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Effect of Lubricant on Die Wall Friction during the Compaction of Pharmaceutical Powders

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The wall friction of pharmaceutical powders (lactose, sucrose, sodium chloride, potassium chloride and sodium bicarbonate) was measured. The sample powders were compressed against a stainless steel wall. Then, keeping the normal pressure (P_N) constant, the wall was slid. During the sliding, the displacement of the wall and the normal and tangential pressures were recorded. For lactose and sucrose powders, the coefficients of wall friction (μ_w) were higher than those for the other powders. The effects of lubricants (magnesium stearate and talc) on μ_w and on the sliding process were examined. The values of μ_w for lubricated powders were 0.22 or less, being slightly dependent on P_N. The residual layer on the wall surface and the replicas of particles on the surface of the compact at various stages during the sliding process were examined. It was found that the junctions between the particles and the die wall grew in size during the course of sliding, and that the lubricants prevented this growth, resulting in decreases of the friction and adhesion forces.

Keywords——tablets; die wall friction; powders; lubricant; magnesium stearate; compaction

Introduction

There are many phenomena of powder flow in pharmaceutical processes, including gravity flow from the hopper and the constrained flow caused by the applied pressure during tabletting. Lubricants are used to improve powder flow properties, and it is important to select a suitable lubricant, to determine the appropriate amount, and to study the lubrication mechanism. In the case of tabletting processes, many effects of lubricants have been pointed out,[1] for example, the decrease in the friction between the tablet and the die wall, and the prevention of sticking to the surfaces of the punch and the die wall.

Using the pressure-transmission ratio from the upper to the lower punch, Higuchi *et al.*[2] evaluated the friction of pharmaceutical powders on the die wall, and then correlated the effectiveness of lubricant to its amount. They proposed two mechanisms for lubrication, *i.e.*, boundary and fluid types, according to the Bowden–Tabor theory.[3]

Adopting the same theory for the friction during die compaction, Train *et al.*[4] expressed the true contact area between the powder bed and the die wall in terms of relative volume, and they compared the shear strength of the compacts prepared from some pharmaceutical powders with the friction force on the die wall.

Müller[5] reported that solid lubricant admixed with powder followed the boundary type mechanism, and that the effectiveness was controlled by the shear strength of the lubricant layer. Furthermore, they discussed the dependence of effectiveness on the structure.

In the previous paper,[6] the effects of magnesium stearate admixed with potassium chloride on the mechanical properties of the mixture were examined. It was found that the lubricant decreased the coefficient of die wall friction more effectively than that of internal friction. In the present paper, the coefficients of die wall friction for some pharmaceutical powders with and without lubricants are presented, and the mechanisms of friction and lubrication are discussed.

Experimental

I. Preparation of Powders——The pharmaceutical powders employed were lactose, sucrose, potassium chloride, sodium chloride and sodium bicarbonate, which were graded from 42 to 65 mesh. Magnesium stearate and talc were used as lubricants without sieving. The powders were mixed with lubricant at 2 wt% in a V-type mixer at 50 rpm for 20 min.[7] The powders were left for at least a week in a room of almost constant temperature (25°C) and humidity (60%RH), and then used for measurements.

II. Apparatus and Procedures——The apparatus and the experimental procedures were the same as described in the previous paper.[8] The sliding plate of stainless steel was polished with a No. 400 sand cloth.

Results

I. Effect of Normal Pressure on the Coefficient of Wall Friction

At first, the effect of pressure normal to the sliding surface (P_N) on the coefficient of wall friction (μ_W) was examined. The results for all sample powders are summarized in Figures 1 and 2. The maximum and minimum values of the coefficient (μ_{max} (○) and μ_{min} (●)) are plotted. Their definitions are described later in detail.

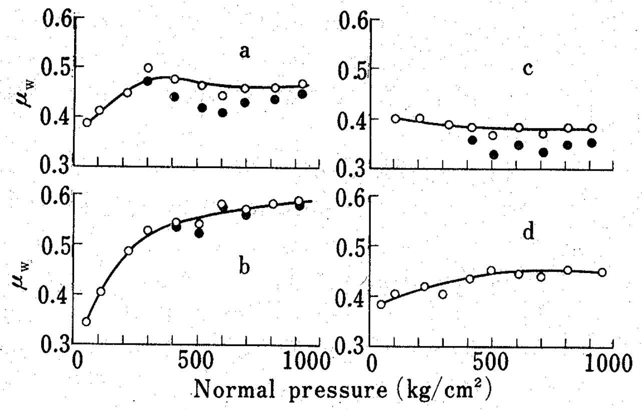

Fig. 1. Effect of Normal Pressure on the Coefficient of Wall Friction

a, lactose; b, sucrose; c, sodium bicarbonate; d, sodium chloride.

