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Compaction of Powders. I. Compression under the Constant Upper Punch Pressure and Behavior of Particle Filling in Void

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To compare the compaction behavior of potassium chloride and lactose powder of 42 to 65 mesh, they were compressed under the constant pressure of upper punch (about 1000 kg/cm^2 in the maximum) by compaction apparatus with a cylindrical die (8.02 mm inside diameter), in which the powder of constant net volume (0.7545 cm^3) was filled. During the compression continued for ten hours, the thickness of powder bed and the pressure transmitted to lower punch were measured. And then the replicated surfaces of compacts were observed by microscope so as to examine the packing structure.

In the case of potassium chloride powder, under the lower pressure than the critical pressure which exists between 391 and 625 kg/cm^2, particles seem to be deformed plastically in appearance. Under the higher pressure than the critical pressure, many fragments or cracks are produced and these small fragments move to fill up voids very closely, as if original particles were deformed plastically. Therefore the values of the porosity of powder bed and the ratio of lower to upper punch pressure approach to zero and 100%, respectively.

In the case of lactose powder, as particles are brittle, they are fractured and small voids are filled with the fragments. The thickness and the ratio take the almost constant values after about five hours in spite of considerable amounts of void left still.

Introduction

The main process of tablet formation is the compression of powders in the cylindrical die, the especially important problems of which are the relationship between the upper punch pressure (in this paper, we deal with the compression from one side by upper punch) and the thickness (apparent volume) or porosity of powder bed, and the transmission of pressure to the lower punch.

Up to this time, as to the relationship between the upper punch pressure and the porosity of powder bed at the compression from one side, Kawakita's,[2] Athy's[3] experimental equation and so on[2] have been used generally to make the data fit, and the correspondence to mechanisms of the volume change or the filling behavior in void has been examined. For the filling behavior in voids, Cooper and Eaton[4] proposed three behaviors: filling of large voids by rearrangement of particles, filling of small voids by fragmentation and plastic flow. But they didn't confirm those filling behaviors experimentally. However, it will be very useful to decide the filling behaviors in voids for various pharmaceutical powders in order to choose the appropriate powders which may give the necessary hardness and the adequate disintegration time to tablets. The differences of filling behaviors in voids proposed by Cooper and Eaton depend on the mechanical properties of powders. Particularly, in the case of powder which may flow plastically, the apparent volume of powder bed will change according to the compressing time under the constant upper punch pressure.

Generally, the apparent volume of powder bed compressed by the upper punch is determined not only by mechanical properties of material, but also by the transmission of pressure

1) Location: *Yoshida-Shimoadachi-cho, Sakyo-ku, Kyoto.*
2) K. Kawakita and K.H. Lüdde, *Powder Technol.*, 4, 61 (1970).
3) L.F. Athy, *Bull. Am. Assoc. Petrol. Geologists*, 14, 1 (1930).
4) A.R. Cooper and L.E. Eaton, *J. Am. Ceram. Soc.*, 45, 97 (1962).

into the bed of powder. As the transmission of pressure to the lower punch is a particularly important problem, it was investigated by Janssen,[5] Mori[6] and so on.[7] Although they dealt with the equilibrium state or the critical state of stress, the changes of pressure transmitted during the process up to those states must come also into consideration in tablet formation. For example, Higuchi, *etc.*,[8] emphasized the complicated relationship between the upper and the lower punch pressure during compression of various powders.

We compressed the powders of potassium chloride and lactose at the constant rate of displacement of upper punch and measured both punch forces and the thickness of powder bed. Further, we continued the compression under the constant load of the upper punch for ten hours, and measured the changes of lower punch pressure and the thickness of powder bed. After the compression, the replicated surfaces of the compacts were examined by microscope. From these experimental results and the models proposed by Cooper and Eaton, the differences of filling behaviors in small voids for potassium chloride and lactose were made clear, and also as to the transmission of pressure to lower punch some comments for Janssen's equation were discussed.

