Cohesion of Particulate Solids. VII.[1] Influence of Particle Size on Compression by Tapping

Eihei FUKUOKA and Shintaro KIMURA*,[2]

Toho University School of Pharmaceutical Sciences, 2-1-1 Miyama, Funabashi-shi, Chiba 274, Japan. *Received September 26, 1991*

An experimental theorem (1) is proposed for conversion of the curve representing powder bed compression by tapping into a straight line. The theorem was found to apply to a variety of particulate solids:

$$\varepsilon_e = -b \log a + k \qquad (1)$$

where ε_e is the porosity of the powder bed at a stable state for specific acceleration levels, a is the acceleration caused by tapping at constant height differences (Δh), and b and k are constants.

If the speed of pouring the powder bed with the sample was high, two continuous straight lines (lines 1 and 2) were obtained. Line 1 was thought to represent the porosity primarily due to the interaction between the particles and the walls of the container, in addition to the porosity at a stable state for the mostly loosely filled sample. Line 2 was considered to represent the maximum porosity under a constant acceleration when there is no interaction between the container and sample.

The two lines could be approximated by line 3 at low filling speeds. This straight line is believed to represent the porosity for the most dense packing under a constant acceleration.

The three straight lines tend to converge into a single line as the particle size is reduced. A correlation was also found between the slope of line 3 and the particle size.

Keywords powder; granules; compaction; tapping; experimental theorem; particle size; loose packing; dense packing; porosity; filling rate

The characteristics of compression of particulate solids by tapping differ from those exhibited by piston compression, especially in the fact that the particle itself, excluding particularly brittle types, is not usually subjected to pressure strong enough to cause it to be deformed. In theory, the applied pressure distribution from tapping is not concentrated in any one area. This observation is particularly important for forecasting phenomena likely to occur during transportation and storage since it eliminates the need for considering breakage and deformation of particles.

A similar method, centrifugal compression,[3] is also available, however, it has been pointed out that the acceleration of particles in this method varies with the distance from the center of rotation.

If it were possible to quantify the compression caused by tapping, it would be possible to forecast the compression present during daily handling of powders and during the compounding process at pharmaceutical production facilities. This could lead to a method of quantifying the characteristics found in loosely compacted pharmaceutical powders under bulk storage conditions.

The authors conducted tests, using methods proposed by Jimbo *et al.*[4] to quantify compression of pharmaceutical materials by tapping, and suggest the possibility of expressing quantitative characteristics of powdered pharmaceuticals in bulk storage.[5]

Conventionally, compression due to tapping has been expressed by tap count n, which was based on the kinetic method.[6] The method proposed by the authors eliminates the kinetic factor.

To the best of our knowledge, there have been no studies on the characteristics of individual particles or high order characteristics, *i.e.*, tapping compaction characteristics.

This study suggests an equation to convert the tapping compression curve into a straight line. It also presents results obtained by the authors' experimental theorem for evaluating the low order characteristics of powdered materials or the correlation between particle size and tapping compression.

Experimental

Methods Testing equipment is illustrated in Fig. 1.

As in our previous work,[5] a commercially available tapping device was used for this experiment. The vibration pickup, a barium titanate lead ceramic sensor which measures the acceleration at the time of collision, was fixed with adhesive to the mounting facility for a graduated glass cylinder (100 ml in capacity) (hereafter referred to as cylinder). The vibration signal was amplified with a vibration meter (Lion Co., Ltd.) and the maximum value was read from an electromagnetic oscilloscope.

Increasing the tapping impact caused the surface of the sample within the cylinder to become irregular and/or sloped, resulting in increased difficulty and decreased accuracy in measuring the apparent volume of the particulate solid.