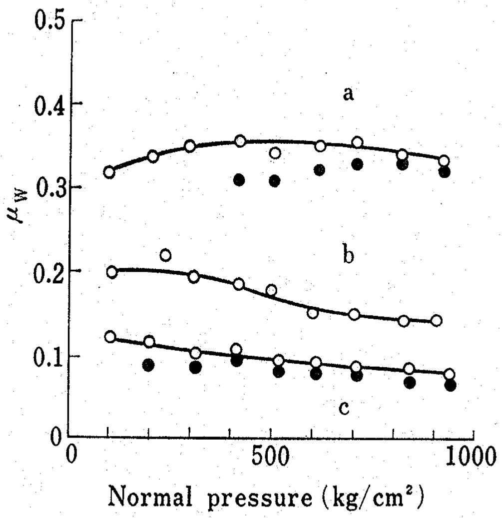

Fig. 2. Effect of Normal Pressure on the Coefficient of Wall Friction

a, lactose with 2 wt% talc; b, lactose with 2 wt% magnesium stearate; c, sodium bicarbonate with 2 wt% magnesium stearate.

For two organic powders, lactose and sucrose, the changes of μ_w were similar, as shown in Figs. 1-(a) and (b), respectively. The value of μ_w increased rapidly in the low P_N range, then approached a constant value at about 300 kg/cm²; 0.45 for lactose and 0.55 for sucrose.

Sodium bicarbonate showed nearly constant μ_W of 0.40 over the examined range of P_N (Fig. 1-(c)). For sodium chloride, μ_W increased gradually (0.40—0.45) as shown in Fig. 1-(d).

In common with the lubricated powders, significant decrease in the wall friction force was observed. The dependence of μ_W on P_N was slight (Fig. 2). For lactose lubricated with talc, μ_W was nearly constant (0.32—0.36) and lower than that for unlubricated lactose. The values of μ_W for powders lubricated with magnesium stearate were much lower than those for powders lubricated with talc, being in the range of 0.04 to 0.22.

II. Sliding Process (Plot of the Coefficient of Wall Friction versus Displacement)

The sliding process may be expressed by plots of μ_w versus displacement, while the powder bed (the compact) is sliding on the wall. The displacement refers to the distance from the point of the initially applied tangential force. The results are shown in Figures 3, 4 and 5

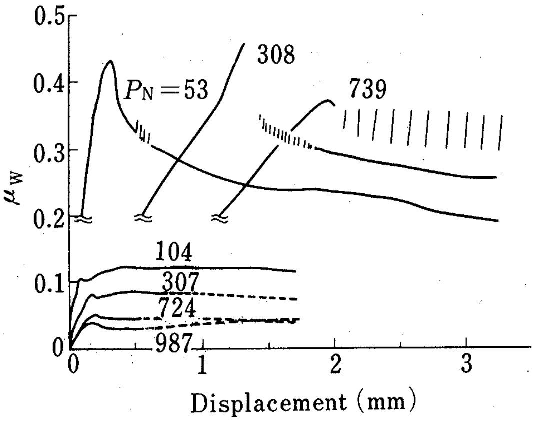

Fig. 3. Records of the Coefficient of Wall Friction during Sliding of Potassium Chloride Powders

Upper curves: unlubricated powders. Lower curves: lubricated powders with 2 wt% magnesium stearate. P_N: [kg/cm^2].

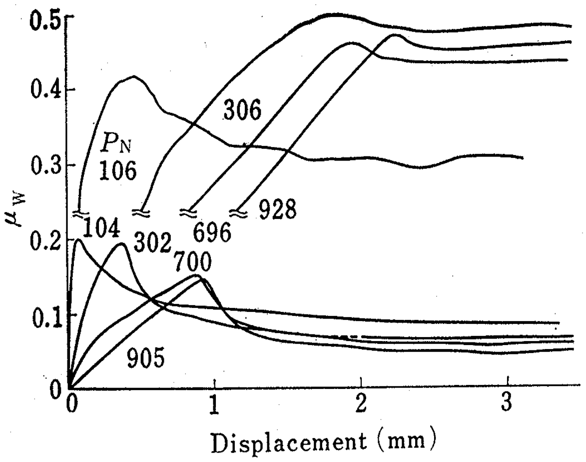

Fig. 4. Records of the Coefficient of Wall Friction during Sliding of Lactose Powders

Upper curves: unlubricated powders. Lower curves: lubricated powders with 2 wt% magnesium stearate. P_N: [kg/cm^2].

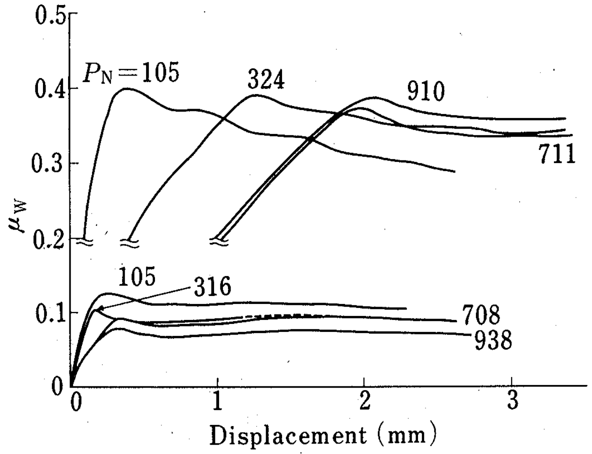

Fig. 5. Records of the Coefficient of Wall Friction during Sliding of Sodium Bicarbonate Powders

Upper curves: unlubricated powders. Lower curves: lubricated powders with 2 wt% magnesium stearate. P_N: [kg/cm^2].

for potassium chloride, lactose and sodium bicarbonate, respectively. In these figures, the upper curves are for unlubricated powders, and the lower curves are for lubricated powders.

