Cohesion of Particulate Solids. VIII.[1] Influence of Particle Shape on Compression by Tapping

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The influence of particle shape of granules or powder on tapping compaction behavior has been studied. Several kinds of samples, glass beads and glass powder, intact lactose granules and spherical lactose granules *etc.*, where the same material was made into different shapes, were used.

The diameter of the particles and the shape factor were measured. Most of the samples showed three compaction lines (lines 1—3), irrespective of the shape of the granules.

The segments of the three compaction lines (k_1, k_2 and k_3) of glass powder were significantly larger than those of glass beads. The segments of line 3, (k_3), of the rounded granules were smaller than those of intact granules.

Dependency on the shape of the particles was noted in the three tapping compaction lines.

A case in which tapping compaction behavior could not be adapted to a straight line was reported. In this case, special particles were considered.

Keywords powder; granules; tapping; compaction; empirical equation; particle shape; porosity; loosest packing; most closed packing; filling rate

In a previous report,[1] the authors proposed an experimental formula making it possible to linearize tapping compaction behavior, reporting the feasibility of approximating the tapping compaction of many kinds of granules to Eq. 1:

$$\varepsilon_e = -b \log a + k \tag{1}$$

where ε_e is the porosity in powder bed which has reached a steady-state at a certain level of acceleration "a," and b and k are constants.

Although Eq. 1 is similar to Balshin's equation,[3] the mechanism of compression is different and so Eq. 1 is not the same as Balsin's equation.

It was possible to represent only two pouring speeds of the samples prior to the beginning of tapping compaction, the fastest and the slowest.

When the particle size is relatively large, it is possible to approximate it to two straight lines (lines 1 and 2) connected to each other on the fastest filling, and to a single independent straight line (line 3) at some distance from these two on the slowest filling.

As the granule size becomes smaller, it becomes more difficult to distinguish line 1 from line 2 and they begin to approach line 3.

After consideration of the physical significance of these three straight lines, the dependence of the slope of the straight line b and segment k on particle size will be discussed.

Ridgeway *et al.*[4] have attempted a shape classification of granules utilizing the difference in their movement on a slanted vibrating plate according to the shape of the granules. From natural sand, these investigators selected particles with shape factors within a certain range to be used as samples. Shape dependency was found in the apparent density, angle of repose and flow rate through an orifice.

Ridgway *et al.*[5] have also reported on the relationship between a granule's weight, and its standard deviation, upon entering the die on tabletting as well as their shape.

Fonner *et al.*[6] reported on the apparent density and shape of granules. Aoki *et al.*[7] studied and reported on the relationship between the angle of repose, angle of internal friction, and porosity and the shape of the particles.

In the previous paper,[8] we defined the surface tensile strength of one particle as the cohesiveness of the powder and we devised a particle shape sorting apparatus.[9] The principal of this apparatus is based on discrimination using the ease of rolling of the granules.

Using various kinds of model granules and classified practical granules as samples, we conduct "measurement of the vertical tensile strength of a powder layer" according to the method we devised. The dependency of the surface tensile strength of a powder on the particle shape was reported.[10]

The dependency on granule shape is thus one of the characteristics of a powder. Because of the difficulty in obtaining samples consisting of the same material with similar particle size, but different particle shape, experimental confirmation has been accomplihsed only rarely.

In the present report, samples made from an identical material with a similar degree of granularity, but of different shape, were subjected to tapping compaction. The dependence of the segment and slope of the tapping compaction line proposed by authors on the shape of the granule was evaluated.

Finally, we report a case using a sample of single component particles where the measured values cannot be fitted to linear Eq. 1.

To our knowledge the relationship between the behavior in tapping compaction and particle shape has never been reported before.

