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Internal Friction of Compressed Pharmaceutical Powders observed in Terms of the Die Wall Pressure

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The internal friction of powders was examined during tablet preparation for lactose and phenacetin as base materials and for potassium chloride, potassium bromide and sodium chloride as model samples.

The coefficient of internal friction decreased with the porosity, even if the voids were filled by fragmentation of particles or by plastic flow. When the porosity had reached zero, the state of the whole bed of powder was found to be plastic, where the coefficient of internal friction was zero, for magnesium stearate, phenacetin, potassium bromide and potassium chloride. The plastic behavior of magnesium stearate and phenacetin was considered to be related to the "capping" of tablets containing them.

For lactose granules, the porosity did not reach zero even at a compacting pressure of 4000 kg/cm^2, the maximum used in this work, because of its crystal hardness. The coefficient of internal friction was large and decreased only gradually with the porosity.

Mixtures of two components exhibited intermediate internal friction, corresponding to the composition. However, when hydroxypropyl cellulose (HPC) was added to phenacetin powder as a binder, HPC increased the yield stress from 2000 to 3100 kg/cm^2 even at a content of only 2% w/w on a dry basis. In contrast, lubricants (magnesium stearate and talc) added to granules had little or no effect on the internal friction, although this small amount was sufficient to reduce the wall friction.

Keywords——internal friction of powders; tablets; plastic flow of solid; potassium chloride; potassium bromide; sodium chloride; lactose; phenacetin; binders; lubricants

Introduction

The compaction of powder druing tabletting was studied on the basis of powder mechanics by Fukumori and Okada.[2] They reported that the fundamental properties of compressed powders were the powder yield locus (internal friction), the wall yield locus (die wall friction) and the one-dimension consolidation curve. With the potassium chloride powder, these properties were measured at each maximum principal stress below 1000 kg/cm^2. The internal friction was evaluated from the die wall pressure. In pharmaceutical technology, the die wall pressure has often been examined in connection with "pressure cycles" or the die wall friction.[3] These results should be useful in powder mechanics studies in connection with the internal friction of powders.

Most studies involving additives to tablets are concerned with the properties of the final products. However, their functions in the compacting (loading) process should also be considered. For example, a binder may act in the compacting process not as a "binder" but as a lubricant because of its softness.

1) Location: *Arise, Igawadani-machi, Tarumi-ku, Kobe.*
2) Y. Fukumori and J. Okada, *Chem. Pharm. Bull.*, **25**, 1610 (1977).
3) *a*) J.J. Windheuser, J. Misra, S.P. Eriksen, and T. Higuchi, *J. Pharm. Sci.*, **52**, 767 (1963); *b*) T. Higuchi, T. Shimamoto, S.P. Eriksen, and T. Yashiki, *ibid.*, **54**, 111 (1965); *c*) K. Ridgway, J. Glasby, and P.H. Rosser, *J. Pharm. Pharmac.*, **21**, 24S (1969); *d*) J.E. Carless and S. Leigh, *ibid.*, **25**, 289 (1973); *e*) E. Shotton and B.A. Obiorah, *J. Pharm. Sci.*, **64**, 1213 (1975); *f*) M.P. Summers, R.P. Enaver and J.E. Carless, *J. Pharm. Pharmac.*, **28**, 89 (1976); *g*) E. Doelker and E. Shotton, *ibid.*, **29**, 193 (1977).

The purpose of the study reported here was to examine the internal friction of compressed powder in terms of the die wall pressure. The compacting pressure (the maximum principal stress) was raised to 4000 kg/cm^2 to clarify the plasticity of the pharmaceutical solids under high pressures. The effects of additives on the internal friction were also examined.

