A Review of the Investigations into Powder Bed Mechanics Based on a Microscopic View in Japan

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1. Introduction

Mechanical behaviors of powder beds have been investigated from different viewpoints with various methods according to their individual objects. In the present paper notable results of works in such a field published in several academic journals in Japan will be reviewed especially from a viewpoint of a microscopic analysis based on individual single particles.

As is widely recognized, a number of investigations have been developed to elucidate the mechanical behaviors of powder beds essentially based on two different viewpoints; taking powder beds as a uniform contininuum and a discontinuum consisting of individual particles. Although these different approaches are popular to both fluid and solid mechanics, the case is slightly different with powder mechanics. It is because the mechanical behaviors of powder beds would depend on the specific properties of single particles which are constituent units not so small as those of the other continuums, i.e. molecules of the materials. This microscopic analysis of powder behaviors started nearly as early as the macroscopic analysis such as the measurement of a adhesive force^{5,15)}, but there remain a number of unsolved problems in both of them.

This review, from a viewpoint of combining individual single particles with behaviors of powder beds, will outline as follows:

- relationship between the stress on powder beds and the local force at a contact point
- mechanical behavior at a contact point
- structure of powder beds
- relationship between the mechanics at a contact point and that of powder beds

2. Relationship between stress and force

Taking each individual particle into consideration, the analysis of a mechanical behavior of powder beds makes a demand for the information of forces acting on interparticle contact points. Unfortunately it is impossible in general to measure these forces directly, thus, if possible, the determination of them by measuring the corresponding stresses would provide sufficient convenience in the analysis.

A typical expression was presented by Rumpf⁴⁷⁾ based on the theoretical concept which referred the tensile strength as follows:

$$\sigma_z = \frac{1 - \epsilon}{\pi} k \frac{H}{d_p^2} \tag{1}$$

where σ_z = tensile strength

 ϵ = porosity

k = average coordination number

H =cohesive force at a contact point

 d_n = particle diameter

Based on the early concept of Rumpf 45, 46), which involved a few mistakes in its derivation process, Molerus²⁵⁾ showed the stress – force relation might be expressed by Eq. (1) under the isotropic or hydrostatic pressure condition, and applied this conclusion to the analysis of shearing mechanism. On the other hand, Nagao²⁸⁾ derived a general expression of the stress - force relation assuming that a distribution of forces acting on interparticle contact points would be similar to that of stresses acting on powder beds. In addition Kanatani²⁰⁾ derived a similar expression of the correlation based on the principle of virtual displacement. These three expressions may have been considered to be essentially different since they are different in their forms.

The author et al. 58) originally found that both expressions derived by Nagao and Kanatani would agree completely with Rumpf's

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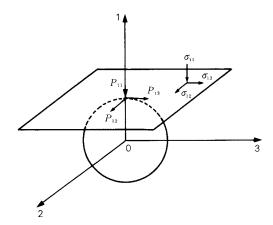


Fig. 1 Schematic representation of P_{ii} and σ_{ii}

equation. Consequently this can refer that the equation would be available in general to any expression of stress conditions. Therefore the stress acting on the plane vertical to *i*-axis, σ_{ij} can provide the *j*-axial component P_{ij} of the force acting on the contact point which is on the plane vertical to *i*-axis as shown in Fig.1⁵⁸, then Eq. (1) can be rewritten by:

$$\sigma_{ij} = \frac{1 - \epsilon}{\pi} k \frac{P_{ij}}{d_p^2} \tag{2}$$

Except the tensile test, very few applications of Rumpf's equation have been reported: Molerus²⁵⁾ made an analysis of the powder yield locus of cohesive powders and Nagao^{26,27,28,29,30,32,34,36)} derived a general theory on the powder mechanics, which will be referred later. On the other hand the author *et al.* ^{58,59)} established the way of analysis in which the effects of the porosity (the coordination number) on the powder layer mechanics and the force at a contact point separately discussed.