(1) **Unlubricated Powders**——Sodium chloride and potassium chloride gave the same sliding pattern, a "stick-slip" as described by Fukumori and Okada.[8] On the other hand, sodium bicarbonate, lactose and sucrose gave a very different sliding pattern from the above-mentioned two powders. The value of μ_W increased with sliding and reached a maximum, followed by a gradual decrease. At high P_N, it increased again (at P_N=928 kg/cm^2 for lactose). In contrast, at low P_N it decreased irregularly and did not reach a steady value (for example, at P_N=106 kg/cm^2 for lactose and at P_N=105 kg/cm^2 for sodium bicarbonate).

(2) **Lubricated Powders**——The sliding process was also influenced markedly by the lubricants. The admixture of magnesium stearate with potassium chloride gave a smooth sliding without "stick-slip", though it gave curves of very small amplitude in the region of large displacement. The value of μ_W for lubricated potassium chloride showed a maximum, a minimum, and then reached a higher plateau successively, over the whole range of P_N. The behavior of μ_W for lubricated sodium bicarbonate was similar to that for lubricated potassium chloride. For lubricated lactose, μ_W was different from that for the above two lubricated powders. With sliding, a rapid decrease in μ_W was observed after the maximum, and then a nearly constant value was reached.

For all the powders lubricated with magnesium stearate, the displacement to the maximum μ_W was nearly constant and less than 1 mm over the range of P_N. On the other hand, for unlubricated powders the displacement was considerably dependent on P_N.

In the previous section, the maximum and minimum values for μ_W, which can be read from the curve, are represented by μ_{max} and μ_{min}, respectively.

Discussion

Coefficient of Die Wall Friction

According to the Coulomb–Amontons law, the friction force between solids is independent of the apparent contact area between the two opposing surfaces, and is proportional to the load at the interface. The coefficient of friction is independent of the load. Bowden and Tabor[3] showed that the true contact area at the interface was much smaller than the apparent area, and that the true area increased with increasing load. The friction force is the tangential one, which is required to cut the junctions.

When the micro asperities on the surfaces deform plastically, the true contact area (A), which corresponds to the summation of the areas of all junctions, is proportional to the load (W),

$$A = W/\sigma_Y \qquad \text{Eq. 1}$$

where σ_Y is the yield stress of the material. The total friction force (F) may be expressed as the product of the true contact area and the average stress required to cut the junctions (S).

$$F = A \cdot S \qquad \text{Eq. 2}$$

Combining equations 1 and 2,

$$F = W \cdot (S/\sigma_Y) \qquad \text{Eq. 3}$$

Then, the coefficient of friction (μ) is

$$\mu = F/W = S/\sigma_Y \qquad \text{Eq. 4}$$

As S and σ_Y are inherent properties of the material, the friction force is dependent on W and independent of apparent contact area.

Bowden and Tabor[3] described some exceptional examples that deviated from this proportionality, such as the frictional system consisting of a metal slid on a hard metal with a thin layer of soft metal, and that of a soft metal slid on the surface of a hard metal. The deviation can be interpreted as follows. The true contact area depends not only on the normal pressure, but also on the mechanical properties of materials and on the atmosphere.

The frictional systems consisting of the pharmaceutical powder bed (the compact) and the metal die wall in the present study may be more complicated. As regards the mechanism of contact, the ratio of the true area to the apparent area increases rapidly with increasing pressure normal to the die wall. The former is the actual area over which the particles are in contact with the die wall, and the latter is the apparent area which can be evaluated from the geometrical dimensions of the powder bed. This effect is due to the rapid dentification of the powder bed with increasing compression pressure. In the previous paper,[9] it was shown that, for potassium chloride powders in contact with a stainless steel surface, the ratio increased from 40 to 70% with increasing normal pressure in the range of about 200 to 900 kg/cm^2. Furthermore, particle deformation on the die wall affected the true contact area. As pharmaceutical particles deform not only plastically but also fragilely,[10] Eq. 1 can not always be applied. The particle deformation also influences the surface structure of the powder bed in contact with the die wall. According to Okada *et al.*,[11] the surface of a bed of lactose (as a brittle body) was smoother than that of potato starch (as a plastic body). The mechanism of sliding must be different from that of a metal/metal system because the mechanical properties of the opposing surfaces (of powder bed and wall) are very different. Thus, the shear strength and the yield stress of the particles will become important.