Experimental

Apparatus and Methods As has been reported previously,[1] a tapping apparatus, modified to make it possible to monitor the acceleration of the impact on tapping, was used to obtain the maximum acceleration at the impact of tapping (subsequently called simply acceleration "a") and the porosity at a steady state "ε_e" under the said acceleration. The relationship between a and ε_e was plotted graphically to obtain the slope of curve b and segment k to evaluate their relationship to particle shape.

The rate of pilling of the sample layer was measured at only two extreme speeds, the fastest and slowest, as has been reported before.[1] No other rate of pilling was measured because of the relatively minor physical significance, as already reported.[1]

As in the previous report, all experimental procedures were conducted in a thermostatic chamber (Tabai Co., Ltd.) with constant humidity, at

20 ± 1 °C and relative humidity (R.H.) 50 ± 5%. The capacity of a measuring glass used to measure an apparent volume of powder was 100 ml and the apparent volume of the samples used was 90—100 ml.

Samples 1) Glass Powder and Glass Beads: Glass powder (GP): Commercially available glass powder (Toshiba Co., Ltd.) was separated into two fractions using a standard sieve (average particle diameter, $d=570$ and 250 μm respectively).

Glass beads (GB): Commercially available glass beads (Toshiba Co., Ltd.) were also separated into two fractions using a sieve ($d=490$ and 200 μm respectively).

Since the shape of the GB and GP was different, the concept of "identical granules" is not valid, but the influence of the particle diameter was kept to a minimum.

2) Spherical Lactose Granules: To lactose, passed through a standard sieve of 200 mesh, was added a small amount of 3% starch solution with kneading. Using a 16 mesh sieve, granules were prepared by pushing them out by hand. About 1/4 were dried immediately at about 70 °C in a hot air desiccator (fraction I). The remaining undried 3/4 were transferred to a 200 mesh silk sieve placed on a "Gyro shifter" (Kotobuki Co., Ltd.) which produced a horizontal circular movement. Granules rolled on the silk sieve, gradually becoming rounded. After 15 s, approximately 1/4 of the granules were taken out to dry (fraction II). The remaining 1/2 of the granules were subjected to further circular movement for another 15 s and again about 1/4 of the granules were then taken out and dried in the same way (fraction III). Finally, the remaining 1/4 were left in the apparatus for a further 90 s and then dried (fraction IV). The four dried fractions were fractionated using standard sieves, and granules ranging in size from 14 to 18 mesh were selected. Each even-grained fraction was divided into a certain range of shape factor using the shape sorting apparatus devised by us.[9]

The shape-sorted granules of fr. I are referred to in Figs. 4 and 5 as "intact granules," those of fr. II as "15 s rounded granules," those of fr. III as "30 s rounded granules" and those of fr. IV as "120 s rounded granules," respectively.

3) Cylindrical Granules: The commercially available granules of *para*-aminosalicylate calcium (PAS calcium®, Sankyo Co., Ltd.) are cylindrical in shape with extreme geometrical regularity, with a shape factor of $\psi=0.43$. The particle size distribution and shape distribution are almost neglegible, so these were used as model granules.

Method of Measurement of the Shape Factor The method of measurement of the shape factor and the method of its expression was the same as described by Aoki,[11] using the ratio between the diameters of the internal contact sphere d_i and that of the external contact sphere d_o; $\psi=d_i/d_o$.

Both d_i and d_o were measured under a microscope and their ratio calculated; this was followed by the calculation of ψ, based on the mean of the measured values of 50 particles.

Table I summarizes the mean granular diameter of each sample (Feret's diameter) and shape factor.

Table I. Particle Size and Shape Factor of the Granules Used

Sample	Particle size (μm)	Shape factor (ψ)
GB (coarse)	490	1.00
GB (fine)	200	1.00
GP (coarse)	570	0.71
GP (fine)	250	0.71
para-Aminosalicylate calcium	3000	0.43
Intact granule	1500	0.50
15 s spherized granule	1300	0.59
30 s spherized granule	1200	0.62
120 s spherized granule	1200	0.66
MCC (coarse)	150	—
MCC (fine)	75	—

Results and Discussion

The results plotting $\varepsilon_e=$ *vs.* a on a semi-log scale, by tapping GP with a mean diameter of 250 μm and of GB with a mean diameter of 200 μm, are shown in Fig. 1.