In general experimentally obtained results have been discussed using a relation of the porosity and the stress of powder beds. Eq. (2) can be rewritten assuming of $\pi \simeq k\epsilon$ as:

$$\sigma_{ij} = \frac{1 - \epsilon}{\epsilon} \frac{P_{ij}}{d_p^2} \tag{3}$$

It has been frequently shown that the relationship between a stress and a porosity in a consolidation test or between a tensile strength and a porosity could be a straight line on a semi-logarithmic paper. If the porosity ranges from 0.3 to 0.8, the term $(1 - \epsilon)/\epsilon$ in Eq. (3) can be modified using an exponential function

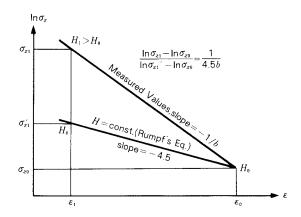


Fig. 2 Relationship between tensile strength σ and porosity ϵ

with an error of not over 10 percent;

$$\frac{1-\epsilon}{\epsilon} \simeq 10 \exp\left(-4.5\epsilon\right) \tag{4}$$

Thus Eq. (3) transforms to

$$\sigma_{ij} = 10 \exp(-4.5\epsilon) \frac{P_{ij}}{d_p^2}$$
 (5)

This equation is available to the analysis of tensile tests. A number of existing experimental data concerning them may correlate the tensile strength σ_z with the porosity ϵ by the equation of the form:

$$\sigma_z = k_1 \exp\left(-\frac{\epsilon}{h}\right)$$
 (6)

where k_1 denotes a constant. The $\sigma_z - \epsilon$ relation is expressed by a straight line on a semilogarithmic graph with a slope of -1/b as shown in Fig. 2. It is also shown in this figure that a tensile strength can have a straight relation to the porosity ϵ with a slope of -4.5, if a cohesive force is constant independently of a porosity. When the powder beds with the porosity of ϵ_0 and the cohesive force at a contact point H_0 are consolidated to the porosity of ϵ_1 , the tensile strength increases from σ_{z_0} to σ_{z_1} if the cohesive force H_0 is kept constant, as shown in Fig.2. It follows that this increase in the tensile strength can be expressed by nothing but the decrease in the porosity or the increase in the number of contact points. The ratio:

$$\frac{ln\sigma_{z_1} - ln\sigma_{z_0}}{ln\sigma_{z_1'} - ln\sigma_{z_0}} = \frac{1}{4.5b}$$
 (7)

thus provides the way of quantitative estimation in which the change of the tensile strength

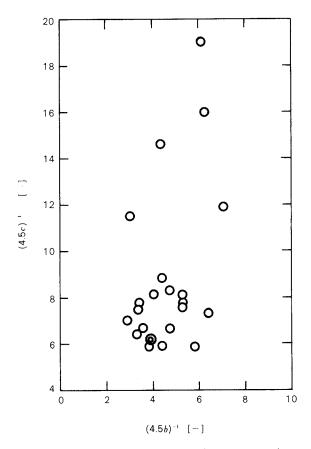


Fig. 3 Comparison of $(4.5c)^{-1}$ and $(4.5b)^{-1}$

due to the increase in the number of contact points and in the cohesive force at a contact point can be separately examined.

With respect to consolidation tests, a number of existing measurements may correlate the consolidation pressure p with the porosity ϵ using the following exponential function.

$$p = k_2 \exp\left(-\frac{\epsilon}{c}\right) \tag{8}$$

As is similar to the solution of Eq. (7), the

value $(4.5c)^{-1}$ refers how much the consolidation stress required to decrease the porosity is transmitted by increasing compaction force at a contact point.

Figure 3 shows the experimental results of the relationship between $(4.5b)^{-1}$ and $(4.5c)^{-1}$ which were obtained from 22 runs of tensile tests using adjusted porosity with pre-consolidation. Since it is remarkably seen that the value $(4.5b)^{-1}$ approximately ranges from 3 to 7 and the value $(4.5c)^{-1}$ from 6 to 19, it follows that the contribution of the porosity change is slight and the behavior will depend mainly on the force at a contact point.