Experimental

Sample powders were phenacetin, lactose, potassium chloride (KCl), potassium bromide (KBr), sodium chloride (NaCl), magnesium stearate, talc and hydroxypropyl cellulose (HPC): commercial crystalline powders were used without making their particle sizes uniform. Phenacetin tablets readily show "capping." Lactose powder forms relatively hard tablets and has often been used as a diluent. KCl, KBr and NaCl were taken as model samples for which plastic deformation of the particles, followed by bonding of adjacent surfaces, has been described.[4)]

Granules of phenacetin and of lactose were prepared following the flow chart shown in Fig. 1. The method is an ordinary wet granulation procedure. HPC used as a binder was first mixed in as a powder and then kneaded with water. However, in granules with 2% w/w of HPC calculated on a dry basis, HPC was added as an aqueous solution. The powder composed of phenacetin alone was kneaded with ethanol. The granules of plain phenacetin were prepared with a disintegrating granulator (speed mill; Okano Seiko Co., Tokyo, Japan) and all the others were prepared with an extrusion granulator (pelleter; Fuji Paudaru, Osaka, Japan). Wet granules were dried to a water content of less than 1% w/w. Dried granules were sieved with standard sieves. Granular samples were mainly the 20—200 mesh fractions, but for lactose the 20—32 and the 50—80 mesh fractions were also used. The difference in the fraction size had no effect on the die wall preseure. Magnesium stearate or talc was mixed into the dried granules as a lubricant as required.

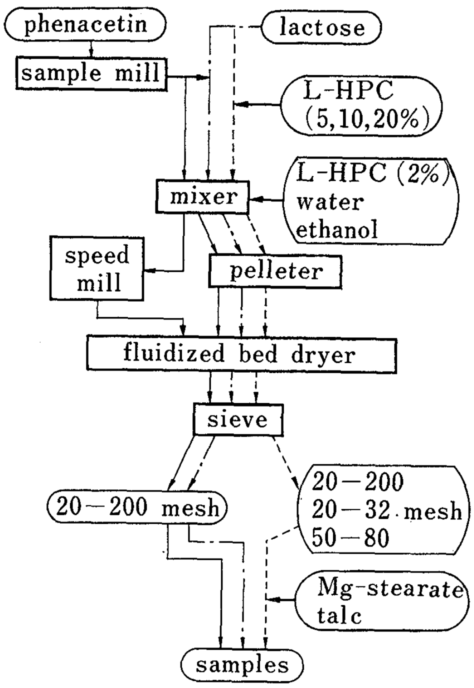

Fig. 1. Flow Chart of the Preparation of Granules

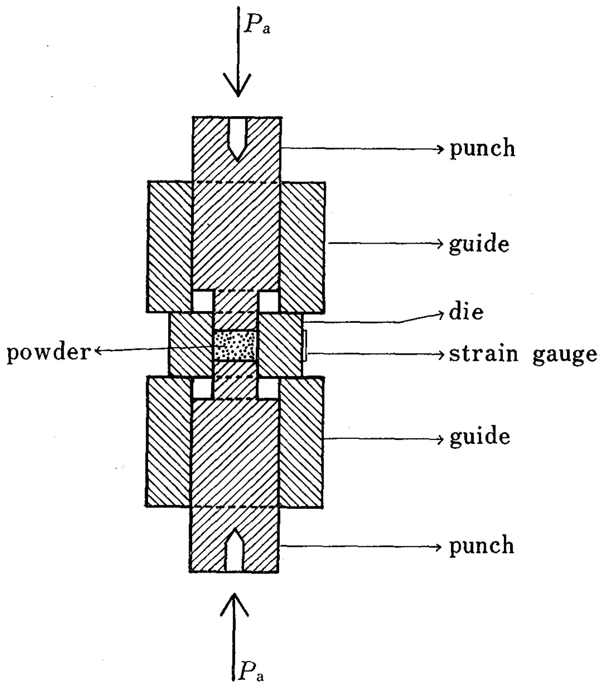

Fig. 2. Die and Punch Assembly

A diagram of the apparatus for measuring the die wall preseure is shown in Fig. 2. The die was of 10 mm inner diameter and was lubricated with magnesium stearate powder. The amount of powder or granules which formed a bed of 5 mm thickness at zero porosity was poured into the die and was compressed from both sides with flat punches at a constant velocity of 0.45 mm/min. Under such conditions, the compacting