It is expected that the changes in both true contact area and friction force may be different for the pharmaceutical powders used in the present work. In addition, this experimental system is quite different from solid/solid and metal/metal systems. However, the deformation of particles on the die wall under the normal pressure can be discussed on the basis of the above-mentioned theory on the deformation of micro asperities.[12] The true contact area would be the actual area over which the particles are in contact with the die wall. σ_Y in Eq. 1 could be regarded as the physical property determining the true contact area. The effect of the plastic deformation of particles could be included in this property. Accordingly, the friction and lubrication for this experimental system will hereinafter be discussed on the basis of the theory of friction of solids.

For two kinds of organic powders (lactose and sucrose), μ_W increased rapidly with P_N but attained a steady value in the range of high P_N (Figs. 1-(a), (b)). After sliding, the surface of the wall was examined with the naked eye. An irregular cohesive residual layer was observed on the surface at $P_N=100$ kg/cm^2. In addition, a distinct sheared residual layer was observed on the surface at $P_N \geqq 300$ kg/cm^2. In comparison with potassium chloride[8] and sodium chloride powders, for which no residual layer was observed, it was considered that the adhesion force was strong at the sliding interface between the powder bed and the wall. The original particles were noticed on the surface of the residual layer at low P_N, but not at $P_N \geqq 300$ kg/cm^2. These observations indicate that these organic powders behave as a brittle body.[10]

For sodium bicarbonate and sodium chloride powders, μ_W was relatively low and increased slightly with P_N (Figs. 1-(c), (d)). For sodium chloride, the so-called stick-slip was detected during sliding. No residual layer could be seen after sliding. This means that sodium chloride does not behave as a brittle body but as an elastic body. In the case of sodium bicarbonate, a cohesive residual layer was observed on the surface at $P_N \geqq 200$ kg/cm^2, but no sheared residual layer. The original particles were not apparent on the surface of the residual layer at $P_N \geqq 500$ kg/cm^2. Thus, in analogy with lactose, the adhesion force might increase in the range of high normal pressure.

For two kinds of organic powder, during the process of formation of the contacting surface of the powder bed by the normal pressure, the true contact area of particles increases with increasing fracture of particles caused by the increasing normal pressure. At the initial stage of compaction, the increase is rapid, in accord with the dentification of the powder bed.[10a] The friction and adhesion forces increase with increasing true contact area of particles. On the other hand, for powders which do not behave as a brittle body, the dentification is rather slow. These considerations would suggest that μ_W for the organic powders increased more rapidly than that for the inorganic powders in the low normal pressure ranges, and also that, even at $P_N=300$ kg/cm^2, sliding occurred within the lactose powder bed due to the increase of friction force on the wall surface.

Strijbos studied the wall friction during powder compaction.[13] The wall friction for powders and granules of ferric oxide was measured. The results were discussed on the bases of the ratio of particle hardness (H_p) to wall hardness (H_w) (V.H.N.) and of the ratio of particle size (d_p) to roughness size (R_w). Two mechanism of sliding were proposed, *i.e.*,

(1) true friction between the powder and die wall (in the case of $d_p/R_w > 1$)
(2) friction between powder and powder, due to the formation of a cohesive particle layer on the wall (in the case of $d_p/R_w < 1$, and/or $H_p/H_w < 1$)

Mechanism (2) gave a higher value of the coefficient of friction than (1). For pharmaceutical powders used in the present study, $H_p/H_w < 1$. The fracture of particles occurs more easily for lactose and sucrose powders, so that the particle size is reduced more easily than in the cases of potassium chloride and sodium chloride. These results imply mechanism (2), and consequent high friction, for the organic powders.

The friction force and the coefficient of friction are expressed by equations 2 and 4, respectively, for the unlubricated case. There may be two ways to decrease the friction force,

(1) reducing the true contact area (A)

(2) reducing the shear strength (S)
For the mechanically isotropic materials such as general metals, a hard material gives a small value of A and a large value of S, and *vice versa.* This is the reason that the values of μ for metals remain in a narrow range.[3]

Lubricants are applied to decrease the friction force. When the contact area is covered by a lubricant layer with low shear strength, the shear strength of the system is controlled by that of the layer. On the other hand, the true contact area is controlled by the mechanical strengths of the opposing materials. Accordingly, from Eq. 4, the coefficient of friction for lubricated powders on the die wall may be written as,

$$\mu_{\mathrm{lub}} = S_{\mathrm{lub}}/(\sigma_{\mathrm{Y}})_{\mathrm{powd}} \qquad \text{Eq. 5}$$

where $(\sigma_{\mathrm{Y}})_{\mathrm{powd}}$ is the mechanical strength of the powders. It is clear that S_{lub} is smaller than S_{powd}. As described above, the true contact area depends not only on the plastic deformation of the particles, but also on the dentification and deformation of the powder bed. The dentification was affected by the mechanical properties of powders,[7] though the relative volume increased in the lubricated powder system. The use of $(\sigma_{\mathrm{Y}})_{\mathrm{powd}}$ in Eq. 5 is appropriate. Many mechanisms for lubrication have been proposed.[14] The mechanisms suggested in pharmaceutical processes[4,5] can be summarized as follows.

