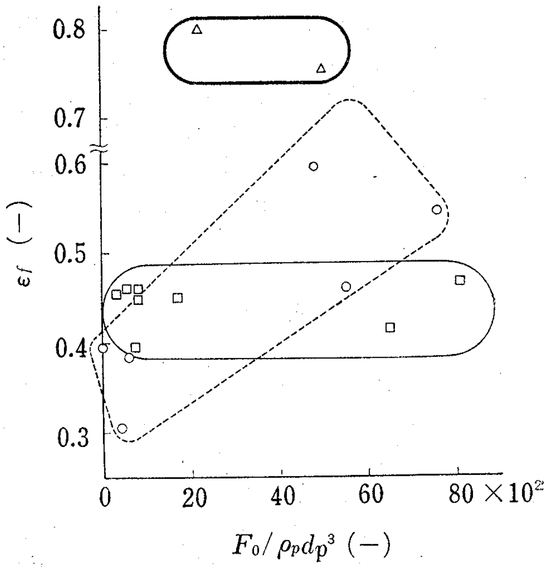

Fig. 3. Relation between Final Fractional Voidage attained by Tapping, ε_f, and $F_o/\rho_p d_p^3$

(□) : irregular particle
(△) : rod-like particle
(○) : spherical particle

Final Fractional Voidage, εf

The relations between εf and $Fo/\rho_p dp^3$ are shown in Fig. 3. These indicate that in the dense packing the structural resistance becomes pronounced as expected and the relation is dependent on the particle-shape which is irregular, rod-like or spherical. For the irregular particles, εf appears to be independent of $Fo/\rho_p dp^3$ and nearly constant, whereas for spherical particles εf seems to increase with $Fo/\rho_p dp^3$ though it is not so obvious as in the case of εo. The same discussion on the strong mechanical resistance as in εo applies to εf of the rod-like particles.

Rate Constant of Tapping Compaction, Kt

The change of the apparent density, ρ_n, with the number of tappings, n, is expressed as

$$\log(\rho_f-\rho_o)-\log(\rho_f-\rho_n)=K_t n \qquad \text{Eq. 1}$$

by Kuno.[6] This equation can be derived from a equation similar to the first order rate equation and hence Kt corresponds to the rate constant or the specific rate of the compaction. Eq. 1 can be rewritten with the fractional voidage by putting

$$\rho_n=\rho_p(1-\varepsilon n) \qquad \text{Eq. 2}$$

into eq. 1, and expressed as

$$\log(\varepsilon f-\varepsilon o)-\log(\varepsilon f-\varepsilon n)=K_t n \qquad \text{Eq. 3}$$

where εn is the fractional voidage at n tappings. Thus it can be seen that Kt is the rate constant for the change of the fractional voidage as well.

As shown in Fig. 1, Kt in the initial step is different from that in the later step. The relation between Kt in the later step and $Fo/\rho_p dp^3$ was examined but a good correlation was not found between them and Kt in the later step was roughly constant regardless of $Fo/\rho_p dp^3$. The relations between Kt in the initial step and $Fo/\rho_p dp^3$ are shown in Fig. 4. The constant Kt for the pherical particles is smaller than that for the irregular ones when $Fo/\rho_p dp^3$ is small, while the difference between them fades out as $Fo/\rho_p dp^3$ increases.

The initial or final fractional voidage for the rod-like particles is considerably higher than that for the irregular or spherical particles, as shown in Fig. 2 or 3, but in the case of *Kt*, the data of the rod-like particles are in an order similar to those of the irregular or spherical particles.