4) *a*) J.A. Hersey and J.E. Rees, *Nature* (London), *Phys. Sci.*, **230**, 96 (1971); *b*) J.S. Hardman and B.A. Lilley, *ibid.*, **228**, 353 (1970); *c*) J. Okada and Y. Fukumori, *Chem. Pharm. Bull.*, **22**, 493 (1974); *d*) E.T. Cole, J.E. Rees, and J.A. Hersey, *Pharm. Acta Helv.*, **50**, 28 (1975); *e*) A.H. DE Boer, G.K. Bolhuis, and C.F. Lerk, *Powder Technol.*, **20**, 75 (1978); *f*) H. Hess, *Pharm. Technol. Intern.*, **1**, 18 (1978).

preseure and the die wall pressure can be regarded as the maximum σ_1 and the minimum principal stress σ_3, respectively.[5] The thickness of the bed of powder was measured with differential transformers. The output readings of strain gauges bonded to the die were converted to values corresponding to the 5 mm bed thickness. The gauges on the die wall were calibrated by compressing a silicon rubber plug.[3c]

Results

Fig. 3 shows the die wall pressure P_r plotted against the compacting pressure P_a for KCl, KBr, and NaCl powders. The corresponding Mohr's envelopes are shown in Fig. 4, where σ and τ are the normal and the shear stress, respectively. With KCl and KBr, the slope of the curve in Fig. 3 reaches unity at high pressure and then the Mohr's envelope becomes parallel to the σ axis. Hence, the material can then be regarded, at least from a mechanical point of view, as being in the plastic state. The extrusion of material from the gap between the die and the punch was then observed.

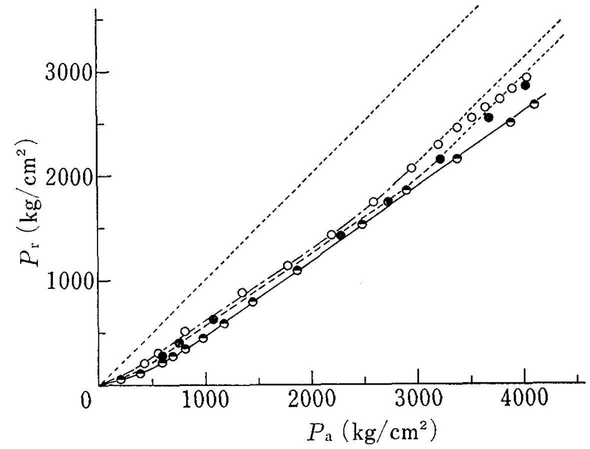

Fig. 3. Die Wall Pressure exerted by KBr, KCl and NaCl Powders

○ : KBr; ● : KCl; ◓ : NaCl; ---- : $P_r = P_a$.

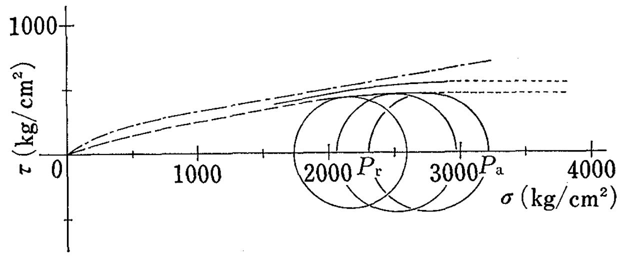

Fig. 4. Mohr's Envelopes in Terms of Die Wall Pressure and Compacting Pressure for KBr, KCl and NaCl

---- : KBr; ——— : KCl; –·–·– : NaCl.

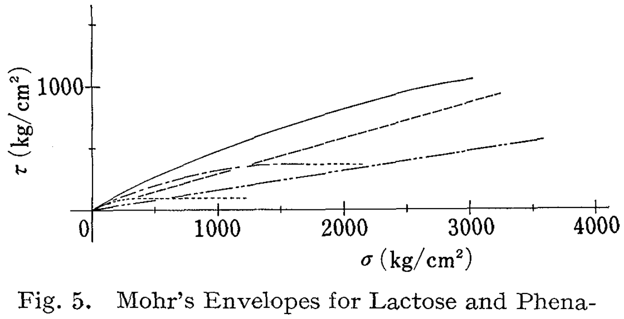

Fig. 5. Mohr's Envelopes for Lactose and Phenacetin Granules and for Magnesium Stearate, HPC and Talc Powders

——— : lactose; –·–·– : phenacetin;
——— : magnesium stearate;
——— : HPC; ---- : talc.