3. Mechanical behavior at a contact point

As described above, the suitable method could be derived to obtain any force at a contact point. As the next step, here is discussed the mechanical behavior at a contact point resulted from such a force.

Most fundamental of the mechanical behavior at a contact point is a deformation of a particle. Nagao²⁶⁾, using grain model with surface roughness as shown in Fig.4, related the elastic and plastic deformation w with the maximum vertical stress P_{N0} at a contact point, and thus presented the following equation:

$$w = b_m P_{N_0}^{\frac{1}{m}} \tag{9}$$

The values of b_m and m for plastic deformation are indicated in **Table 1**. Nagao³⁶⁾ also derived the following expression which states a plastic recovery,

$$w_e = b_{em} P_{N_0}^{1 - \frac{1}{2m}} \tag{10}$$

The values b_{em} and m are also indicated in **Table 1**.

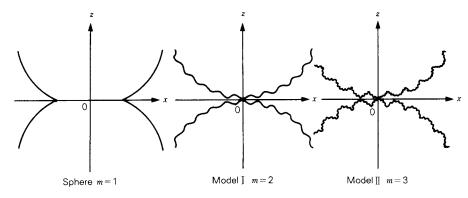


Fig. 4 Models of contact of grains

Table 1. The value of b_m , b_{em} and m for the interactions between the adjacent grains.

Shape of grain	m	b_m	b _{em}	Remarks
Plastic contact of spheres	1	$1/(2\pi p_m)$	$\frac{2(1-\nu^2)\sqrt{p_m}}{\sqrt{\pi} E}$	 P_m: Yield pressure. N: Number of (small) asperities per unit area. R: Mean radius of curvature of (small) asperities. N': Number of large asperities per unit area. R': Mean radius of curvature of large asperities. r: Mean radius of grains
Plastic contact of Model I (Spheres with asperities)	2	$\frac{1}{2r} \left(\frac{2r}{\pi^2 NRp_m}\right)^{1/2}$	$\frac{8(1-\nu^2)}{3\sqrt{\pi}E}\left(\frac{NRp_m}{2r}\right)^{1/4}$	
Plastic contact of Model II (Spheres with large asperities covered by small asperities.)	3	$\frac{1}{2r} \left(\frac{6r^6}{\pi^3 NN'RR'p_m} \right)^{1/2}$	$\frac{\frac{16(1-\nu^2)}{5\sqrt{\pi}E}(\frac{NN'RR'p_m}{6r^2})^{1/6}}{\frac{1}{6}}$	

As for the deformation or the flow behavior of powder beds, not only the deformation at a contact point but the friction there is of importance. Unfortunately, however, there have been few researches on the friction problems except that of Umeya^{11,62)} in which the friction factor between polystylene pellets and the plane was measured. Nagao²⁹⁾, making use of Eq. (2) by which the distribution of forces at contact points could be obtained, quantitatively determined the surface area of spheres which would cause a slip or not.

Since the cohesive force relatively increases with a decrease in particle size, observed phenomena would become complicated. The adhesive force of single particles have been frequently measured using vibration^{17,18,19)} or impact method^{17,18,19)} as well as classical methods of balance^{3,6,66)} or centrifugal force^{5,8,13,48)}. It is because the obtained results are valid for actual applications such as dust collection and surface contamination.

The measurement of the adhesive force of a single particle was found to be greatly dependent on the particle shape and the surface roughness, which was pointed out by Jimbo^{14,16,17)}. Jimbo et al. ⁵⁾ and Sano et al. ⁴⁸⁾ experimentally found that the adhesive force between glass sphere and smooth glass plane is almost ten times larger than that between crushed glass particle and smooth glass plane, and the former distribution would be considerably broad. Sano et al. ⁴⁸⁾ correlated the adhesive force to the shape factor defined by them. However in the case of discussing the shape factors from the viewpoint of particle separation mechanism, it should be necessary to pay

attention to the rotating moment around contact points, which Jimbo *et al.* pointed out in their reports^{18,19)}. Jimbo *el al.*^{18,19)} found that the separation force (adhesive force) of tangential direction is of an order to 1/10 of that of