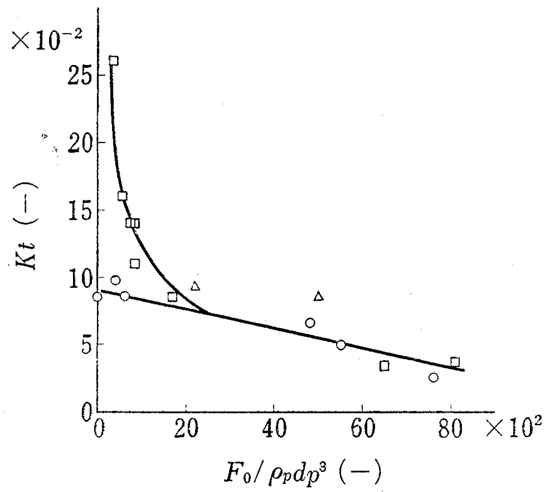

Fig. 4. Relation between *Kt* of Kuno's Equation and $Fo/\rho_p dp^3$

□: irregular particle
△: rod-like particle
○: spherical particle

Discussion

The relations between εf and $Fo/\rho_p dp^3$ depend on the particle-shape and this finding may be explained as follows. In the dense packing the structural resistance is a predominant factor governing the fractional voidage. The rod-like particles, in which the mechanical resistance caused by interlocking of particles is considerably strong, have much greater εf than that of the irregular or spherical particles. Although in the case of the irregular particles εf is smaller than that of the rod-like ones, a contribution of the mechanical resistance is still comparatively large and the influence of $Fo/\rho_p dp^3$ on εf is small. Not *Fo* but *F* should be used as the interparticle cohesion in the dense packing because under such conditions fairly high pressure acts at point of contact. An interparticle cohesion, *F*, between two particles under a compacting force, *p*, at point of contact, is represented by[5)]

$$F = Fo + \gamma_1 p \qquad \text{Eq. 4}$$

Although the absolute value of *p* caused by tappings can not be determined, the relative value may be determined on the basis of the assumption that *p* is proportional to $\rho_p dp^3$, *i.e.*,

$$p = \gamma_2 \rho_p d_p^3 \qquad \text{Eq. 5}$$

From eq. 4 and 5 it follows that

$$F/\rho_p dp^3 = Fo/\rho_p dp^3 + \gamma_1\gamma_2 \qquad \text{Eq. 6}$$

The interparticle cohesions, F_0 and *F*, determined in this experiment are the shear-forces necessary to separate two particles. Therefore, γ_1 is considered not only to consist of the contributions of plastic deformations or of the irreversible change of interparticle distances by pressing but also to be affected by the frictional mechanism. Table III shows γ_1 for the spherical particles. In the case of No. 12- and 14-sample, $F/\rho_p dp^3$ is equal to $Fo/\rho_p dp^3$, because these γ_1's are zero. No. 13-sample's εf has a value between εf's of No. 12- and 14-sample, and therefore in the plots of εf *vs.* $F/\rho_p dp^3$, if the point of No. 13-sample was plotted at an intermediate position between the points of No. 12- and 14-sample, an estimated value of 5×10^5 was obtained for γ_2. From this value of γ_2 and γ_1, $F/\rho_p dp^3$ for the other spherical samples was calculated by using eq. 6 and the relation between εf and $F/\rho_p dp^3$ for the spherical particles is shown in Fig. 5. This relation is similar to that between εo and $Fo/\rho_p dp^3$. The same discussion on the interparticle cohesion in the dense packing as in the case of the spherical particles applies to both cases of the irregular particles and of the rod-like ones. However, in these cases the influence of $Fo/\rho_p dp^3$ on εf is small and εf is nearly constant. Therefore, εf is considered to be still nearly constant regardless of $F/\rho_p dp^3$.

The rate-constant of tapping-compaction, *Kt*, is a dynamic characteristic of the powder-bed. *Kt* suffers from little influence of the particle-shape——this finding is considerably

Table III. The Slope of F vs. p Plot for Spherical Particle

Sample No.	γ_1 (−)
11	0.0
12	0.0
13	2.9×10^{-3}
14	0.0
15	2.4×10^{-3}
16	46×10^{-3}

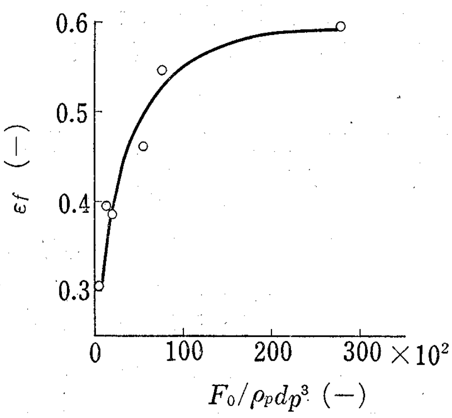

Fig. 5. Relation between εf and $F/\rho_p d_p^3$ ($=Fo/\rho_p d_p^3+5\times10^5\ \gamma_1$) for Spherical Particles

different from those of εo and εf. As reported previously,[5] the rate of discharge through orifices is also hardly affected by the particle-shape. The rate of discharge, as well as the rate of tapping-compaction, is a dynamic characteristic of powder. From these findings, it is considered that in dynamic states the influence of the particle-shape is smaller than in static states.