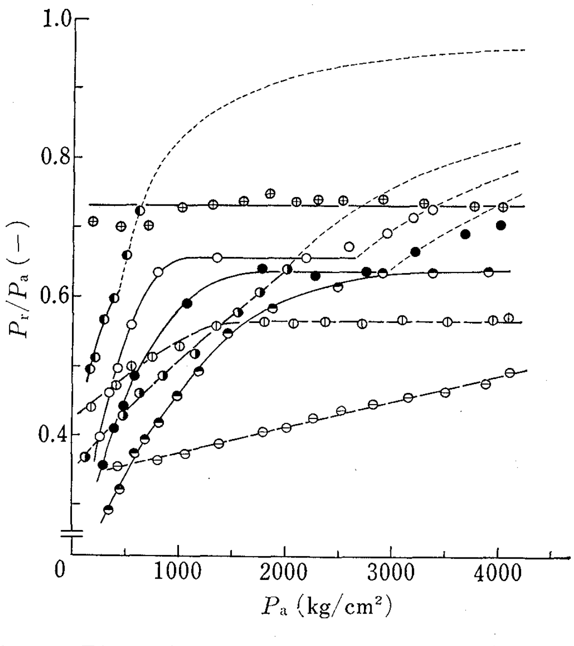

Fig. 6. Plots of Stress Ratio P_a/P_r against the Compacting Pressure

○ : KBr; ● : KCl; ◓ : NaCl; ⊖ : lactose; ◑ : phenacetin; ◐ : magnesium stearate; ⊕ : HPC; ⦶ : talc.

5) Y. Aketa and H. Tsuwa, *Technol. Reports Osaka Univ.*, **18**, 489 (1968).

Fig. 5 shows the Mohr's envelopes for lactose, phenacetin, magnesium stearate, HPC and talc. The plastic state is also apparent for phenacetin and magnesium stearate. Lactose granules exhibit the greatest internal friction.

In Fig. 6. P_r/P_a is plotted against P_a. In the ideal plastic state, P_r/P_a can be expressed by Eq. 1,

$$P_r/P_a = 1\text{–}2S/P_a \qquad (1)$$

where S is the maximum shear stress. The dotted hyperbolic lines in Fig. 6 correspond to the ideal plastic states. The measured values of P_r/P_a deviate from these lines because of the extrusion of material from the gap between the die and the punch. The values of P_a at the yield point (the yield stress) and the maximum shear stress are shown in Table I. The maximum shear stress was calculated by substituting the values of P_a and P_r at the yield point into Eq. 1.

For KBr, KCl, NaCl and talc powders, P_r/P_a initially increases with P_a and then becomes constant, as can be seen in Fig. 6. In the case of lactose, P_r/P_a increases only linearly. HPC powder takes a constant value of P_r/P_a over the whole pressure range under study.

Table I. Powders Exhibiting Plastic Flow

Material	Yield stress (kg/cm^2)	Maximum shear stress (kg/cm^2)
KCl	3000	535
KBr	2650	455
Phenacetin	2000	360
Magnesium stearate	440	85

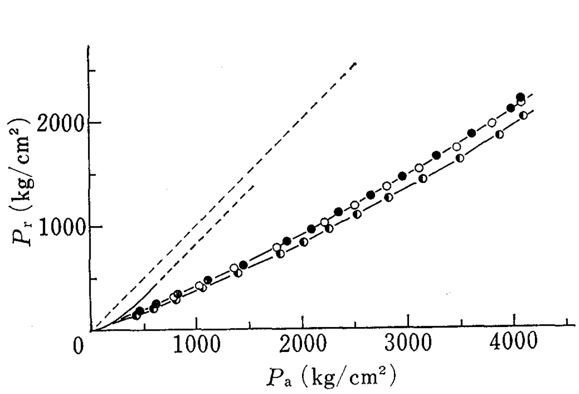

Fig. 7. Die Wall Preseure exerted by Lactose Granules (20—32 mesh) admixed with Magnesium Stearate Powder

◐ : plain lactose granules;
content of magnesium stearate (%):
○ : 2; ● : 5.
———— : plain magnesium stearate.