A similar plot of GP with a mean diameter of 570 μm and GB with a mean diameter of 490 μm is shown in Fig. 2.

As is clearly seen in Figs. 1 and 2, three compaction curves (lines 1—3) are evident for GB and GP.

As was shown in a previous report,[1] line 1 was made by piling the sample at the highest speed in the cylinder. The loosest porosity of filling an infinite vessel, further modified by a mutual interaction with the vessel wall, is probably expressed here. Because of the presence of the vessel wall, the degree of directional freedom of the granule is restricted, and this can result in unstable piling. This would increase the proportion with a large porosity.

Even in the spherical granules which lack such directional constraint, theoretically preventing interaction with the vessel wall, slight but definite evidence of line 1 was noted.

Granules which should achieve stable positions under gravity are prevented from doing this by the presence of a vessel wall. This can be called "restriction of the degree of freedom on the position of particles."

All particles, except those that are spherical, have a directional quality. Consequently, in addition to the restriction on the degree of positional freedom, restriction on the degree of directional freedom will operate. Probably as the result of this, the slope of line 1 of GP can be greater than that of GB.

The line 1 produced by samples with a relatively large diameter, assuming non-spherical shape, is thought to represent the result of the continuous mutual effect of these two actions.

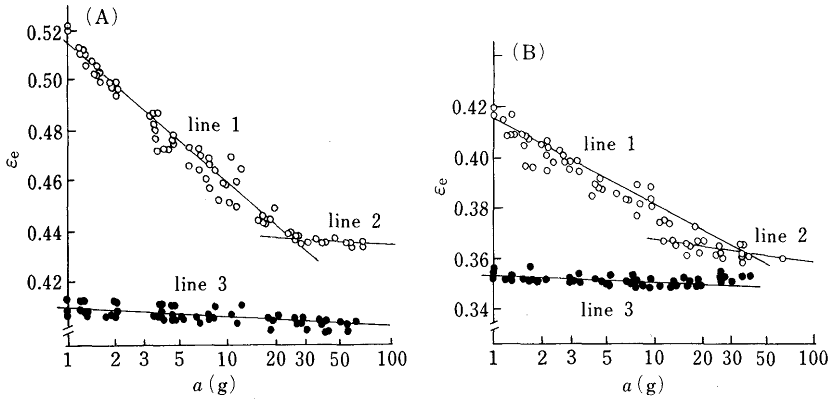

Fig. 1. Influence of Particle Shape on ε_e *vs.* a Plot for GP (250 μm) (A) and GB (200 μm) (B)

Key; pouring speed of initial powder bed, ○, fastest (GP: 14 g/s, GB: 21 g/s); ●, slowest (GP: 0.13 g/s, GB: 0.12 g/s).

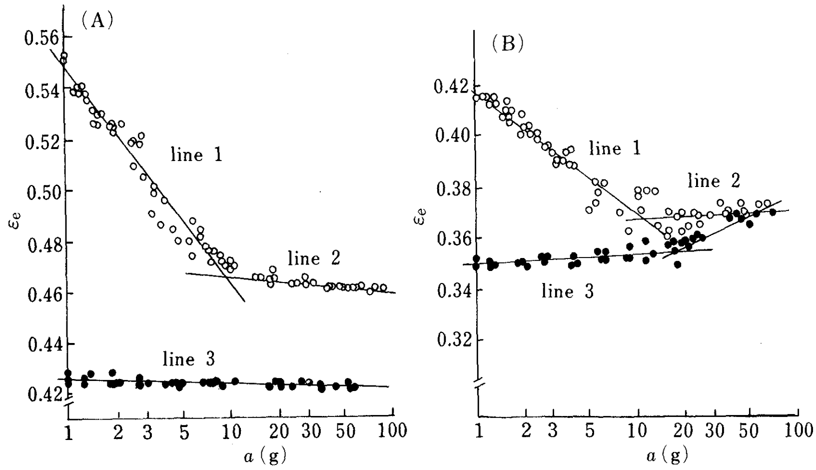

Fig. 2. Influence of Particle Shape on ε_e *vs.* a Plot for GP (570 μm) (A) and GB (490 μm) (B)