It has been reported that the equation proposed by Kawakita, eq. 7, which is not a rate-equation, applies well to tapping-data.[7]

$$n/r = 1/ab + n/a \qquad \text{Eq. 7}$$

$$r = (Vo - V)/Vo$$

Vo: initial volume of a sample

V: volume at n

The data in the present study were applied to eq. 7 and n/r was plotted against n in Fig. 6. As shown in Fig. 6, the data in the earlier parts of tapping deviated from a straight line for some samples. From the intercept and the slope of the straight line ,b was determined. As it has been reported that b is related to the interparticle cohesion,[2a)] b was plotted against Fo but no correlation was found. When it was plotted against $Fo/\rho_p dp^3$ (see Fig. 7), the rela-

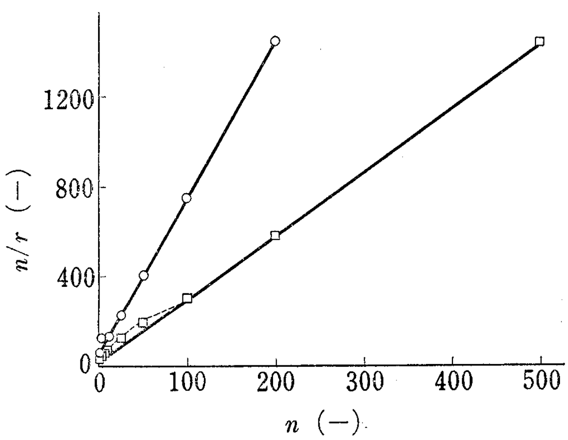

Fig. 6. An Example for Relation between n and n/r

□: No. 1 ○: No. 16

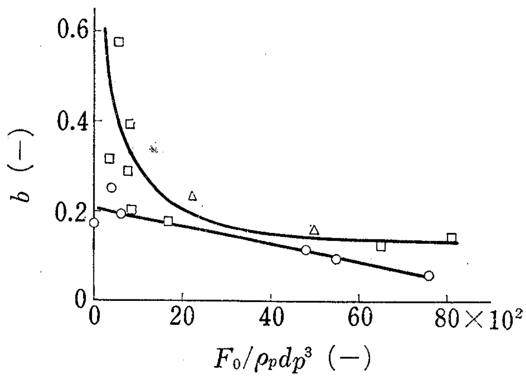

Fig. 7. Relation between b of Kawakita's Equation and $Fo/\rho_p d_p^3$

□: irregular particle
△: rod-like particle
○: spherical particle

7) T. Morioka, Y. Ikegami, and E. Nakajima, *Yakuzaigaku*, **15**, 119 (1959).

tion was found to be almost the same as that between Kt and $Fo/\rho_p dp^3$. This finding is explained on the basis of the fact that the driving force in tapping-compaction is proportional to $\rho_p dp^3$.

Arakawa, *et al.* have reported a correlation between εo and the angle of repose.[2a)] On the other hand, Nakajima[2b)] and Aoki, *et al.*[2c)] have reported correlations between ΔV (represented in eq. 8) and the angle of repose or the sliding angle.

$$\Delta V = (Vapp.i - Vapp.f)/Vapp.i \qquad \text{Eq. 8}$$

$Vapp.i$: apparent specific volume before tapping
$Vapp.f$: apparent specific volume after tapping

The plots of the angle of repose, θ, *vs.* $Fo/\rho_p dp^3$ in the present study are shown in Fig. 8, together with the solid lines obtained in the previous paper.[5)] From the relations shown in Fig. 2