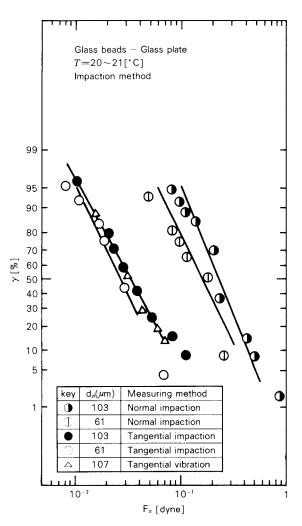
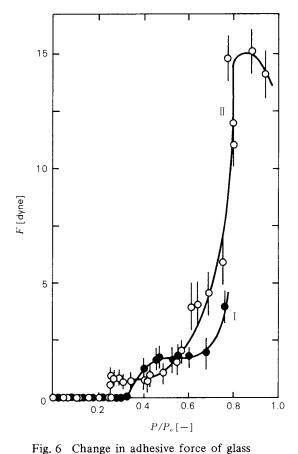


Fig. 5 The results of impaction separation method Comparison of normal and tangential separation



bead (GB503M) at a contact point
with water vapor pressure
I: Glass bead—commercial plate glass

II : Glass bead -plate glass specimen prepared from glass beads

vertical direction as shown in Fig. 5.

Atmospheric conditions also have a significant influence on the adhesive force of a single particle. The measurement of a single particle adhesive force is also dependent on atmospheric condition such as humidity and temperature. As for humidity, the influence of vapor pressure or relative humidity of water and methylalcohol was experimentally examined in detail by Chikazawa et al. 6,66). It is shown in Fig.66) that the adhesive force rapidly increased when the relative humidity exceeded 60 percent or so at which the absorbed water formed liquid surface on particles^{6,8,39} and it took a maximum value at about 80%, and it would decrease with increasing condensate water or p/p_0 Similar tendency was obtained by other studies^{5,8,13,48}.

As for temperature, on the other hand, the magnitude of a adhesive force was found to be greatly dependent on the kind of material.

Jimbo et al. 17,18,19) reported that the adhesive force of inorganic powder would increase with increasing temperature, with a minimum value at the temperature ranging from 100 to 150°C. As for organic powder, Danjo et al. $^{7)}$ found that the adhesive force would increase with increasing temperature below 0.9 T_m K (T_m : melting point) and rapidly decrease over such a temperature.

4. Structure of powder layers

The combination of the mechanics at a contact point with that of powder beds should require the investigation into the structure of powder beds. A number of researches have been performed using a powder layer model in which monodispersed sphere particles were packed with isotropical and random arrangement. However the analysis based on such a primary hypothesis as described above might be unsatisfactory to obtain the detail information on mechanics of powder layers, and thus it follows that quantitative description of powder bed structure should be required from the micromechanical viewpoint.

Makino et al. 21,22,23,51) tried to develop a description of powder bed structure with the help of statistic mechanics. They 21) regarded random packing beds indicated in Fig. 7 (a) as satistic powder beds illustrated in Fig. 7 (b), in which individual particles were overlapped one another. They also presented a new concept called a particle density original function 22, 23) which could provide the particle density distribution of powder beds depending on time and space. From the viewpoint that a particle arrangement would have a significant influence on powder mechanics, also they derived the probability distribution of the first layer in

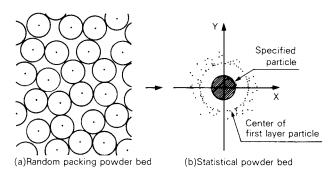


Fig. 7 Statistical representation of particle arrangement in powder bed

satistical powder beds⁵¹⁾ as shown in Fig. 7 (b).

A number of computer simulations for random packing beds have been performed to obtain radial distributions^{9,51)}, relationships between porosity and coordination number^{37,38,52,53,54)}, and other^{10,49)} which will be referred later. The porosity was related to the coordination number experimentally^{1,2)} and with model simulation^{42,44)}.