Experimental

1. **Preparation of Powders**——Two crystalline powders were employed: potassium chloride and lactose. Commercial powders were crushed by a pestle and a mortar, and then screened. The fraction of 42 to 65 mesh was left for a week at least in the room where temperature and relative humidity were maintained almost constant: $25\pm0.5°$ and $60\pm2\%$, respectively.

The reason why we chose these two materials, is as follows: potassium chloride powder has better compressibility than lactose powder, on the other hand the former seems to be harder than the latter when they are crushed in mortar.

2. **Specific Gravity of Material**——The specific gravities of potassium chloride and lactose were measured by Beckmann's air comparison pycnometer. The measured values were 1.988 and 1.537 g/cm³, respectively.

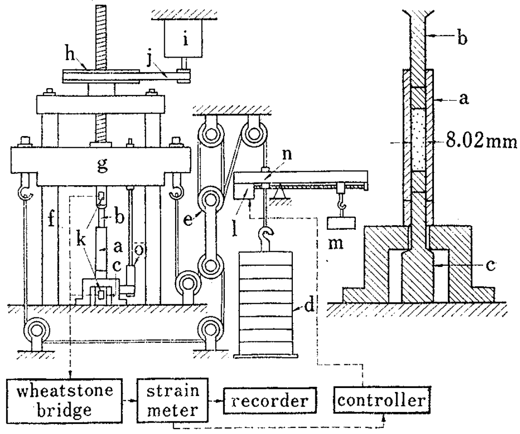

Fig. 1. Compaction Apparatus

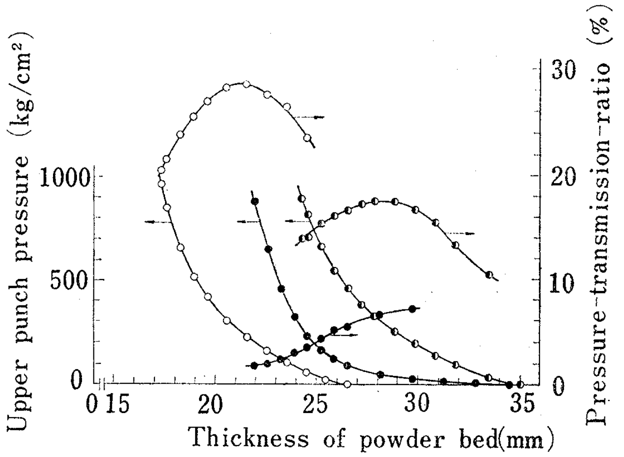

Fig. 2. Relationship between the Thickness of Powder Bed and Upper Punch Pressure or Pressure-Transmission-Ratio (the Ratio of Lower to Upper Punch Pressure) when compressed at the Constant Rate of Displacement of Upper Punch Equal to 0.332 mm/sec

Symbol	Material	Powder weight (g)	Net volume (cm³)
○	potassium chloride	1.500	0.7545
◐	potassium chloride	2.000[a)]	1.0060
●	lactose	1.158[a)]	0.7545

a) the thickness of powder bed before the compression: about 35 mm

3. **Compaction Apparatus and Procedure**——Fig. 1 shows the schematic diagram of compaction apparatus made in our laboratory and the details of the die and punches. The cylindrical die a (8.02 mm inside

5) R. Aoki, "Funtai Kogaku Handbook," ed. by K. Iinoya, Asakura-shoten, Tokyo, 1958, p. 109.
6) Y. Mori, "Shin Kagaku Kogaku Koza IV-1," Nikkan-kogyo-shinbunsha, Tokyo, 1958, pp. 37—53.
7) S. Miwa, "Funryutai Kogaku," Asakura-shoten, Tokyo, 1972, p. 263.
8) E. Nelson, S.M. Nagvi, L.W. Busse, and T. Higuchi, *J. Am. Pharm. Assoc.*, 43, 596 (1954).