(1) decrease of the direct contact area between the powder bed and the die wall by the interposition of the lubricant layer at the interface
(2) decrease of the shear strength by the low shear strength of the lubricant layer formed on the interface

On the basis of Bowden's theory,[3] Lewis *et al.*[4] studied the die wall friction of pharmaceutical powders during compaction. They measured the shear strengths of diluent and lubricant powders by the punch penetration method. The lubricants (potassium stearate, sodium stearate, magnesium stearate, *etc.*) gave lower shear strength than the diluents (sodium chloride, sucrose, *etc.*). In the same experiments, it was found that the strength of lubricant was independent of the pressure applied to the compact, while that of diluent increased with increasing pressure. Fukuda *et al.*[15] also found that the maximum shear stress of magnesium stearate (85 kg/cm^2) was much smaller than those of potassium chloride and phenacetine (535 and 360 kg/cm^2, respectively).

In the present study, the wall friction force for lubricated powders was determined. The three types of powders[7] were admixed with lubricant of 2 wt%. The lubricant reduced μ_{W} to the range of 0.04 to 0.22, and the value was little affected by the normal pressure in any lubricated system. No residual layer was observed over the whole range of pressure. Kitamori *et al.*[16] studied the lubrication effects of magnesium stearate, measuring the residual lateral force and the ejection force. In their studies, the lubricant obviously decreased the die wall friction. The value of μ_{w} for lubricated powders agreed with the values in the present study.

These experimental results can be used to interpret the results of the compression test described in the previous paper.[7] The pressure-transmission ratio from the upper to the lower punch did not always increases with increasing compaction pressure for some types of powders, such as lactose and potassium chloride, indicating that the results did not follow Janssen's equation. On the other hand, for the powders lubricated with magnesium stearate of 2 wt%, the ratio increased monotonously with the pressure, and followed the equation. As expected, the results of lubrication in the compression test were due to the low and almost constant coefficient of die wall friction for the lubricated powders.

The values of μ_{W} of lactose lubricated with talc of 2 wt% were from 0.32 to 0.36 over the whole range of pressure. These values were higher than those of lactose lubricated with magnesium stearate. The residual layer was observed at $P_{\mathrm{N}} \geqq 400$ kg/cm^2. The effects of talc on the friction and adhesion forces were lower than those of magnesium stearate. For lactose powder with talc and with magnesium stearate, Kitamori *et al.* obtained coefficients of die wall friction of 0.25 and 0.04, respectively.

Sliding Process

The sliding process was expressed by plots of the value of μ_W versus displacement. The pattern for potassium chloride, shown in the previous paper,[8] varied with the normal pressure to the sliding surface. The patterns for lactose, sucrose and sodium bicarbonate powders were different from that for potassium chloride (Figs. 3—5). Furthermore, the addition of magnesium stearate of 2 wt% made the patterns for these powders very different. The value of μ_w for lubricated powders was independent of the normal pressure. In this respect, the lubricated powders are distinct from the unlubricated powders. The mechanisms of friction and lubrication on the die wall for these powders will be discussed below, based on the sliding process.

The patterns of sliding can be classified as shown in Fig. 6. In the study on ferric oxide powders by Strijbos,[13] three types of curves, (A), (B) and (C), were observed. These curves were distinguished in terms of the changes in μ_W after the "transition point," corresponding to the transition from static to dynamic friction with sliding. Namely, in (A) μ_W decreases, while in (B) it increases after a decrease, and in (C) it increases, after this point. He found that (A) was the case of ideal sliding, (B) was the case of sliding with the fragmentation of particles, and (C) was the case of sliding with the brittle fracture of particles.

The results obtained in the present study are shown in Figs. 3 to 5. The unlubricated powders gave type (B) patterns, expect for potassium chloride and sodium chloride powders which gave type (D). On the other hand, lactose lubricated with magnesium stearate gave a typical pattern (A). In Figs. 1 and 2, the values of μ_{max} are plotted. From the response on the displacement recorder used, this maximum point can be regarded as the starting point of the macro slide between the powder bed and the wall. This maximum point corresponds to the "transition point," defined by Strijbos, and also to the point of maximum static friction.

Ideal sliding did not occur, because the surface conditions changed with sliding after the maximum point, and because μ_W sometimes showed a high value beyoned the maximum. This is, for example, the case for the unlubricated lactose, which adhered more strongly to the wall than the lubricated lactose, and which fractured more easily than potassium chloride. These experimental results are at least partially consistent with the observations on shear pattern and on sliding behavior made by Strijbos *et al.*[13] However, it is impossible to relate