The bed structure of fine powders is more complicated due to the formation of secondary construction caused by agglomerated particles. It was experimentally found that effects of this secondary structure were considerable by Arakawa⁴⁾ and the author *et al.*⁵⁷⁾ using tensile tests, and by Hirota *et al.*¹²⁾ using shearing tests.

The change of powder bed structure is also important when the deformation or the flow behavior of powder bed is taken into account.

Makino et al.²¹⁾ described the deformation of packed particles using the first particle layer deformation in the statistical powder bed shown in Fig. 7 (b).

It is known that a shear deformation would destroy a isotropic structure of powder layers. This fact was ascertained experimentally by Umeya et al. ⁶⁴⁾ (Fig. 8) and by Matsuoka²⁴⁾ with a shear test on a two-dimensionally packed bed model with a photoelastic rod, and by

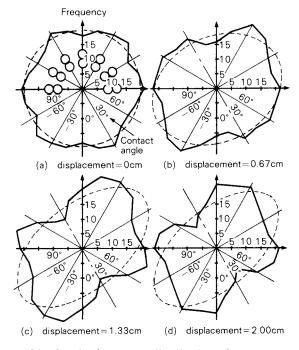


Fig. 8 The frequency distribution of contact angles (θ) (PVC rods)

Oda^{40,41)} with a two-dimensional visual observation of the yield behavior in a packed particles. Such a problem as an isotopy of powder bed structure is being discussed mainly in the field of soil mechanics.

It has been also reported that not only macroscopic but microscopic change of powder bed structure would have a significant influence on the rheological behavior such as stress-relaxation of powder beds.^{66,60,65)}

5. Relationship between mechanics at a contact point and that in powder beds

As above mentioned, works on mechanical behavior at contact points and structure of powder beds have been devoted to combine the macroscopic-microscopic relation of powder mechanics. Here it is shown to what extent the mechanics at contact points could form a connection with the mechanics of powder beds.

Nagao^{26,28,29,30,36)} and Makino *et al.*^{21,22,23,50)} presented the theoretical concepts which could be applied to more general problem of powder behavior.

Nagao, based on the stress-force relation²⁸⁾, Eq. (2) and deformation at contact points²⁶⁾, presented a theoretical relationship between stress and strain under the condition that; slip eixsts between individual contact particles^{29,30)}, it is negligible²⁸⁾, and consolidation pressure is released³⁶⁾. In the existence of slip motion²⁹⁾, the friction factor between individual particles was correlated with the internal friction factor in powder beds as illustrated in Fig.9, which implies the internal friction angle could not decrease to less than 30°. He also performed

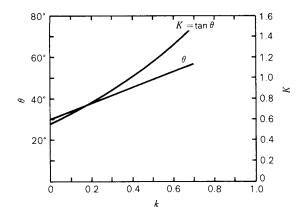


Fig. 9 Relationship between the macroscopic internal friction coefficient K and the friction coefficient between the adjacent grains k.

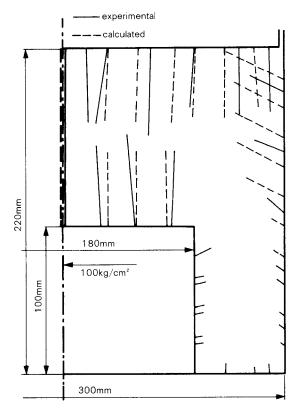


Fig. 10 Resultant stress distributions on the surface of the box (Silica sands are compressed under the mean pressure of 100kg/cm² on the lower surface of punch)

various consolidation tests^{27,31,32,33,34,35)} and ascertained his theories by experiments^{27,34)}. The simulation of the finite-element approach based on the concept developed by Nagao could provide a stress distribution on the box surface in good agreement with measured results as illustrated in Fig.10³⁴⁾. Although Nagao's concept made a frontal attack on the powder mechanics with rigorous logic, the problems of cohesive forces at constant points and a slip mechanism have not been made clear yet.