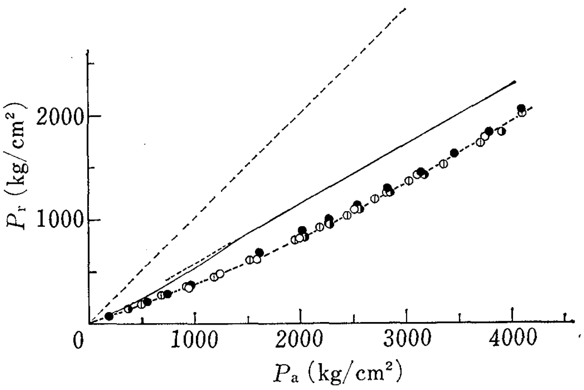

Fig. 8. Die Wall Pressure exerted by Lactose Granules (20—32 mesh) admixed with Talc Powder

---- : plain lactose granules;
content of talc (%):
● : 2; ○ : 5, ⦶ : 10; ◑ : 20;
—— : plain talc.

Fig. 7 and 8 show the results for lactose granules (20—32 mesh fraction) admixed with magnesium stearate and talc powder, respectively. With the magnesium stearate powder, P_r took about 1.1 times larger values, but the addition of talc powder had no effect on P_r. Fig. 9 shows that P_r exerted by phenacetin granules is not affected by the presence of either magnesium stearate or talc.

Fig. 10 and 11 show the effects of incorporated HPC on P_r for lactose and phenacetin granules, respectively. For lactose, the greater the content of HPC, the larger the value of P_r over the whole pressure range. That is, the internal friction is reduced by HPC, which gives a larger die wall pressure than lactose. On the other hand, Fig. 11 shows that for the phenacetin granules both the yield stress and the maximum shear stress are increased by the addition of HPC even at a level of 2%. With increase in HPC content, P_r has a tendency to increase at low pressures and to decrease at high pressures. As a result, P_r exerted by the granules approaches that of plain HPC powder.

Fig. 12 shows the results for granules prepared from a mixture of lactose powder with phenacetin powder. It appears that the lactose powder is lubricated by phenacetin, which exerts a larger die wall pressure than lactose.

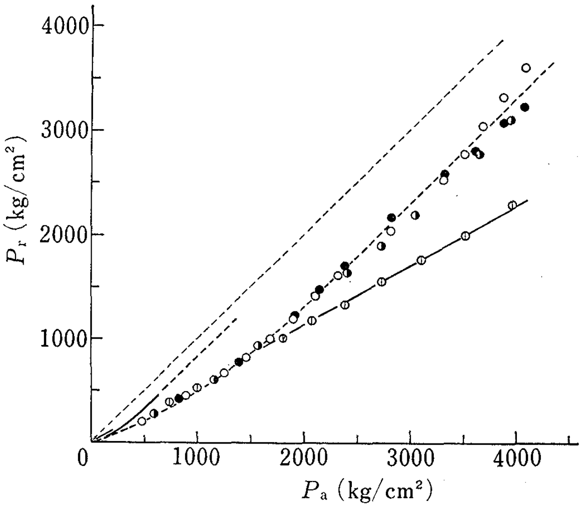

Fig. 9. Die Wall Pressure exerted by Phenacetin Granules admixed with Magnesium Stearate or Talc Powder

---- : plain phenacetin granules
content of magnesium stearate (%): ● : 2; ○ : 5;
content of talc (%): ◑ : 10;
-------- : plain magnesium stearate;
—⦶— : plain talc.

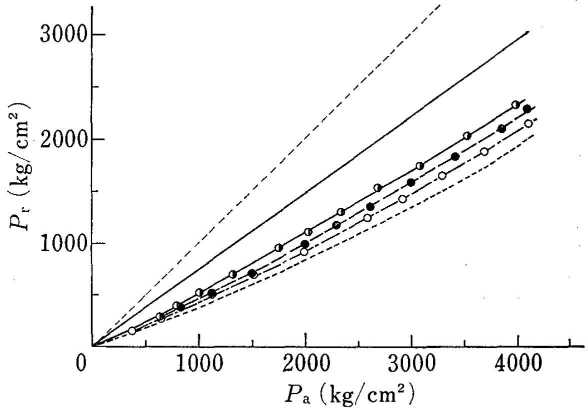

Fig. 10. Die Wall Pressure exerted by Lactose Granules Containing HPC

---- : plain lactose granules;
content of HPC (%): ◑ : 20; ● : 10; ○ : 5;
—— : plain HPC.