diameter, 20 mm outside diameter and 80 mm length) was previously washed with benzene and ethanol in Soxhlet's extractor in order to remove stains on the surface, dried in a current of dry air, and left overnight in the room maintained at the constant temperature and humidity (25° and R. H. 60%, respectively). The powder is poured into this die a and compressed by the upper punch b at the fixed state of lower punch c. The weight d is 100 kg in the maximum. This load is multiplied by about five times with pulleys e and is transmitted to the ram g sliding on guides f. The ram g is held by a pulley h before the operation. When the pulley h is rotated at the constant speed (29 rpm) by a motor i and a belt j, the upper punch b fixed to the ram g comes down until the upper punch force becomes equal to the load given by d, and then the bed of powder is compressed under the constant upper punch pressure for ten hours. The upper and the lower punch force are measured by wire resistance strain gauges k fixed on punches (four gauges method). Further, as the upper punch force is not kept constant on account of the friction of pulleys, the output voltage of strain meter is adjusted to the required level by giving a small load to the wire rope at the point n. This small load is given by the balance weight m which can be moved by a controller and a motor l.

The displacement of upper punch is measured by a differential transformer o calibrated previously by cathetometer.

Result

1. Compression at the Constant Rate of Displacement of Upper Punch

Fig. 2 shows the relationship between the thickness of powder bed and the upper punch pressure or the ratio of the lower to the upper punch pressure (written hereafter by pressure–

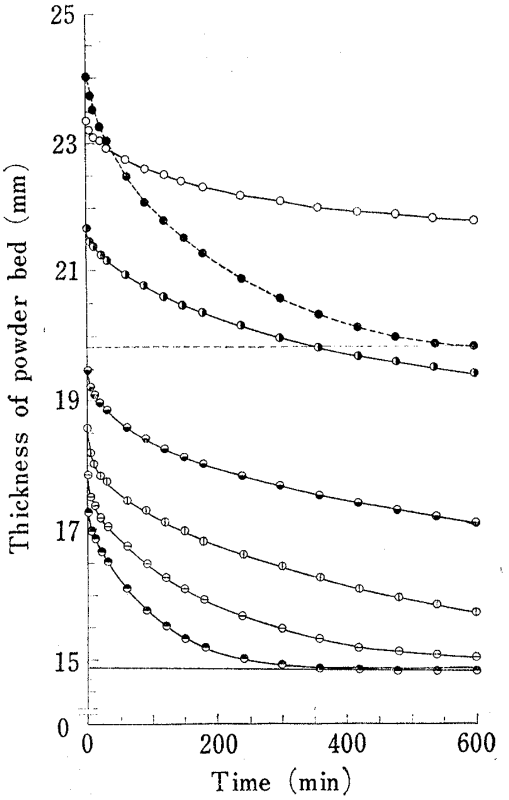

Fig. 3. Relationship between the Thickness of Powder Bed and Compressing Time when Potassium Chloride Powder is compressed under the Constant Upper Punch Pressure

powder weight (g): ——; 1.500, ······; 2.000 upper punch pressure (kg/cm²):
—○—; 83, —◑—; 183, —◒—; 391, —⦶—; 625, —⊖—; 813, —◓—; 983, ···●···; 1017
the thickness of powder bed (mm) at porosity equal to zero (shown by horizontal line): ——; 14.87, ······; 19.83