To overcome this problem, a suppression plate, a very light (approximately 1 g) circular acrylic disk connected to an acrylic rod (Fig. 1)

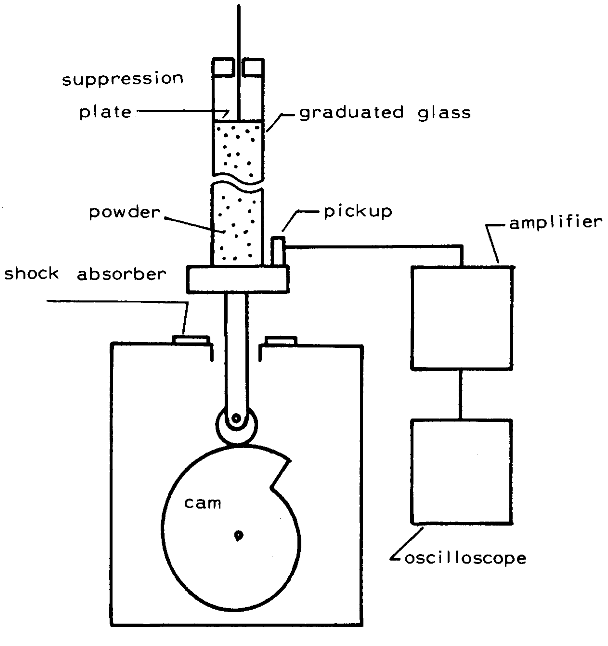

Fig. 1. Tapping Apparatus

TABLE I. Feret's Diameter (μm) of the Powder Particles

Mesh number of the screens	GP[a)]	Sulfisoxazole	Borax	Lactose
20— 28	—	—	950	—
28— 36	700	700	670	—
36— 48	565	570	470	—
48— 70	—	450	350	—
70—100	255	200	190	220
100—145			135	150
145—170			100	110
—				—
270—325				60

a) Glass powder.

was inserted through a small hole drilled in the lid of the cylinder and allowed to rest on the surface of the particulate solid sample. This slight modification allowed accurate measurement of nominal volume. As long as the weight of the plate was less than 1/100 of the gross weight of the sample, it was assumed to have a negligible effect on compression of the powder.

A method similar to that previously reported[5)] was used in this study. An optimally charged and loosely filled sample was subjected to repeated tapping at constant height difference Δh and a constant acceleration a_i. The porosity of the stable state ε_{ei} with the acceleration of a_i was measured.

The stable state, (stable mechanical state), is the state at which the sample powder layer ceases to exhibit changes in its apparent volume at the desired acceleration.

Although the stable condition was normally reached at a tapping count of approximately 200, the count was increased to 750—1000 for this study.

Δh was increased gradually and the porosity ε_{ei} measured at the stated levels of acceleration a_i after the stable state was achieved. This allowed the authors to clarify the correlation between acceleration a and porosity ε_e at the stable state.

A relationship is normally expressed in terms of the correlation between acceleration and porosity, however, the method employed here allows the following interpretations: assuming the mass of the sample to be m, the force f applied to the entire particle layer at a specific acceleration level a can be expressed by, $f=ma$, if the friction between the cylinder wall and the particle can be ignored.

Assuming the cross sectional area of the cylinder to be A, and compression pressure to be p, the relationship, $p=f/A$, is established. This is equivalent to piston pressure and allows direct comparison whenever necessary.

With the tapping count sufficiently large as described above, the assumption will be made that friction between the cylinder wall and the particle of the sample could be ignored.

Unlike the earlier paper,[5)] this entire experiment was carried out in an environmentally controlled chamber (Tabai Co., Ltd.) in which the temperature was maintained at 20 °C and relative humidity at 50%.

Samples Both organic and inorganic pharmaceuticals and inorganic materials, about 90—100 ml in apparent volume, were used as test samples; these included potato starch, lactose, sulfisoxasol, microcrystalline cellulose (MCC), glass powder (GP) and borax. In all cases, the materials were sifted through a standard sieve and divided into several fractions to be used as samples. Talbe I shows the average particle size of the fractions as measured through an optical microscope.

Results and Discussion

Tapping Compression Equation Figure 2 shows a typical curve representing the relationship obtained by plotting the maximum tapping acceleration a (hereafter referred to as tapping acceleration) and the porosity at stable state ε_e.

Potato starch was used as the sample and was redispersed using a 200 mesh standard sieve after compaction. Tapping was repeated ten times using the same sample.