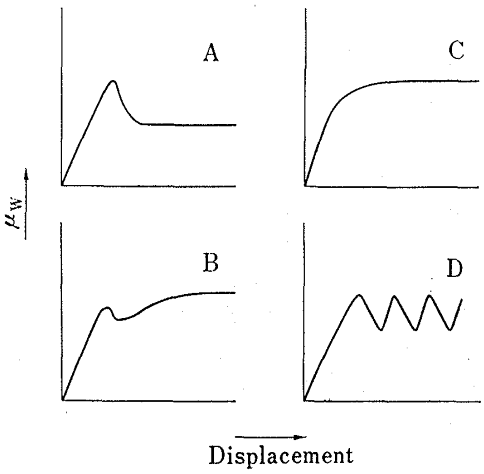

Fig. 6. Typical Models of the Sliding Process on the Die Wall

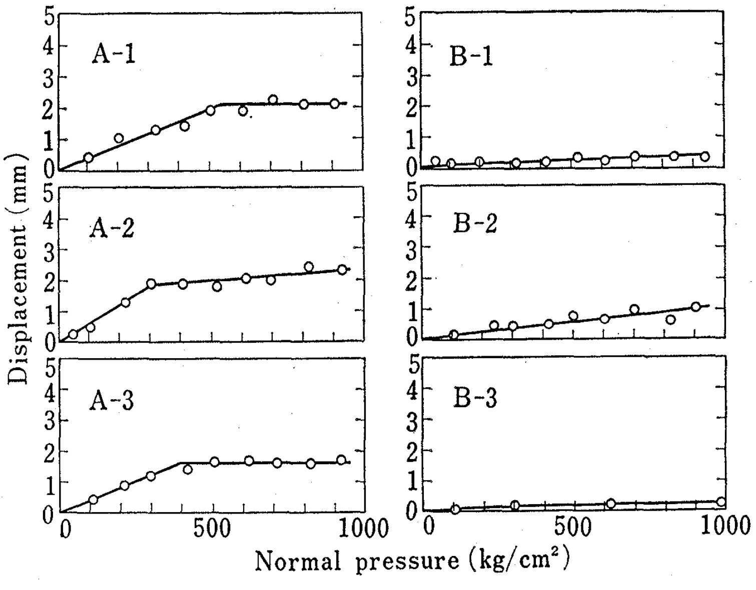

Fig. 7. Displacement to the Maximum Point

A-1: sodium bicarbonate. A-2: lactose. A-3: lactose with 2 wt% talc. B-1: sodium bicarbonate with 2 wt % magnesium stearate. B-2: lactose with 2 wt% magnesium stearate. B-3: potassium chloride with 2 wt % magnesium stearate.

the pattern to the mechanical properties of the powders used in the present study. It was also impossible to interpret in detail the characteristic pattern of the lubricated powders. Three types of sliding process were observed for three different lubricated powders. This must be due to the lubricant effect on the mechanical properties of each powder. By removing factors such as the adhesion of particles and the roughness of surfaces in the lubricated systems, the mechanical behavior of powders during sliding will become clearer.

The sliding patterns were examined in detail. Namely, the behavior in the state of static friction and immediately after the transition point was examined. The difference between the lubricated and unlubricated systems is as follows. For unlubricated powders, the displacement to the maximum point of μ_w increased with increasing normal pressure, and μ_w decreased slowly after the maximum point at every pressure. For the lubricated powders, the displacement did not depend on the pressure, and μ_W decreased rapidly. Plots of the displacement to the maximum point versus the normal pressure are shown in Fig. 7. The displacement (*dD*) was determined as the distance from the zero point where the tangential force was first applied on the apparatus to the maximum point read from the records of μ_W.

Figure 7-A shows the plots for the powders which gave sheared residual layers in the range of high normal pressure, *i.e.*, which sheared within themselves during sliding. This range, in which *dD* remained constant, was in agreement with the range in which the sheared residual layers appeared. In the range of low pressure, *dD* increased linearly with normal pressure. For lactose and sodium bicarbonate powders, the slopes were 6.0×10^{-3} and 3.7×10^{-3} mm per unit normal pressure, respectively. This difference indicates that the displacement in the static friction state is mainly affected by both the normal pressure and the mechanical strength of particles. On the other hand, for the powders lubricated with magnesium stearate (Fig. 7-B), the values of *dD* were very low in comparison with those of the A-group, and were slightly dependent on the normal pressure. The slopes were much lower than those of the A-group (1.1×10^{-3}, 4.7×10^{-4} and 2.6×10^{-4} mm per unit normal pressure for lubricated lactose, lubricated sodium bicarbonate, and lubricated potassium chloride, respectively).

Soda[17)] described the micro mechanism of the initial stage of sliding, from the application of the tangential force to the transition point to dynamic friction. According to him, tangential force causes the micro junctions to deform elastically until about 10^{-6} mm displacement, then to fracture partially, and finally to break completely at 10^{-3}—10^{-1} mm. When the displacement becomes equal to the average diameter of micro junctions, static friction passes into dynamic friction. On the basis of this concept, a larger normal pressure or a smaller mechanical strength of material results in a lager junction area, and a larger displacement. In the present study, the average particle size was about 300 μm, and the maximum displacement was about 2 mm. So it can not be accepted that the diameter of the junction between the particle and the wall coincides with the displacement. This concept can not interpret the increasing friction at the initial stage. However, Soda also stated that soft metals showed larger displacement than hard metals. This means that the mechanical properties of materials affect the displacement in the static friction state. The experimental results in the present study are in agreement with this result.