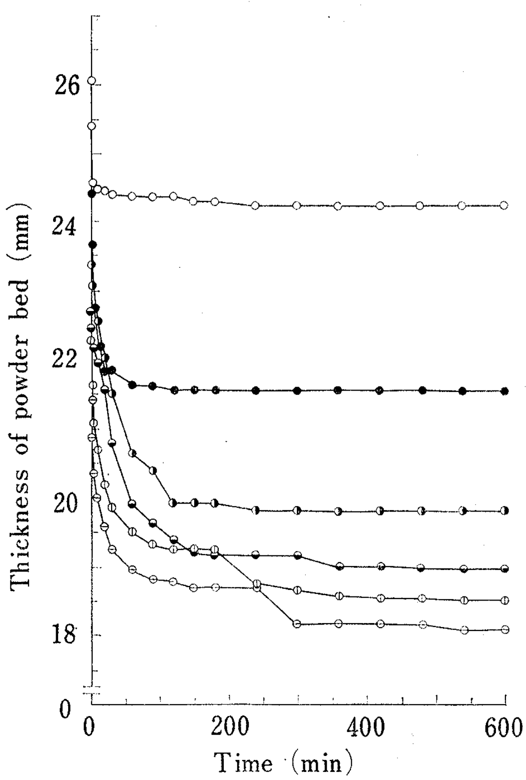

Fig. 4. Relationship between the Thickness of Powder Bed and Compressing Time when 1.158 g of g Lactose Powder is compressed under the Constant Upper Punch Pressure

upper punch pressure (kg/cm²):
—○—; 88, —●—; 199, —◑—; 394, —◒—; 595, —⦶—; 795, —⊖—; 990
the thickness of powder bed (mm) at porosity equal to zero: 14.87

transmission-ratio), when potassium chloride and lactose powder were compressed at the constant rate of displacement of upper punch (0.332 mm/sec).

It is obvious from Fig. 2 that the bed of lactose powder has larger porosity than potassium chloride during the compression, because 1.5 g of potassium chloride and 1.158 g of lactose have the same net volume (0.7545 cm³) of materials. In the case of lactose powder, the upper punch pressure increases slowly in the first stage of compression and rapidly in the range of pressure over 100 kg/cm².

The pressure–transmission-ratio in potassium chloride powder has wholly larger values than lactose, and takes the maximum value under the upper punch pressure equal to about 250 kg/cm² in both cases of 1.5 and 2.0 g of powder weight, while, in lactose, the ratio decreases monotonously during the compression.

2. Continuous Compression Under the Constant Pressure of Upper Punch

When potassium chloride and lactose powder were compressed continuously under the constant pressure of upper punch reached finally by the compression at the constant rate of displacement of upper punch (0.332 mm/sec), the thickness of powder bed changed as shown in Fig. 3 and 4, respectively.

As shown in Fig. 3 for potassium chloride, the thickness of powder bed reaches to 14.87 mm under 983 kg/cm² of upper punch pressure in case of 1.5 g of powder weight and to 19.83 mm under 1017 kg/cm² in case of 2.0 g. These thicknesses mean zero porosity. The thickness seems to approach to 14.87 mm under 625 and 813 kg/cm² in case of 1.5 g. On the other hand, in the case of lactose powder (Fig. 4) the thickness ceases soon to change, although

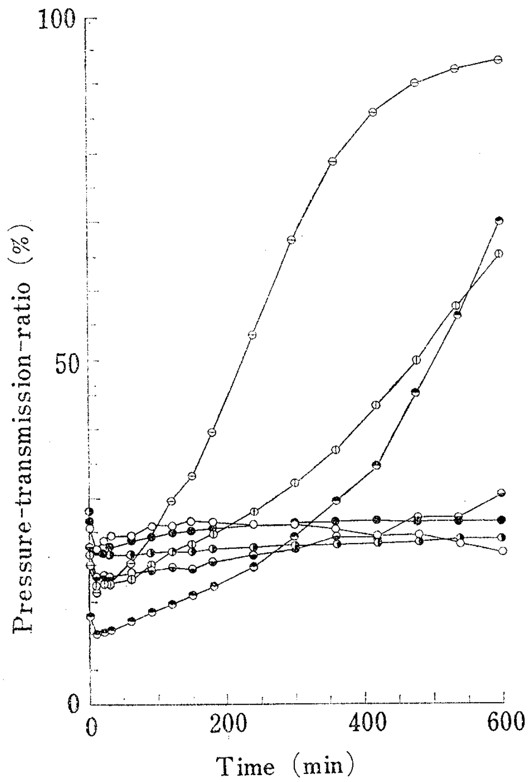

Fig. 5. Relationship between Pressure–Transmission-Ratio and Compressing Time when Potassium Chloride Powder is compressed under the Constant Upper Punch Pressure