Since the possibility exists that the particles may be defaced or deformed by repeated compression and sifting, the identical operations were repeated using another sample.

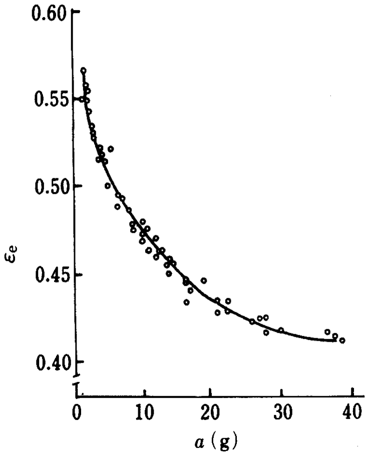

Fig. 2. Relation between ε_e and Acceleration (a) by Tapping

Potato starch at R.H. 50%.

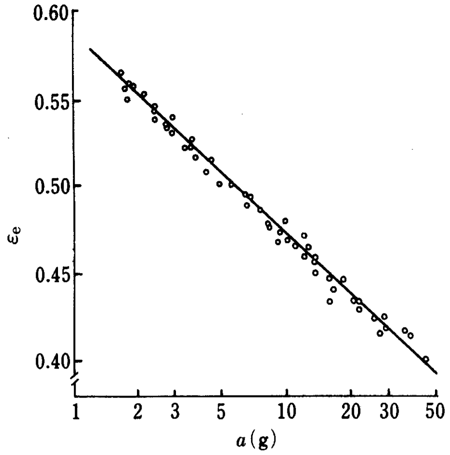

Fig. 3. Linear Relation between ε_e and Acceleration by Tapping (a) on Semi-log Scale

Potato starch at R.H. 50%.

Results were plotted on the same graph and no significant differences were found between the two samples.

The plotting of the graphs started from "1 g", the gravitational acceleration field of the earth.

Figure 2 reveals a great deal of porosity for the most loosely filled potato starch sample at 1 g. It also shows a considerable reduction in bulk volume resulting from tapping shocks and gives rise to a uniquely characteristic gradual curve represented as ε_e *vs.* a.

These tapping compression characteristics indicate that potato starch exhibits relatively strong cohesive properties. Moreover, since the particle shape is simple and the particles have a smooth surface, dense compacting occurs readily under high acceleration.

Several linear equations[7)] have been proposed to describe the phenomena observed during piston compression of particles. On the other hand, no simple equations showing relationships for tapping compression between acceleration or compression pressure and apparent volume or porosity have yet been proposed.

Figure 3 was obtained by plotting tapping acceleration a, on the horizontal axis of a logarithmic scale and the porosity of stable state ε_e, on the vertical axis. This produced a regression line which proves the experimental Eq. 1:

$$\varepsilon_e = -b \log a + k \qquad (1)$$

where b and k are constants.

Although two or three exceptions were recognized, Eq. 1 was found to apply to a very large number of pharmaceutical powders. Here, we interpret the results of the experiment using Eq. 1 and we study the slope of the straight line and significance of intercepts from the physical characteristics standpoint.

Effects of the Pouring Speed of Initial Powder Bed on Tapping Compression It was earlier pointed out[8)] that bulk density varies with the pouring speed of powder into a cylinder, therefore the relationship between compression by tapping and the pouring speed for the initial powder bed was evaluated.

Using 674 μm borax as a sample, the initially filled powder bed, (hereafter called initial powder bed), was formed at six different pouring speeds ranging from 100 g/600 s to 100 g/s. Figure 4 is the semi-log plot of ε_e *vs.* a at the minimum and maximum pouring speeds.

Figure 5 shows only the regression line of tapping compression for intermediate pouring speeds.

It is evident from Fig. 4 that the lowest straight line represents the pouring at the minimum speed. At higher speeds, the lines seem to break down into two straight lines with an intersecting point at approximately, $a = 7$ g.