Makino et al. presented a theoretical model of powder bed yielding based on the following assumptions. One is the yield condition of constant displacement that powder beds yield when the specified particle reaches a point on the circular with a center of S(0, s) and a radius of d/2 as shown in Fig.11²¹⁾. The other is the Lennard-Jones typed potential energy which provides an interparticle force. They found the yield condition of constant displacement to be adequate using tests on consolida-

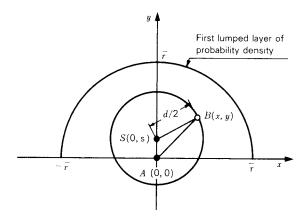


Fig. 11 Yield condition of constant displacement

tion, shear, and tensile strength, and estimated the PYL obtained from his concept as illustrated in Fig. 12. Although the model made by Makino et al. 50) can be applied to any yield phenomenon of powder beds, it seldom seems to provide a sufficient information of the relationship between such a mechanism and the powder or particle properties which have been referred by a number of researchers. It is because the particle behavior at contact points and the strain of powder beds were daringly expressed using the potential function and the concept of statistical powder beds.

There have been several researches on individual powder bed behaviors. The author *et al.*⁵⁹⁾ found by a semi-theoretical method that a compressive force would have a dominant effect on a tensile strength. Takahashi *et al.*⁵⁵⁾ also made a similar point on the strength of a granule. Suzuki *et al.*⁴⁹⁾ made a computer simulation for obtaining the relationship between a tensile strength and a porosity in the beds packed with spheres.

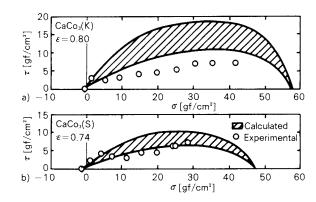


Fig. 12 Comparison between calculated yield loci and experimental results

It was shown by Gotoh et al. 10) using computer simulation that the unisotropical construction of the beds packed with spheres would result in the bottom weight distribution distorted to M shape. Danjo et al. indicated that a porosity of a powder bed packed by means of a centrifugal⁴³⁾ or an impact⁷⁾ force could be a function of the ratio of a force applied to a single particle to a cohesive force at a contact point. Basing his concept largely on the deformation at a contact point, Umeya et al. 63) stated that PYL shape as well as a consolidation characteristic (the relationship between a porosity and a consolidation stress) might change at a certain consolidation stress. Umeya et al. investigated the qualitative relationship between stress and strain in two-dimensional model of packed beds using a straw⁶¹⁾. They⁶⁴⁾ also drew the conclusion that in a shearing twodimensionally arrayed bed model with photoelastic rods, stress values calculated from a force at a contact point would tend to agree generally with measured results.

6. Conclusion

The present paper reviewed recent researches in Japan which microscopically investigated the mechanics of powder beds. Most of them have confined themselves chiefly to a microscopic analysis for specific objects. On the other hand Nagao and Makino *et al.* derived theoretical concepts relevant to general problems in powder mechanics which are highly estimated as a precursory investigation, though they have a few problems to be solved. The experimental researches by Umeya *et al.* are also valid because they have systematically analyzed the microscopic mechanics including rheological behaviors.

In order to develop the mechanics based on a microscopic view to more raised level, a model of powder bed structure should be required which is able to describe the deformation and/ or the flow of a powder bed as the structure change. Investigations into beds structure have been actively performed in the field of soil mechanics as introduced partially in the present review.

It may be also essential to establish the appropriate measuring methods for mechanical or surface characteristics based on geometry and physical-chemistry of single particles. In par-

ticular fundamental works on friction should be required because their reports have been very few.

In this review the author tried to introduce as many studies performed in Japan as possible. Although this review involves the articles printed mainly in the field of powder technology, the author considers there are some other outstanding researches especially in the fields of pharmaceutics, ceramics, and soil mechanics. As for soil mechanics, essential reports of powder bed structure have been printed in the English journal 'Soils and Foundation' published by The Japan Society of Soils Mechanics and Foundation Engineering.