powder weight (g): only ◓; 2.000, others; 1.500 upper punch pressure (kg/cm²):
—○—; 83, —●—; 183, —◑—; 391, —◒— 625, —⦶—; 813, —⊖—; 983, —◓—; 1017

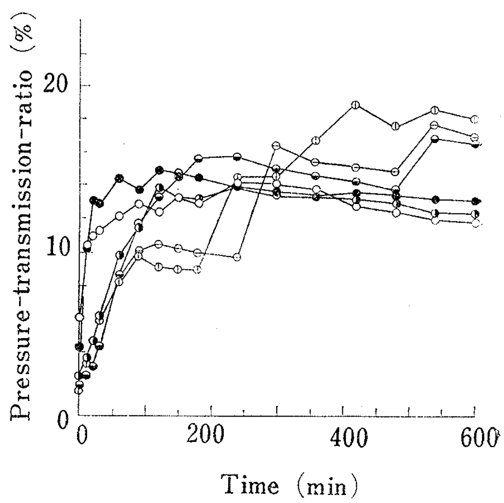

Fig. 6. Relationship between Pressure–Transmission-Ratio and Compressing Time when 1.158 g of Lactose Powder is compressed under the Constant Upper Punch Pressure

upper punch pressure (kg/cm²):
—○—; 88, —●—; 199, —◑—; 394, —◒—; 595, —⦶—; 795, —⊖—; 990

considerable amounts of void are still left in the bed of powder, and moreover the early changes are not smooth.

Fig. 5 and 6 show the changes of the pressure–transmission-ratio (%) during compression of potassium chloride and lactose powder, respectively. It is found from Fig. 5 that the pressure–transmission-ratio (%) increases remarkably whenever the porosity of potassium chloride powder has reached or has been approaching to the zero value (in cases of 1017, 983, 813 and 625 kg/cm^2 of upper punch pressure). Particularly, it seems to reach to 100% when the powder of net weight 1.5 g was compressed under the upper punch pressure 983 kg/cm^2. On the other hand, the pressure–transmission-ratio of lactose powder (Fig. 6) changes irregularly and reaches the nearly constant value after ten hours. Moreover, the sudden increase of the pressure–transmission-ratio in Fig. 6 corresponds to the decrease of the thickness of powder bed in Fig. 4. From this fact, we may conclude that the structure of powder bed was destroyed at the moment.

Discussion

1. Microscopic Observation of Replicated Surfaces of Compacts

Fig. 7 shows the photomicrographs of potassium chloride particles before the compression (Fig. 7,a) and of replicated surfaces of compacts (Fig. 7,b—e).[9] The particles before the compression are roundish owing to grinding in a mortar. As shown in Fig. 7,b, every particle on the surface of upper punch, when compressed under 183 kg/cm^2 of upper punch pressure for 600 min, seems to be not almost fractured and be deformed plastically in appearance so that the flat surfaces are produced in the regions contacted directly with the punch. Furthermore, on these flat surfaces of particles, man can see the concentric scratches which are