Tsuya[14)] and Bowden *et al.*[3)] described the initial stage in further detail. The application of force normal to the interface generates junctions. Next, with the application of tangential force along the interface, the junctions are deformed plastically by the combined effect of the normal and tangential stresses, resulting in increase of the junction area. When the tangential force approaches the limit under a certain condition, the junctions are cut, and the macro slide starts simultaneously. The growing stage of junctions corresponds to the stage of increasing friction force in the sliding process. At the starting point of macro slide, the junction area is maximum.

For the present frictional system between the pharmaceutical powder bed (the compact) and the metal wall, the growth of junctions is discussed below.

The photographs shown in Fig. 8 are the wall surfaces which were obtained by removing the powder beds of unlubricated lactose by hand. Figure 8-(b) is the surface obtained after the application of normal force, but in the absence of tangential force. There is only a thin layer of powders on the wall. On the other hand, Fig. 8-(a) is the surface slid by 2.8 mm on application of tangential force. On this surface, a marked residual layer can be observed. While unlubricated lactose gave a marked residual layer, the lubricated lactose did not give a residual layer even after the macro slide. These observations indicate that a stronger adhesion force occurred at the interface between the lactose powder bed and the wall after the macro slide than before the application of tangential force.

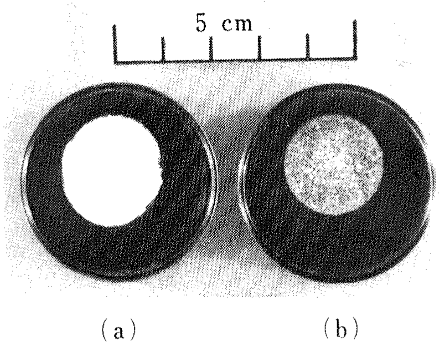

Fig. 8. Residual Layers of Lactose on the Wall

(a) after sliding, (b) before sliding.

Replicas[11] of the potassium chloride compact in contact with the wall were taken to observe the particles on the interface during sliding. Photomicrographs of them are shown in Fig. 9. Figures 9-(a), (b) and (c) are for unlubricated potassium chloride compacts, and Fig. 9-(d) is for a lubricated one. The particle boundaries are clearly discernible in (a) for the replica before the application of tangential force. In contrast, the boundaries are hardly discernible in (b) for the compact slid before the maximum point, that is, where the macro slide has not yet occurred. Furthermore, after the maximum point no boundary can be detected, as shown in (c). While (d) is the replica for the lubricated compact after the maximum point, the boundaries are discernible as in (a). These observations imply that, for unlubricated powders, the tangential force increases the particle contact area over that formed by the normal force, and that the growth of the particle contact area is substantial before the macro slide. On the other hand, for lubricated powders, the tangential force does not increase the area, even after the macro slide.

The above-mentioned observations should help to clarify the mechanisms of friction and lubrication between pharmaceutical powders and the die wall during compaction. It appears that the junctions of particles formed in the interface by the normal force increase with the tangential force. The friction and adhesion forces increase with the growth of junctions and reach the maximum. The lubricants prevent such growth, that is, they prevent the increase in friction and adhesion forces due to the tangential force. This conclusion seems to be consistent with the description of the compaction of magnesium carbonate powder by Train,[4a] *i.e.*, in the powder bed, there was a part with higher strength than the others, where the higher shear stress acted.

Accordingly, the differences in the patterns of sliding can be interpreted by considering this growth of junctions. The measured displacement may be the displacement up to the beginning of the fracture of junctions, and is larger than the average diameter of junctions[18] formed by the normal force. The displacement may be equal to the diameter of junctions after the increase due to the application of tangential force. The start of the fracture of junctions depends on the mechanical properties of the particle materials, along with the external force, the contamination on the surface and so on. In the hard-matal/hard-metal system, the growth is small due to the elastic relaxation.[3] However, in the present system between pharmaceutical powders and the metal wall, both the growth of particle junctions and the displacement to the maximum friction are large.

The transition point to the dynamic friction described by Soda[17] may correspond to the end point of the fracture of junctions. The displacement associated with the diameter of micro