Key; pouring speed of initial powder bed, ○, fastest (GP: 12 g/s, GB: 18 g/s); ●, slowest (GP: 0.14 g/s, GB: 0.17 g/s).

TABLE II. Comparison of the Segment (k_1—k_3) between GP and GB

Sample	k_1	k_2	k_3
GP (570 μm)	0.548	0.473	0.425
GB (490 μm)	0.416	0.367	0.349
GP (250 μm)	0.514	0.442	0.409
GB (200 μm)	0.415	0.376	0.352

These specific phenomena will be discussed later.

One remarkable differences of GP ($\psi=0.71$) and GB ($\psi=1.00$) consists of the difference in the intercept of the three straight lines with the vertical axis ($a=1.0$ g).

The intercept of line 1 to line 3 are expressed as k_1—k_3, and these values are summarized in Table II.

As is evident in Table II, values k_1—k_3 produced by GB were always smaller than GP, indicating the expected tendency of dense packing not only under natural gravity, but also under various levels of acceleration, as the shape becomes closer to spherical, in accordance with the overall model.

The second difference between GP and GB is, as is evident from Figs. 1 and 2, the distance between lines 2 and 3.

As shown in a previous report, it is difficult to pack GP densely, once it has been packed loosely, even under a high intermittent acceleration such as is produced by tapping, similarly to other substances.

By contrast, in the spherical-shaped particles such as GB, 10—20 g acceleration is sufficient to induce dense packing from a state of coarse packing, indicating an intimate relationship between the filling by tapping and particle shape.

As shown in Fig. 2, line 3 produced by GB showed a definite linear rise from a point of acceleration greater than 20 g. (Line 3 also tended to rise slightly from 1 g, the initial acceleration.) This phenomenon is observed in GB of 490 μm but not in GB of 200 μm. Thus, these specific phenomenon are related not only to materials but also to particle diameter.

Figure 3 shows the semi-log plots of ε_e *vs.* a in GB with a granule diameter of 820 μm. As shown in Fig. 3, lines 1 and 2 appear to approximate to a straight line, while line 3 showed no change up to an acceleration of approximately 8 g, then tended to increase after 8 g. This tendency was the same with the line obtained by GB with a diameter of 490 μm, shown in Fig. 2.

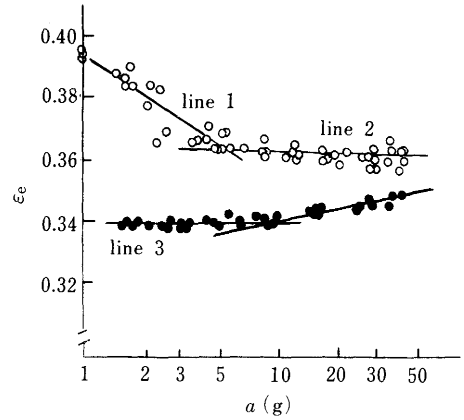

Fig. 3. Scatter Diagram of ε_e *vs.* a Plot for GB (820 μm)

Key; pouring speed of initial powder bed, ○, fastest (18 g/s); ●, slowest (0.31 g/s).

Oishi[12] compared the results of experiments conducted by various groups using steel balls and lead shot, which may be practically considered equal spheres, giving a porosity of 0.363, 0.370 on oscillation filling.