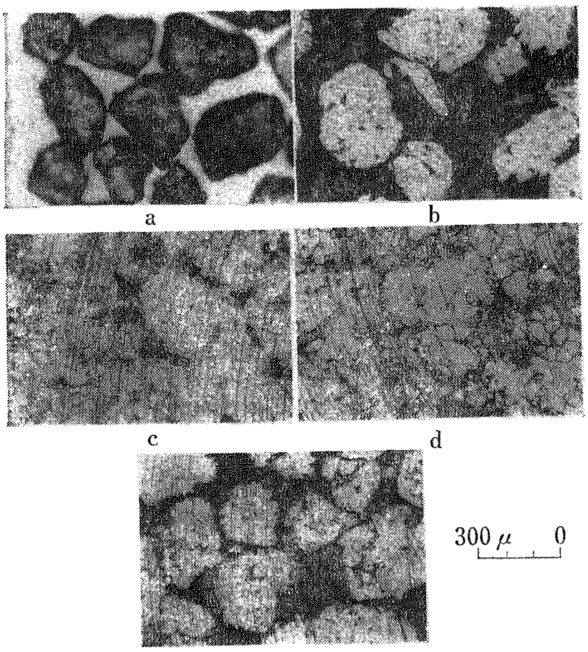

Fig. 7. Photomicrographs of Potassium Chloride Particles before the Compression a and Replicas of Particles in Contact with Upper Punch

b pressure (P): 183 kg/cm^2, compressing time (T): 600 min
c P: 625, T: 600, d P: 983, T: 600, e P: 983, T: 0

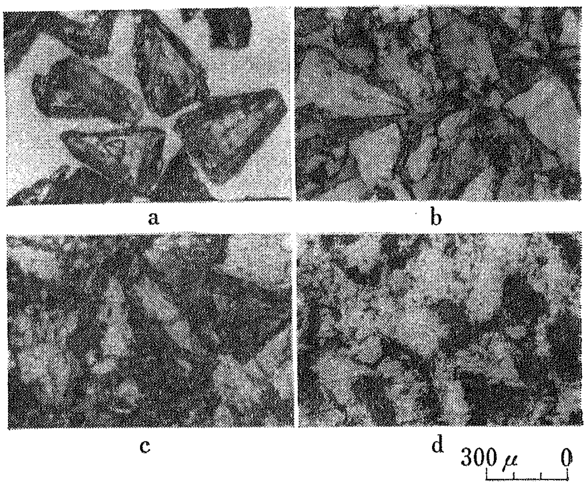

Fig. 8. Photomicrographs of Lactose Particles before the Compressin a and Replicas of Particles in Contact with Upper Punch

b pressure (P): 199 kg/cm^2, compressing time (T): 600 min
c P: 595, T: 600, d P: 990, T: 600

9) J. Okada, Y. Matsuda, and Y. Fukumori, *Yakugaku Zasshi*, **91**, 1207 (1971).

the copies of streakes on punch surface made by lathe bite. Particles compressed under 625 and 983 kg/cm² for 600 min are more remarkably deformed and many fragments or cracks tend to be produced (shown in Fig. 7,c and d). In the bed of powder whose porosity and pressure–transmission-ratio take the values equal to zero and 100%, respectively, as shown in Fig. 3 and 5, small fragments or largely deformed particles with many cracks are closely packed as shown in Fig. 7,d. But, although such the closely packed state is produced as a result of compression under 983 kg/cm² of upper punch pressure for 600 min, particles just after reaching of upper punch pressure to 983 kg/cm² are deformed as shown in Fig. 7,e. Although the particles shown in Fig. 7,b and e may be considerably fractured or many cracks may be produced in particles, the sizes of deformed particles are nearly identical to those of original particles.

On the other hand, in the case of lactose powder shown in Fig. 8, particles compressed under 199 and 595 kg/cm² of upper punch pressure are fractured and the fragments tend to fill the small voids, as authors pointed out in the proceeding paper.[9] Under 990 kg/cm² of upper punch pressure, particles in contact with upper punch are forced to deform or fracture to smaller fragments and apparently flat surfaces are produced.