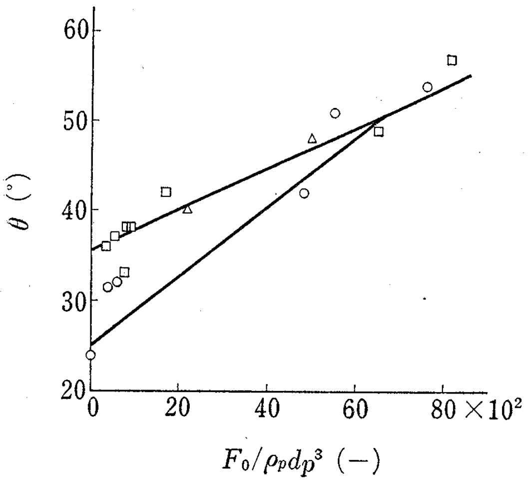

Fig. 8. Relation between Angle of Repose and $F_0/\rho_p d_p^3$

□: irregular particle
△: rod-like particle
○: spherical particle

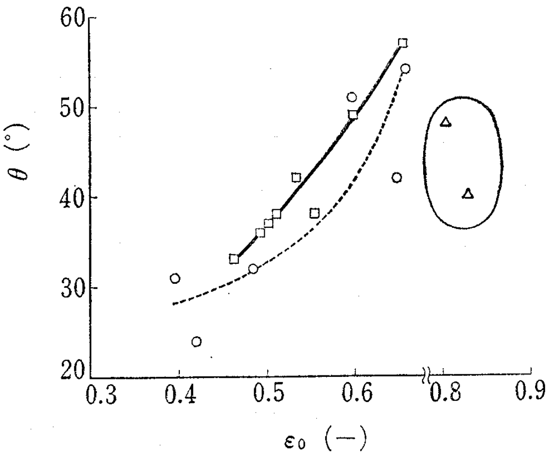

Fig. 9. Relation between Angle of Repose and ε_0

□: irregular particle
△: rod-like particle
○: spherical particle

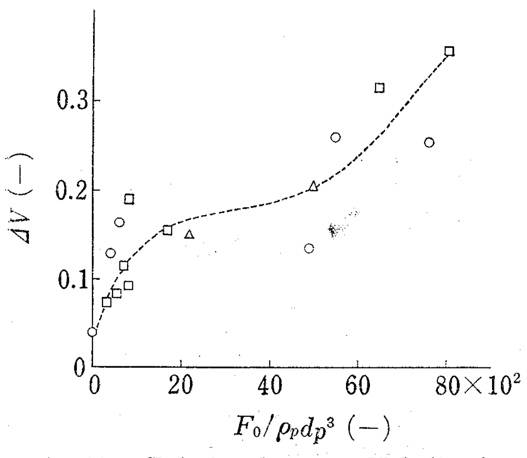

Fig. 10. Relation between Nakajima's ΔV and $F_0/\rho_p d_p^3$

□: irregular particle
△: rofd-like particle
○: spherical particle

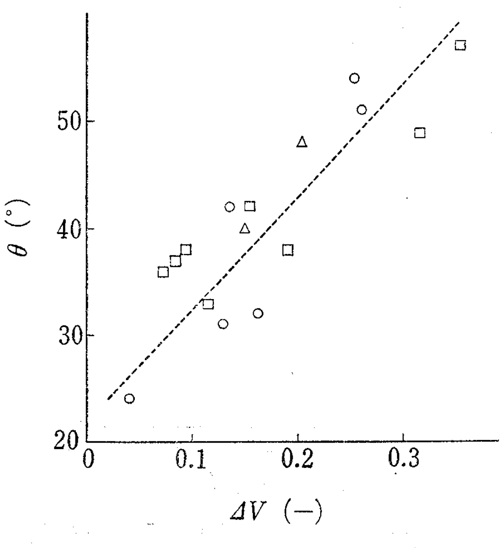

Fig. 11. Relation between Angle of Repose and Nakajima's ΔV

□: irregular particle
△: rod-like particle
○: spherical particle

(plots of εo *vs.* $Fo/\rho_p dp^3$) and in Fig. 8 (plots of θ *vs.* $Fo/\rho_p dp^3$), it is considered that θ is closely related to εo, though this relation depends on the particle-shape. The relation between εo and θ is shown in Fig. 9. Nakajima's ΔV is expressed by using the fractional voidage as

$$\Delta V = 1 - (1-\varepsilon o)/(1-\varepsilon f) \qquad \text{Eq. 9}$$

Both εo and εf suffers from the influence of the particle-shape though the dependence of the one on the particle-shape is different from that of the other. Therefore, the relation between ΔV and $Fo/\rho_p dp^3$ seems to be less dependent on the particle-shape than that between εo and $Fo/\rho_p dp^3$ because in ΔV the influence of the particle-shape is counterbalanced partially. The relation between ΔV and $Fo/\rho_p dp^3$ is shown in Fig. 10. Therefore, the relation between ΔV and θ shown in Fig. 11 is considered to be hardly affected by the particle-shape. On the basis of these results, it is considered that ΔV is suitable for the estimation of flowability of powder, while εo should be used in the investigation on flowability in connection with, for example, the particle-shape.

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