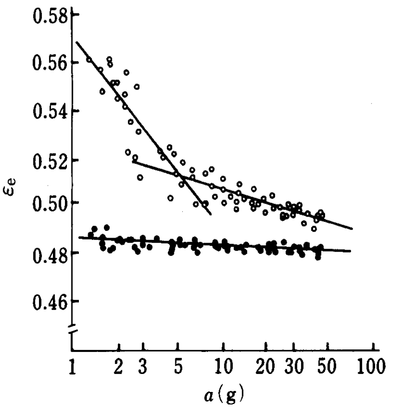

Fig. 4. Scatter Diagram of ε_e *vs.* a for Initial Powder Bed Prepared with Maximum Pouring Rate (○) and Prepared with Minimum Pouring Rate (●)

Sample, borax (670 μm).

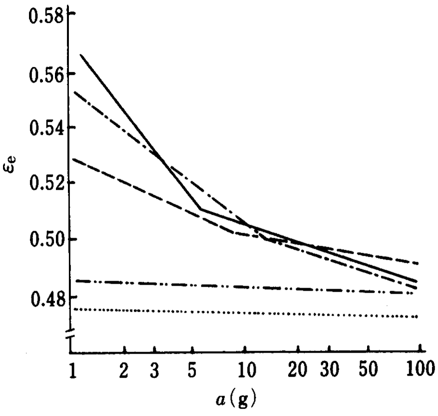

Fig. 5. Influence of Pouring Rate at the Preparation of Initial Powder Bed on Regression Curves of ε_e *vs.* a Plot on Semi-log Scale

Sample, borax (670 μm). ——, 100 g/s; —·—, 100 g/16 s; — —, 100 g/80 s; ——··—, 100 g/225 s; ------, 100 g/400 s and 100 g/600 s.

In the following descriptions, line 1 is the highest line and represents the highest pouring speed. Line 2 is the next line and has a relatively gradual slope, while line 3 represents the slowest pouring speed.

As the pouring speed of the initial powder bed decreases, the slope of both lines 1 and 2 (Fig. 5) become less steep. A change is noted in that the slope of line 1 gradually approaches that of line 2 and the two lines merge to appear as a single straight line.

With further reduction of the pouring speed, a slight change in the intercept with the vertical axis occurs but the slope and the intercepts finally become uniform.

These observations seem to indicate that the tapping compression curve for a powder bed is unlike that gained by compressing particles by piston and that there is no direct correlation between the stress and the state of the sample. This means that compression behavior of particulate solids due to tapping can very widely depending on the pouring speed of the initial sample. In other words, if the progress of compression due to tapping in a particular sample can be represented by a group of straight lines which vary with the pouring condition of the initial powder bed and which are spread over a relatively broad area, these lines can be represented by the three straight lines, lines 1 to 3.

During the compression process, particles interchange positions due to steric obstacles posed by other particles. Moreover, a change in the orientation of particles is impeded. It is believed that this phenomenon increases as particle compression progresses. Since the characteristics of the initial layer are maintained somewhat as compression progresses, it appears that compression behavior depends greatly on the state of the initial powder bed.

Line 1 is one of the limiting compression lines. When filling occurs at high speed, even the slightest shock causes the very unstable state to collapse and the volume of the initial layer to decrease dramatically until it reaches a semi-stable state.

In such case, mutual interaction between the particles and the cylinder wall depends on the diameter of the cylinder. This is particularly true when the particle size is relatively large. This paper, however, does not take the cylinder diameter into account. As explained later, by maintaining the cylinder diameter constant and varying the particle size, the relative ratio between the two is altered and supporting evidence obtained qualitatively.

Line 1 represents the most loosely filled initial powder bed which can be obtained in normal gravity with the mutual interaction between the particles and the cylinder wall taken into account. In such an experiment, the straight line is reproducible as long as the same cylinder is used. The value obtained can be useful for discussing the interaction between the particles and the cylinder wall.

Another line, line 2, represents the porosity, unaffected by the container wall interaction, at the most loosely packed state for a given acceleration. This is expressed as a function of the acceleration to which the sample is subjected. The value of the intercept with the vertical axis represents the porosity of the most loosely packed layer in the absence of interaction with the cylinder wall and under normal gravitational force. Theoretically, this value cannot be obtained without using a cylinder of infinite size.