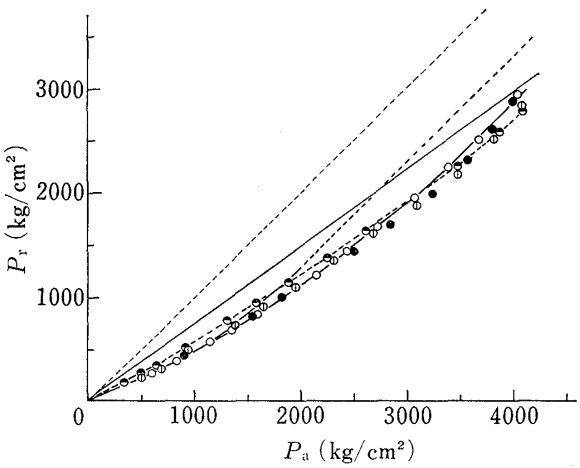

Fig. 11. Die Wall Pressure exerted by Phenacetin Granules Containing HPC

-------- : plain phenacetin granules;
content of HPC (%): ○ : 2; ● : 5; ⦶ : 10; ◓ : 20;
—— : plain HPC.

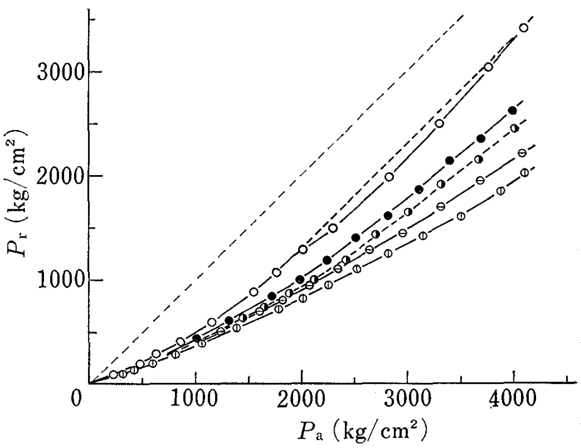

Fig. 12. Die Wall Pressure exerted by Granules prepared from Mixtures of Lactose with Phenacetin

—⦶— : plain lactose granules;
—○— : plain phenacetin granules;
weight ratio (lactose: phenacetin):
● : (2: 8); ◑ : (4: 6); ⊖ : (6: 4).

Discussion

Plastic behavior of KCl and KBr powders had already been observed in compaction at a constant compacting pressure lower than 1000 kg/cm[2,4c,6] From a rheological viewpoint, it is clear that when compressed at constant velocity (0.45 mm/min), a powder becomes plastic at high pressure. It has often been observed that plastic deformation of KCl or KBr particles can take place even at relatively low pressures in the neighborhood of the surface of contact.[4c] Hess[4f] observed by scanning electron microscopy that KCl exhibited almost ideal plastic behavior. Boer *et al.*[4e] reported a similar plastic deformation for NaCl. However, it was observed by the present authors that NaCl particles were more frequently fragmented than KCl and KBr particles. This may arise from the higher yield stress and maximum shear stress in NaCl, as can be estimated from Fig. 6. In addition, it is noteworthy that the whole bed of powder begins to behave plastically at 3000 kg/cm² for KCl and at 2650 kg/cm² for KBr (Table I).

Magnesium stearate powder had a remarkably low maximum shear stress: 85 kg/cm². The function of magnesium stearate as a lubricant arises from this low shear strength. Phenacetin powder also had a relatively low maximum shear stress: 360 kg/cm². These findings are clearly related to the "capping" of phenacetin tablets or tablets containing large amounts of magnesium stearate.