The glass beads used in the present experiment showed some distribution in their particle size, differing somewhat from the random filling of homogeneous spheres. Line 3 of GB with a diameter of 820 μm gave a porosity of $\varepsilon_e=0.340$, which is smaller than the minimum value for the random filling of a homogeneous spheres, 0.363. This value indicates considerable compaction already present, even in the initial filling, prior to tapping compaction. On infliction of impact, by tapping the layer of piled particles in its most dense state, the porosity does not change immediately, when the impact is weakest. As the impact increases, particles repulse each other, making the porosity even greater.

Such a phenomenon occurred in line 3, occasionally in line 2, and never in line 1, because the porosity was greater than in the state described by line 3.

A similar phenomenon has already been reported by Taneya *et al.*[13] and was explained by the repulsing of the particles from each other. Otsuka *et al.*[14] by contrast, reported the absence of such a phenomenon. The occurrence

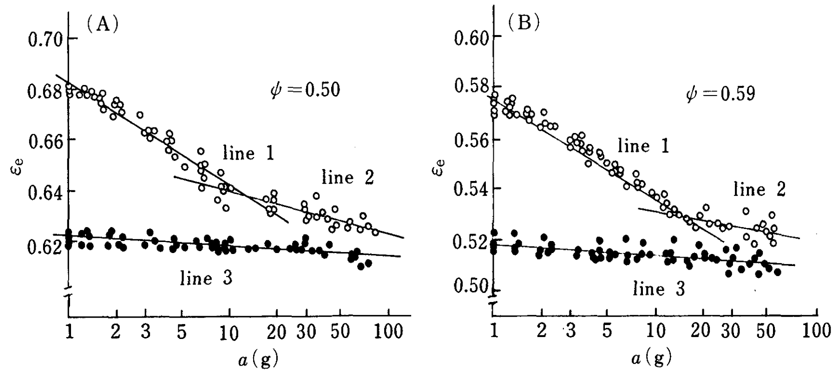

Fig. 4. Influence of Particle Shape on ε_e *vs.* a Plot for Intact Granules (A) and 15 s Rounded Granules (B)

Key; pouring speed of initial powder bed, ○, fastest (intact gr.: 3.8 g/s, 15 s rounded gr.: 6.2 g/s); ●, slowest (intact gr.: 0.11 g/s, 15 s rounded gr.: 0.16 g/s).

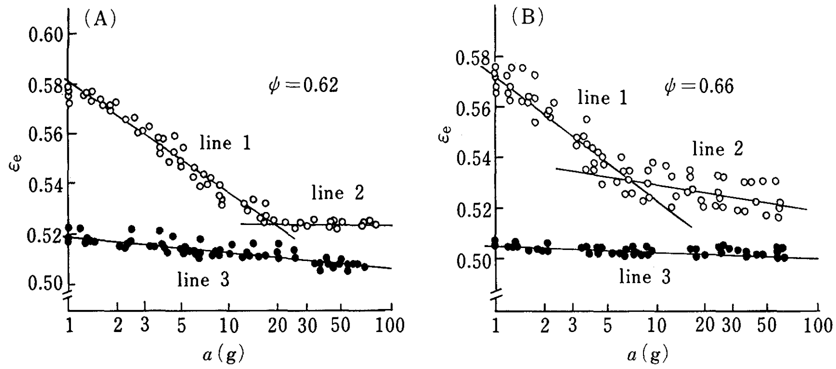

Fig. 5. Influence of Particle Shape on ε_e *vs.* a Plot for 30 s Rounded Granules (A) and 120 s Rounded Granules (B)

Key; pouring speed of initial powder bed, ○, fastest (30 s rounded gr.: 5.8 g/s, 120 s rounded gr.: 5.9 g/s); ●, slowest (30 s rounded gr.: 0.15 g/s, 120 s rounded gr.: 0.15 g/s).