Many researchers[8)] have defined or are trying to create the most loosely packed states using uniform size model particles. Little research, however, has been done on the most loosely packed states of powders under normal gravitational field.

Line 2 demonstrates excellent reproducibility. From the standpoint of physical properties, it can also be considered to be an important representation of the most basic particle characteristics.

Line 3 represents the lower limit and most densely packed state (in which the particle is considered to be in the most stable position) under given acceleration.

Thus, it appears that the most densely packed state can also be defined as a function of acceleration of the environment into which the sample is placed. Furthermore, the group of lines falling between the three lines describes the compression process of the initial layer at various filling speeds. There seems to be no correlation between these lines and other physical characteristics.

From the above observations, it can be discerned that the generally used descriptions "most densely packed" and "most loosely packed" are very vague. To accurately define these states, it is also necessary to indicate the acceleration force at which the particle layer reaches stability.

These findings suggest that when certain methods of filling are employed, it may be difficult to increase the density of the filling at a later date. This would explain the phenomenon observed when a quantity of powder is removed from a container but will not then fit back into the same container. Such information is useful in the design of containers such as hoppers, bottles and cans for storage and transportation of powders.

In addition, the fact that there is considerable variation in the progress of compression throughout a sample can theoretically explain the difficulty in reproducing measurement of apparent volume of powders. A powder layer of a given porosity, when under compression, may have numerous layers of particles of different steric orientation. Compressive forces may affect these layers differently and result in decreased reproducibility of results.