In contrast to KCl and KBr, phenacetin powder was not reported to show plastic behavior in compaction under constant compacting pressure.[6] It can therefore be considered that the particles are brittle and elastic even at the point of contact, at least until the stress reaches the yield point. As regards the state of the yielding bed of powder, it is possible that slippage may take place on the boundary of small crystalline particles, or that the material may be entirely amorphous. If the latter is correct, the addition of a small amount of binder would have no effect on the yield point. As can be seen in Fig. 11, as little as 2% HPC increases the yield stress and the maximum shear stress to about 3100 and 550 kg/cm², respectively. Clearly, the small amount of binder is effective only at the grain boundaries in a polycrystalline structure. Experimentally, phenacetin compacts compressed up to 4000 kg/cm² remained opaque, while KCl, KBr, and magnesium stearate compacts became fairly transparent. X-ray crystallographic observations will be reported in the following paper.

Fig. 6 shows that HPC powder behaves as an elastic body. P_r/P_a is not only constant over the whole range, but also took almost equal values in the unloading process. The constant value of P_r/P_a for HPC powder is large and hence Poisson's ratio is also large. In addition, the residual die wall pressure was so low as to be indetectable. This means that HPC is also effective for the reduction of ejection force. For KBr, KCl, NaCl, and talc, after the porosity had reached zero, P_r/P_a took a constant value (Fig. 6). Clearly, then, these powders were also in the elastic state as a whole. These compressed powders might take a polycrystalline structure composed of small elastic particles.

With the lactose granules, the porosity did not reach zero even at 4000 kg/cm², probably because the lattice structure of lactose crystals does not permit any plastic deformation[4f] and the crystal hardness is very large.[3c] In contrast, talc crystals have a laminar structure, undergoing fragmentation easily.[7] The easy fragmentation of talc particles is also supported by the findings that the porosity had already become zero at 1500 kg/cm², and that P_r/P_a took a large value, close to that for magnesium stearate, at low pressures where the bed of powder had enough void volume to be filled by fragmentation (Fig. 6). Hence, talc can act as an efficient lubricant, but is not effective in the compacting process where the porosity of the compact is low.[7] This may be related to the fact that the bed of talc powder remains elastic at pressures above 1500 kg/cm², as can be seen in Fig. 6.

6) J. Okada and Y. Fukumori, *Chem. Pharm. Bull.*, **23**, 326 (1975).
7) D. Train and J.A. Hersey, *J. Pharm. Pharmac.*, **12**, 97T (1960).

The results shown in Fig. 3 essentially agree with those reported by Ridgway *et al.*[3c] Althought they stated that the force transmitted to the die wall was directly proportional to the compacting pressure, such linearity should be valid only in a narrow range of pressure. Fig. 6 shows that P_r/P_a varies with the porosity of the bed of powder. For KCl powder, it was constant below about 150 kg/cm^2, where the bed of powder had enough void volume to be filled by rearranged particles.[2] In this study, P_r/P_a was constant only in the case where the powder containing no void volume remained elastic (Fig. 6).

The addition of lubricant to tablet formulations is important mainly for the reduction of wall friction. The wall friction is dominated by both the coefficient of wall friction and that of internal friction. The reduction of internal friction, which should arise from the addition of lubricant, results in an increase in the die wall pressure. As a result, the lubricant also acts to increase the friction force on the die wall. However, Figs. 7—9 show that magnesium stearate and talc have little or no effect on the internal friction. This is because only a small quantity of lubricant was added. The small quantity is sufficient, however, to decrease the wall friction force, if added to large particles such as granules. Sufficient lubricant can then be supplied to the die wall

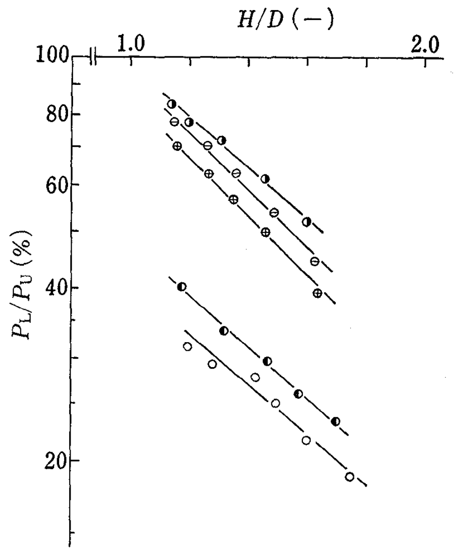

Fig. 13. Effects of Admixed Magnesium Stearate on the Pressure Ratio P_L/P_U for Lactose Granules (50—80 mesh) compressed only from the Upper Side

Weight of compressed powder (g): 9.847; thickness of bed at zero porosity (mm): 20; content content of magnesium stearate (% w/w): ○ : 0; ◐ : 0.1; ⊕ : 0.5; ⊖ : 1.0; ◑ : 5.0.