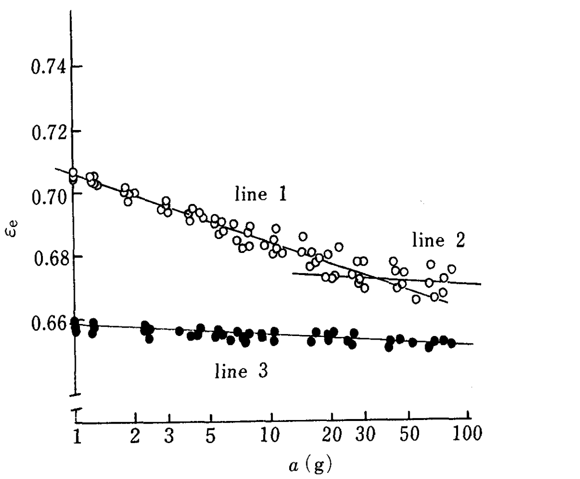

Fig. 6. Scatter Diagram of ε_e *vs.* a for Cylindrical Granules of *para*-Aminosalicylate Calcium (PAS Calcium)

Key; pouring speed of initial powder bed, ○, fastest (4.2 g/s); ●, slowest (0.13 g/s).

of repulsion probably depends not only on the diameter of the granules, but also on the shape of the particles and their nature.

Such a phenomenon does not occur in GP with the same material of similar granularity, probably because of the difference in the filling density.

The GP layer, unlike GB, cannot achieve a high density because of the particle shape. Because of the sharp angle of the particles, friction among the particles is pronounced, preventing repulsion. In addition to the material, the shape of the particles is thus also involved in the repulsion.

TABLE III. Comparison of the Segment (k_1—k_3) between Intact Granules and Spherical Granules of Lactose

Sample	ψ	k_1	k_2	k_3
Intact granules	0.50	0.683	0.656	0.622
15 s spherized granules	0.59	0.576	0.544	0.518
30 s spherized granules	0.62	0.580	0.525	0.518
120 s spherized granules	0.66	0.571	0.540	0.505

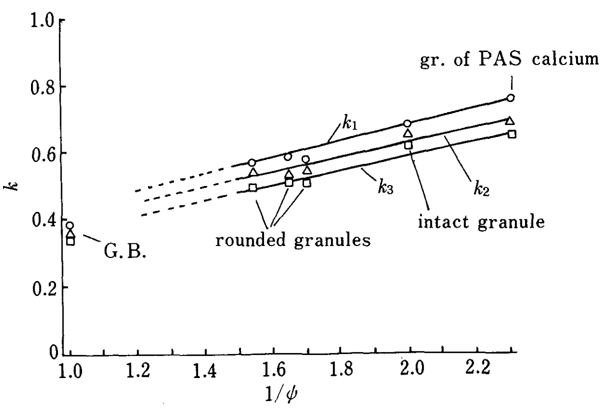

Fig. 7. Relation between Reciprocal of the Shape Factor ($1/\psi$) and the Segment (k_1—k_3)

Sample: hand made granules of lactose, commercially available granules of PAS calcium and glass beads.

The results of tapping compaction using intact granules and 15 s rounded granules are shown in Fig. 4, and those using 30 s rounded granules and 120 s rounded granules are