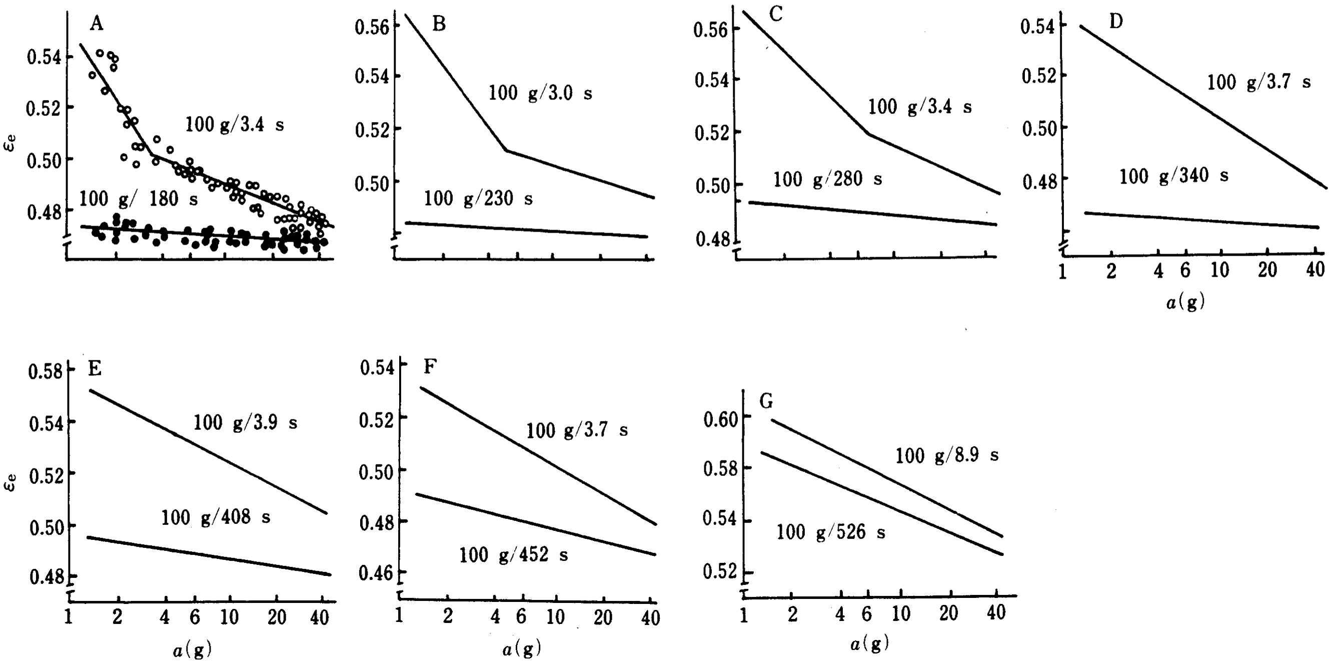

Fig. 6. Influence of Particle Size on Semi-log Plot of ε_e *vs. a*

Sample: borax; A, 950 μm; B, 670 μm; C, 470 μm; D, 350 μm; E, 190 μm; F, 135 μm; G, 100 μm. Key: ○, prepared with maximum pouring rate; ●, prepared with minimum pouring rate.

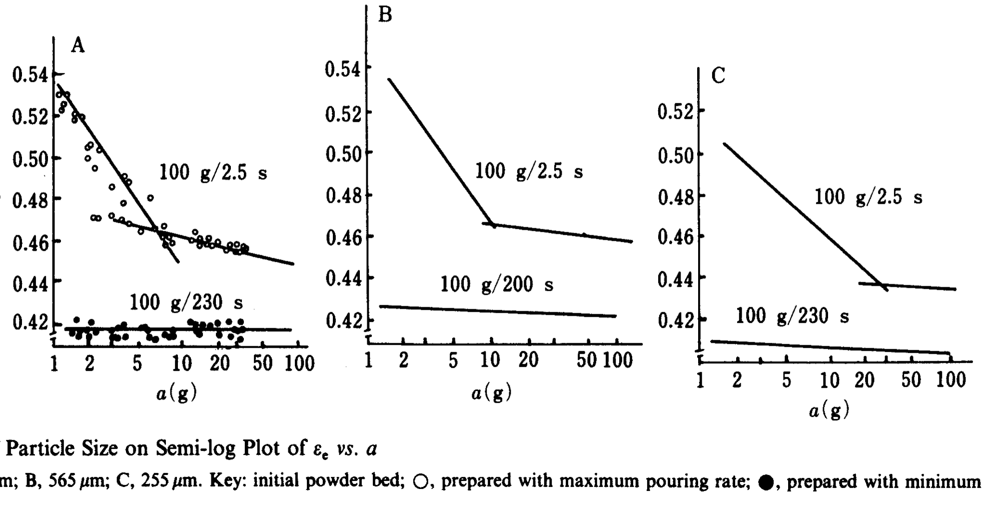

Fig. 7. Influence of Particle Size on Semi-log Plot of ε_e *vs. a*

Sample: GP; A, 700 μm; B, 565 μm; C, 255 μm. Key: initial powder bed; ○, prepared with maximum pouring rate; ●, prepared with minimum pouring rate.

Effects of Particle Size on Compression by Tapping The physical properties of the particles can affect compression by tapping and can therefore influence the lines mentioned above.

Inorganic Pharmaceuticals and Inorganic Materials Figure 6 is a ε_e *vs.* a semi-log plot of fractions of seven types of borax with average particle size ranging from 135 to 950 μm.

Figure 7 shows the semi-log plots of three types of GP with particle size ranging from 255 to 700 μm. In all cases, none of the lines except the three mentioned above appeared to have any particular significance, thus only these three lines are shown.

As is evident from Figs. 6 and 7, as the particle size decreases, the slope of line 1 decreases but the slope of line 2 increases. In the case of borax with a particle size less than 350 μm, line 1 disappears completely. As the particle size decreases, both the slope and the intercepts of line 3 decrease and it approximates line 2. The result is that the lines tend to merge as particle size decreases. When the data from borax and GP were compared, the only difference found was a slight variation in the porosity for the most densely packed state at the acceleration represented by line 3.

Organic Pharmaceuticals Figure 8 is a ε_e *vs.* a semi-log plot of the four fractions of sulfisoxazole whose average particle size varies from 700 to 200 μm.

Figure 9 is a ε_e *vs.* a semi-log plot of four fractions of lactose whose average particle size varies from 60 to 220 μm.