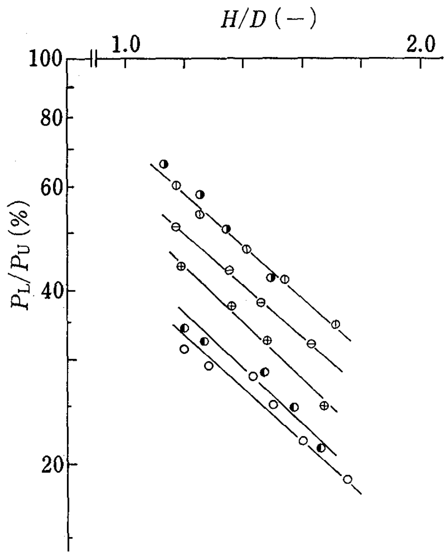

Fig. 14. Effects of Admixed Talc on the Pressure Ratio P_L/P_U for Lactose granules (50—80 mesh) compressed only from the Upper Side

For details and symbols, see Fig. 13.

Fig. 13 and 14 show the axial transmission of pressure in one-sided compression. P_L and P_U are the lower and the upper punch pressures, respectively. The maximum of P_U was about 1000 kg/cm^2. H is the thickness of the bed of powder and D is the inner diameter of the die: 20.02 mm. Each line corresponds to one compression experiment for lactose granules with each content of lubricant. In contrast to P_r, the axial transmission of pressure is greatly affected by the addition of a small amount of magnesium stearate or talc. A small amount is sufficient to lubricate the die wall surface.

The addition of HPC to lactose granules had an effect corresponding to the amount (Fig. 10). Similar results were obtained for mixtures of lactose granules with phenacetin granules (Fig. 12). For phenacetin granules incorporating HPC, P_r also varied with the composition at pressures below the yield stress (Fig. 11). These are cases where the mixture had an intermediate internal friction, corresponding to the composition. The state in which each component is distributed would be important. It is expected that when a random mixture is achieved, the mixture would exhibit an intermediate internal friction. The relatively poor effects of magnesium stearate and talc on the internal friction arose because a random mixture was not achieved as a result of the difference in particle size between the components.

Conclusions

The coefficient of internal friction decreased with the porosity, even if the voids were filled by fragmentation of the particles or by plastic flow. After the porosity had reached zero, the state of the whole bed of powder became plastic, where the coefficient of internal friction was zero, for magnesium stearate, phenacetin, potassium bromide and potassium chloride, whose yield stresses were 440, 2000, 2650, and 3000 kg/cm^2, respectively. The plastic behavior in magnesium stearate and phenacetin can be considered to be related to the "capping" of tablets containing them. In the case of sodium chloride, the yield point was not found at any compacting pressure up to 4000 kg/cm^2, the maximum used in this work.

The force transmitted to the die wall was directly proportional to the compacting pressure when the bed of powder behaved as an elastic body, *i.e.*, with potassium chloride, potassium bromide and sodium chloride with no void compressed below the yield point, with talc at zero porosity and with HPC in the whole range of compacting pressure.

Lactose particles were difficult to fragment. Hence, the porosity did not reach zero even at 4000 kg/cm^2. The coefficient of internal friction was large and decreased only gradually with the porosity. In contrast, talc powder showed low internal friction at low pressures, so that talc acts as an effective lubricant.

Mixture of two components exhibited intermediate internal friction, corresponding to the composition. However, when HPC was added to phenacetin powder as a binder, HPC increased the yield stress remarkably even at a content of only 2% w/w on a dry basis. In contrast, the addition of lubricants, magnesium stearate and talc, to granules had little or no effect on the internal friction, although this small amount was sufficient to reduce the wall friction. HPC added to lactose powder as a binder lubricated the lactose in the compacting process.