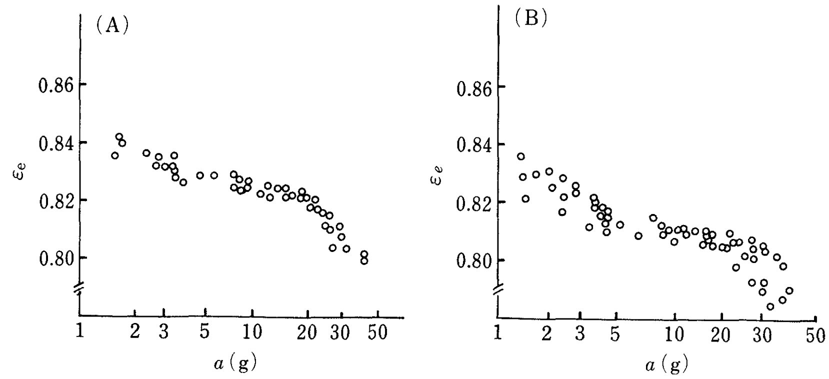

Fig. 8. Exceptional Nonlinear Relation between ε_e and Tapping Acceleration (a) on Semilog Scale for Micro-crystalline Cellulose (MCC)
(A) MCC (150 μm); (B) MCC (75 μm).

shown in Fig. 5.

As is evident from Figs. 4 and 5, the tapping compaction line made by the 4 samples consists typically of three straight lines. The three k values of these samples are given in Table III.

Spherical particles, although there seems to be some scattering, tended to show smaller values of k_1—k_3 as the value of ψ became smaller. As the shape factor (ψ) of the particles become smaller, denser compaction is a possible explanation for such a phenomenon.

No tendency of line 2 approaching line 3 was noted. Probably because the difference in shape was not as pronounced as that between GP and GB, discrimination of tapping compaction behavior between intact granules and rounded granules was not possible. However, there seems to be some distribution in line 2 at 120 s rounded granules in Fig. 5, but the reason is not clear.

The results of tapping compaction using granules of *para*-aminosalicylate calcium are shown in Fig. 6.

This consists of three straight lines as for other granular samples. It is evident from Fig. 6 that ε_e and the k values ($k_1=0.706$, $k_2=0.684$, $k_3=0.658$) are relatively large compared with other samples. This is attributable to the minimal shape factor ($\psi=0.43$) of the granules of *para*-aminosalicylate calcium.

k values were therefore plotted on the vertical axis and the reciprocal of the shape factor ($1/\psi$) was plotted on the horizontal axis. The results including those obtained by the compaction of the granules of the commercially available *para*-aminosalicylate calcium cylinder-shaped granules, were plotted in Fig. 7.

As is clear from Fig. 7, in the absence of a large difference in the diameter of the particles, the three k values showed a dependency on the reciprocal of shape factor ($1/\psi$), unrelated to the sample material.

The values obtained with GB, however, were located a little below of these three straight lines. Compared with other samples, k_1—k_3 lay closer to each other.

Unlike other materials, GB has a smooth surface without negative curvature and is devoid of small holes. Such a differences in the surface state may explain these findings.

Cases Which Do Not Fit the Linear Equation Most of the samples used for tapping compaction could be fitted to the experimental Eq. 1. However, we found one exception. Figure 8 is ε_e *vs.* a semi-log plot of two fractions ($d=150$ and 75 μm respectively) of micro-crystalline cellulose (MCC).

The two fractions of MCC both show ε_e *vs.* a semi-log plot of similar shape, both without linearity. The MCC of other fractions gave similar results.

The powder layer of MCC retained a higher porosity than other samples ($\varepsilon_e=0.830$—0.840) and this persisted even after tapping compaction. This is attributable to the shape of the MCC particles which, unlike potato starch, do not possess a simple shape with a smooth surface.

The MCC layer may also readily absrob the impact on tapping, producing a discrepancy between the acceleration by the vessel itself and the powder.

The acceleration measured by this method is that of the vessel and not the acceleration of the powder particles themselves. Consequently, when the granules absorb the impact, as in the case of MCC, a discrepancy may occur between the accleration of the vessel and the powder.

This may be the cause of the different behavior shown by this material. Tapping compaction, even if it cannot readily be approximate a straight line, may nevertheless reflect a bulk property, not readily expressed as the primary property of a substance.

When a deviation from the tapping compaction line occurs, the distinctive nature of the material and shape of the particles is suggested. Whether the data fits on a straight line or not, useful information about the properties of the powder is provided by this method.

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