Figures 8 and 9 reveal a tendency similar to that found for borax. The larger particles produced three distinct straight lines but as particle size decreased, line 1 became indistinct and lines 2 and 3 showed a merging tendency. This phenomenon was not limited to inorganic materials such as borax and GP, but was also observed in organic materials such as lactose and sulfisoxazole. One of the reasons that line 1 does not appear separate as particle size decreases is that the ratio of particles in contact with the cylinder wall and those not in contact increases as particle size decreases. Thus, as particle size decreases, the influence of the interaction between the particles and the cylinder wall can be ignored.

The main reason that lines 2 and 3 are approximately the same as particle size decreases may be that the speed of sample layer formation is influenced by cylinder size. This suggests a decrease in the flowability of the particles and in the speed of formation of the initial layer, with the result

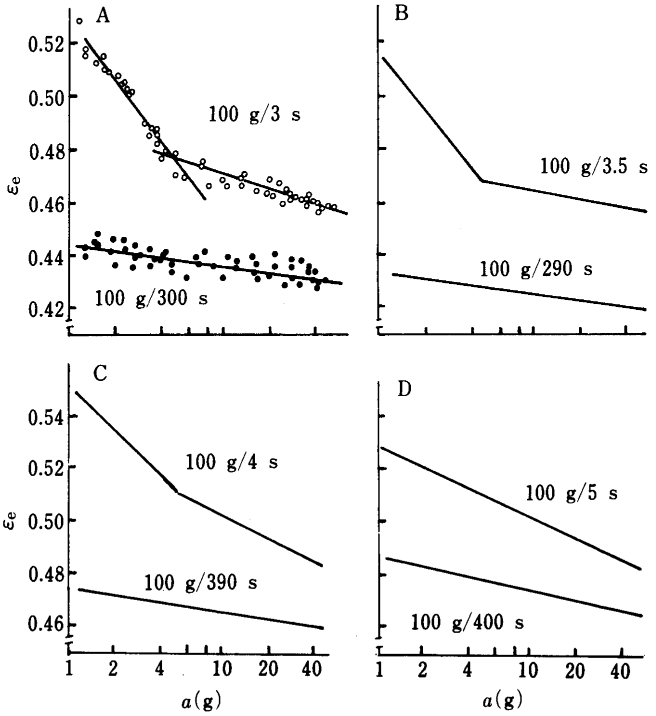

Fig. 8. Influence of Particle Size on Semi-log Plot of ε_e *vs.* a

Sample: sulfisoxazole; A, 700 μm; B, 570 μm; C, 450 μm, D, 200 μm. ○, prepared with maximum pouring rate; ●, prepared with minimum pouring rate.

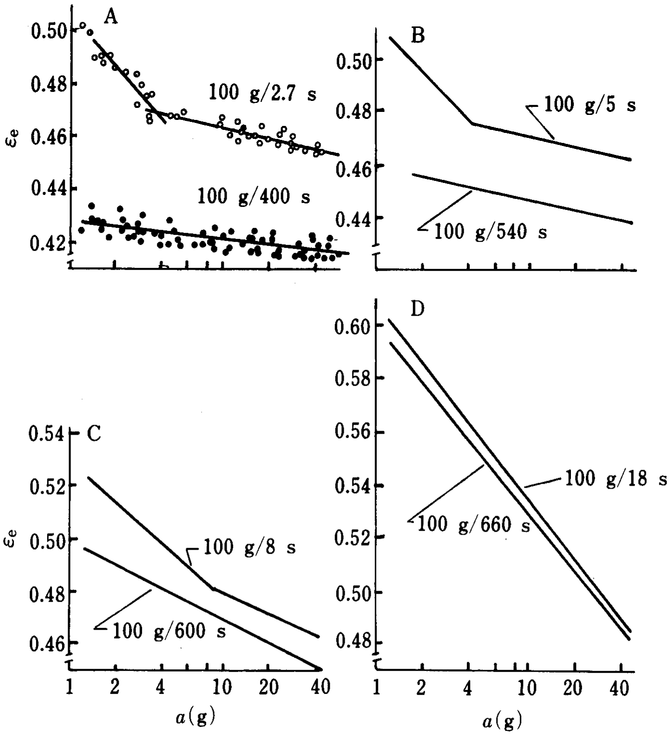

Fig. 9. Influence of Particle Size on Semi-log Plot of ε_e *vs.* a

Sample: lactose; A, 220 μm; B, 150 μm; C, 110 μm; D, 60 μm. Key: initial powder bed; ○, prepared with maximum pouring rate; ●, prepared with minimum pouring rate.