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References

- 1) Arakawa, M.: Journal of the Society of the Material Science, Japan, 16, 319 (1967).
- 2) Arakawa, M. and M. Nishino: ibid, 22, 658 (1973).
- 3) Arakawa, M. and S. Yasuda: ibid, 26, 858 (1977).
- 4) Arakawa, M.: ibid, 29, 881 (1980).
- 5) Asakawa, S. and G. Jimbo: *ibid*, **16**, 358 (1967).
- 6) Chikazawa, M., W. Nakajima and T. Kanazawa: Journal of the Research Association of Powder Technology, Japan, 14, 18 (1977).
- Danjo, K. and A. Otsuka: Yakugaku Zasshi, 100, 893 (1980).
- 8) Emi, H., S. Endo, C. Kanaoka and S. Kawai: Kagaku Kogaku Ronbunshu, 3, 580 (1977).
- 9) Gotoh, K. and T. Chiba: Journal of the Society of Powder Technology, Japan, 16, 179 (1979).
- 10) Gotoh, K., T. Chiba and A. Suzuki: Kagaku Kogaku Ronbunshu, 6, 267 (1980).
- 11) Hara, R., H. Suzuki and K. Umeya: Journal of the Society of the Material Science, Japan, 30, 867 (1981).
- 12) Hirota, M., Y. Iwamoto, T. Kobayashi and T. Oshima: Journal of the Society of Powder Technology, Japan, 21, 69 (1984).
- 13) Jimbo, G. and S. Asakawa: Journal of the Research Association of Powder Technology, Japan, special issue, 6 (1966).
- 14) Jimbo, G.: Journal of the Society of the Material Science, Japan, 16, 291 (1967).
- 15) Jimbo, G. and S. Asakawa and N. Soga: *ibid*, 17, 177 (1968).
- 16) Jimbo, G.: Kemikaru Enjiniyaringu, 13, 631

- (1968).
- 17) Jimbo, G. and R. Yamazaki: Preprints of European Symposium Particle Technology, B, 1064 (1980 Amsterdam).
- 18) Jimbo, G., R. Yamazaki and G. Hong.: Semi-Annual Report of the Asahi Glass Foundation for the Contribution to Industrial Technology, 38, 123 (1981).
- 19) Jimbo, G. and R. Yamazaki: KONA, 1, 40 (1983).
- 20) Kanatani K.: Journal of the Society of Powder Technology, Japan, 17, 504 (1980).
- 21) Makino, K., K. Saiwai, M. Suzuki, T. Tamamura and K. Iinoya: Kagaku Kogaku Ronbunshu, 4, 439 (1978).
- 22) Makino, K.: Journal of the Japan Society of Powder and Powder Metallurgy, 28, 269 (1981).
- 23) Makino, K.: ibid, 28, 276 (1981).
- 24) Matsuoka, H.: Soils and Foundations, 41, 29 (1974a).
- 25) Molerus, O.: Powder Technology, 12, 259 (1975).
- 26) Nagao, T.: Transactions of the Japan Society of Mechanical Engineers, 33, 229 (1967) or Bulletin of JSME, 10, 775 (1967).
- 27) Nagao, T.: ibid, 34, 100 (1968).
- 28) Nagao, T.: *ibid*, **43**, 4038 (1977) or *Bulletin of JSME*, **21**, 1077 (1978).
- 29) Nagao, T.: ibid, 44, 1912 (1978) or Bulletin of JSME, 22, 148 (1979).
- 30) Nagao, T.: *ibid*, **44**, 2967 (1978) or *Bulletin of JSME*, **22**, 636 (1979).
- 31) Nagao, T., K. Chijiiwa, Y. Hatamura, K. Takeuchi and E. Shinohara: *Journal of the Society of Powder Technology*, **15**, 643 (1978).
- 32) Nagao, T., K. Chijiiwa, Y. Hatamura, K. Takeuchi, N. Nakashima and H. Okamoto: *ibid*, **16**, 460 (1979).
- 33) Nagao, T., K. Chijiiwa, Y. Hatamura, K. Takeuchi and E. Shinohara: *ibid*, **16**, 625 (1979).
- 34) Nagao, T. and S. Katayama: *Transactions of the Japan Society of Mechanical Engineers*, **46**, 355 (1980) or *Bulletin of JSME*, **23**, 1763 (1980).
- 35) Nagao, T., K. Chijiiwa, Y. Hatamura, K. Takeuchi, N. Nakashima and H. Okamoto: Journal of the Society of Powder Technology, Japan, 17, 10 (1980).
- 36) Nagao, T.: Transactions of the Japan Society of Mechanical Engineers, 47, 1363 (1981).
- 37) Nakagaki, M. and H. Sunada: Yakugaku Zasshi, 83, 73 (1963).
- 38) Nakagaki, M. and H. Sunada: ibid, 88, 651 (1968).
- 39) Nishino, M. and M. Arakawa: Journal of the Society of the Material Science, Japan, 22, 663 (1973).
- 40) Oda, M.: Soils and Foundations, 12, 1 (1972).
- 41) Oda, M. and J. Konishi: ibid, 14, 25 (1974).