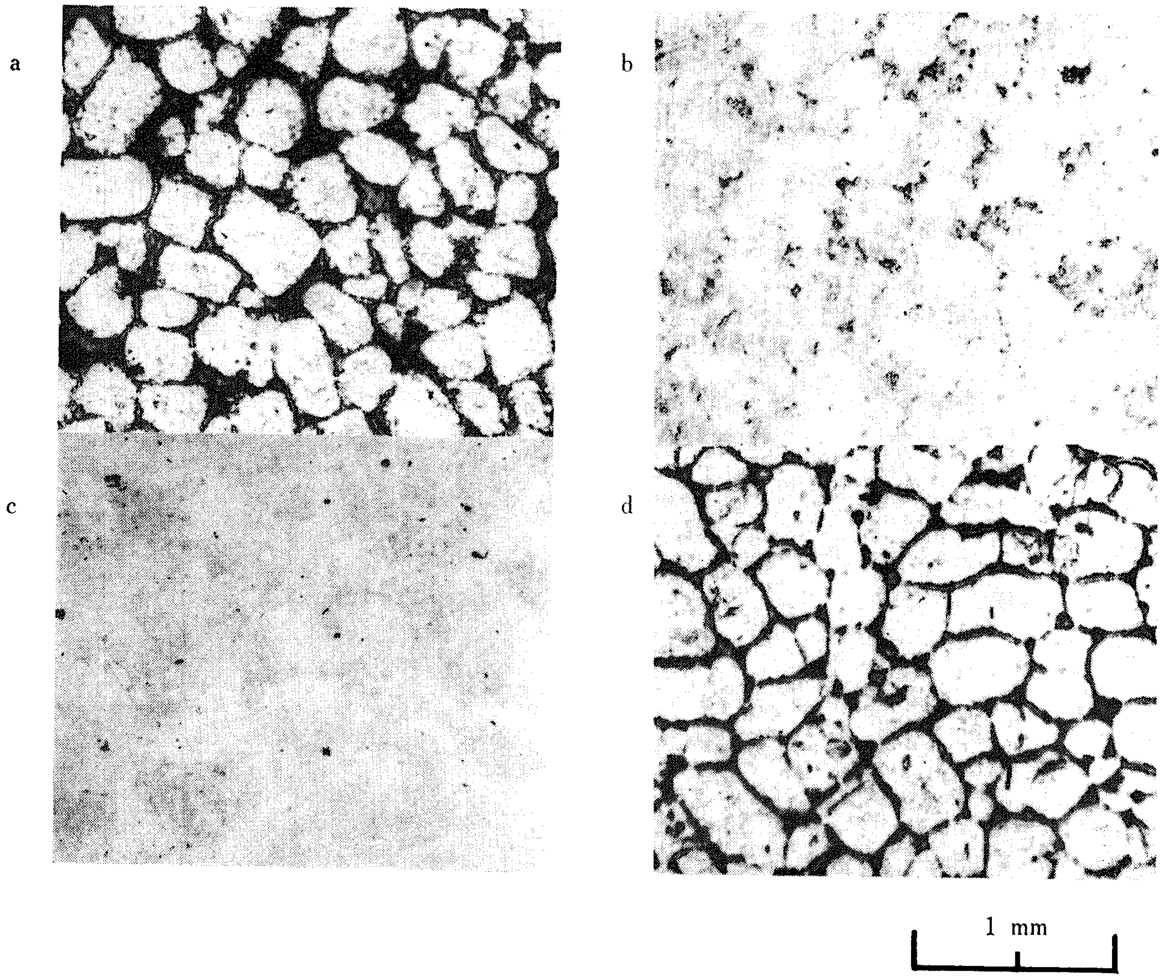

Fig. 9. Photomicrographs of Replicas of Particles on the Surface of Potassium Chloride Tablets

Unlubricated powder: a,b,c. Lubricated powder: d. Normal pressure: about 800 kg/cm^2. Sliding stages: a, displacement (dD)=0 mm (before sliding); b, dD=1.1 (after sliding and before maximum μ_W); c, dD=4.0 (after maximum μ_W); d, dD=4.0 (after maximum μ_W).

junctions by him may be the distance from the starting point to the end point of the fracture of junctions, *i.e.*, it may be the distance from the maximum static friction point to the dynamic friction state. The different behavior seen in this study after the maximum point can be explained in terms of this displacement. The more rapid decrease in μ_w in the sliding process for lubricated powders than for unlubricated powders can explained on the basis of smaller displacement.

Conclusion

The friction force between pharmaceutical powders and the metal wall was measured. The dependence of the coefficient of wall friction (μ_W) on the normal pressure to the interface (P_N) was examined. The values of μ_W for lactose and sucrose powders increased more rapidly with P_N than those for the other powders, and were high, that is, 0.45—0.60. Furthermore, for these powders, cohesive residual layers were observed on the wall surface.

The addition of magnesium stearate of 2 wt% to the powders decreased μ_W to about 0.2 or below. For the lubricated powders, no significant dependence of μ_W on P_N was noted, and no residual layer was discernible.

The die wall friction and the dependence on the normal pressure are apparently related to the mechanical properties of particles and the compaction properties of the powder bed.

The sliding process could be expressed by plots of μ_W versus the displacement in the sliding direction. The sliding patterns for lactose, sucrose and sodium bicarbonate powders were different from those for potassium chloride and sodium chloride powders. The pattern for lubricated powders was also different from that for the unlubricated powders. The mechanisms of friction and lubrication may be related to these sliding patterns.

The residual layer on the wall surface and the replicas of particles on the surface of the compact at each stage of sliding were examined. The examinations showed that the particle junctions grew, and that the growth was associated with the friction and adhesion forces. It was also found that lubricant prevented the growth and, consequently, decreased the friction and adhesion forces. The growth was correlated with the sliding process while the coefficient of wall friction transited from the static to the dynamic friction state through the maximum point of static friction.

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