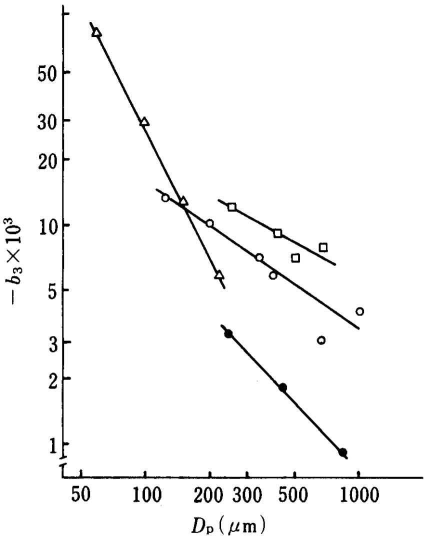

Fig. 10. Relation between Tapping Constant ($-b_3$) and Particle Size (D_p)

△, lactose; □, sulfisoxazole; ○, borax; ●, GP.

that there is little difference in the porosity between the highest and lowest filling rate.

The other possible explanation is that cohesion between the particles increases as particle size decreases. Cohesion may dominate in the early stage of the formation of the initial layer with the result that the porosity of this layer may be unrelated to the filling speed.

A shifting of joint between line 1 and line 2 is observed in Figs. 6—9. This phenomenon is considered attributable to a decrease in slope of line 1 and an increase in slope of line 2 with decreasing particle size.

Slope of Line 3 and Particle Size Observation of Fig. 6 through 9 reveals that intercept and slopes on the three lines vary with both material and particle size. This is especially true of the slope $-b_3$ of line 3, which seems to vary continuously with particle size. Figure 10 is a log plot of the average particle size D_p and slopes $-b_3$ of line 3 for the various samples.

Figure 10 reveals that there is somewhat of a disarray in samples of larger particle sizes. It is evident, however, that there is a fairly close correlation between D_p and $-b_3$ for all samples. The intercepts and slopes are, however, unique to the type of material. This suggests that the slope $-b_3$ of the straight line representing compression by tapping is not affected by the particle size alone but is also influenced by other factors such as cohesion.

References and Notes

1) E. Fukuoka, S. Kimura, M. Yamazaki and T. Tanaka, *Chem. Pharm. Bull.*, **31**, 221 (1983).
2) Present address: *Pharmacy of Hospital attached to the Faculty of Dentistry, Tokyo Medical and Dental University, 1-5-45, Yushima, Bunkyo-ku, Tokyo 113, Japan.*
3) A. Otsuka, H. Sunada and K. Danjo, *Zairyo*, **18**, 509 (1969).
4) M. Jinbo, S. Asakawa and T. Somiya, Abstract of Papers, Symposium of Powder Technology, Fukuoka, 1970, p. 56.
5) E. Fukuoka and S. Kimura, *Yakugaku Zasshi*, **95**, 1333 (1975).
6) Y. Sanoh, K. Motegi and E. Terada, *Zairyo*, **17**, 484 (1968); T. Morioka, Y. Ikegami, E. Nakajima, *Yakuzaigaku*, **19**, 119 (1959).
7) K. Kawakita, *Powder Technol.*, **4**, 61 (1970); M. Hasegawa, A. Otsuka and F. Higashide, *Yakuzaigaku*, **46**, 50 (1986).
8) S. Asakawa, M. Jimbo and O. Sakai, *Funtai Kougaku Kenkyu*, **9**, 171 (1972); F. Takagi and M. Sugita, *ibid.*, **16**, 528 (1979); I. Sugiura, *Oyobutsuri*, **17**, 168 (1948).