- 42) Okazaki, M., T. Yamazaki, S. Gotoh and R. Toei: Journal of Chemical Engineering of Japan, 14, 1983 (1981).
- 43) Otsuka, A., H. Sunada and K. Danjo: Yakugaku Zasshi, 89, 1013 (1969).
- 44) Ouchiyama, N. and T. Tanaka: *Industrial and Engineering Chemistry, Fundamentals*, 19, 338 (1980).
- 45) Pietsch, W. and H. Rumpf: Chemie Ingenieur Technik, 39, 885 (1967).
- 46) Rumpf, H.: ibid, 30, 144 (1958).
- 47) Rumpf, H.: ibid, 42, 538 (1970).
- 48) Sano, S., F. Saito and S. Yashima: Kagaku Kogaku Ronbunshu, 10, 17 (1984).
- 49) Suzuki, A. and K. Gotoh: ibid, 7, 628 (1981).
- 50) Suzuki, M., K. Makino and K. Saiwai, H. Yamamoto and K. Iinoya: *ibid*, **5**, 481 (1979).
- 51) Suzuki, M., K. Makino, T. Tamamura and K. Iinoya: *ibid*, **5**, 616 (1979).
- 52) Suzuki, M., K. Makino, M. Yamada and K. Iinoya: *ibid*, **6**, 58 (1980).
- 53) Suzuki, M. and T. Oshima: *Powder Technology*, **35**, 159 (1983).
- 54) Suzuki, M. and T. Oshima: *ibid*, **36**, 181 (1983).
- 55) Takahashi, M., T. Kobayashi and S. Suzuki: Annual Report of Ceramic Engineering Research Laboratory (Nagoya Institute of Technology) 8, 25 (1981).
- 56) Tanno, K., S. Kikuta and Y. Matsuda: *Journal of the Society of Powder Technology, Japan*, 18, 887 (1981).
- 57) Tsubaki, J., K. Kato and G. Jimbo: *ibid*, **18**, 873 (1981).
- 58) Tsubaki, J.: ibid, 21, 30 (1984).
- 59) Tsubaki, J. and G. Jimbo: *Powder Technology*, 37, 219 (1984).
- 60) Tsubaki, J., G. Jimbo and K. Takagi: Proceedings of 3. European Symposium on Particle Characterization (1984 Nürnberg).
- 61) Umeya, K., N. Kitamori, R. Hara and T. Yoshida: Journal of the Society of the Material Science, Japan, 18, 489 (1969).
- 62) Umeya, K., T. Isoda, R. Hara and J. Kikuta: *ibid*, 23, 549 (1974).
- 63) Umeya, K., T. Isoda, R. Hara and Y. Kato: *ibid*, **22**, 637 (1973).
- 64) Umeya, K., R. Hara: Journal of Chemical Engineering of Japan, 8, 56 (1975).
- 65) Umeya, K., R. Hara and T. Kimura: Journal of the Society of the Material Science, Japan, 25, 623 (1976).
- 66) Yamaguchi, T., M. Tomita, M. Chikazawa and T. Kanazawa: *Journal of the Society of Powder Technology, Japan*, 16, 514 (1979).