2. Filling Behavior in Voids

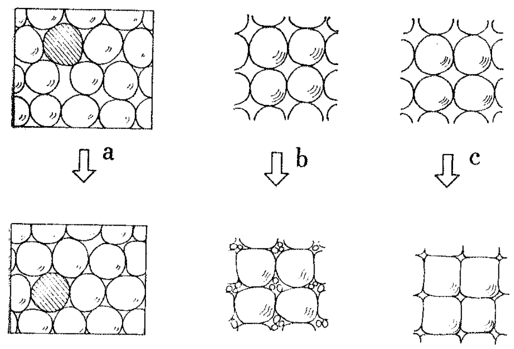

Fig. 9. Schematic Diagram of Different Methods of Particle Compaction by Cooper and Eaton

a : filling of large voids by rearrangement
b, c : filling of small voids
b ; by fragmentation, c ; by plastic flow

The models of filling behaviors in voids proposed by Cooper and Eaton are shown in Fig. 9.

As shown in Fig. 7,b, small voids in the bed of potassium chloride powder are filled by plastic deformation of particles in appearance under relatively low pressure, corresponding to the model Fig. 9,c. Furthermore, under the higher pressure than the critical pressure which seems to be between 391 and 625 kg/cm², the boundaries of particles become indistinct gradually with increase of compressing time and pressure, and many fragments or cracks are produced (Fig. 7,c, d and e). Perhaps, in these states, the fragmentation of particles arises at inside and circumference, corresponding to Fig. 9,b. Then, small fragments move to fill up voids very closely, as if original particles were deformed plastically.

On the other hand, particles of lactose powder are mainly fractured and the fragments fill small voids, corresponding to the model Fig. 9,b. That is, lactose particles behave as the brittle bodies. Accordingly, it may be considered that the rest of voids can not be filled easily, for the structure of powder bed becomes stronger and stronger owing to packing of small voids with produced fragments. Certainly, as shown in Fig. 4 and 6, although the sudden destructures are repeated at the first stage of compression owing to the unstable parts of structure and the fragmentation of particles, the thickness and the pressure–transmission-ratio stop to change after about five hours in spite of considerable amounts of voids left still in the bed of powder.

As shown in Fig. 2, the porosity of lactose powder is larger than potassium chloride in the case of upper punch pressure equal to zero, that is, before the compression. Then, the upper punch pressure increases slowly in the range of pressure lower than about 100 kg/cm² and the change of thickness is very large. Besides, as considered from Fig. 8,b, under such the low pressure, particles seem to be not fractured exceedingly. Accordingly, in this stage of compression, it may be considered that large voids in the bed of lactose powder are mainly filled by rearrangement of particles, corresponding to the model Fig. 9,a.

3. Pressure-transmission-ratio

When the weight of powder itself is neglected in Janssen's equation, the pressure–transmission-ratio is given by the form,

$$P_L/P_U = \exp(-4\mu_w KH/D)$$

where μ_w is the coefficient of friction between the die wall and the powder, K is the ratio of the horizontal to the vertical pressure, D is the diameter of powder bed, H is the thickness of powder bed, P_L is the lower punch pressure and P_U is the upper punch pressure. In this equation, μ_w and K are assumed to be constant.

It is clear from Janssen's equation that the pressure–transmission-ratio is equal to 100% only at the zero thickness of powder bed, but, as shown in Fig. 5, in the case of potassium chloride powder the pressure–transmission-ratio approaches to 100% in spite of some thickness under the high pressure, for example, under 983 kg/cm². Accordingly, this tendency can not be explained by Janssen's equation.

Besides, it is an important problem that, as shown in Fig. 2, the decrease of thickness accompanies the decrease of pressure–transmission-ratio under the high pressure, which is contrary to tendency of Janssen's equation, and also that, as shown in Fig. 5 and 6, under the constant load of upper punch, the pressure–transmission-ratio decreases temporarily in the case of potassium chloride powder, or increase in the case of lactose powder. Its reasons are still unknown.

Conclusion

From the Results described above, we may conclude the followings.

(1) When potassium chloride powder is compressed for ten hours under the constant pressure lower than the critical pressure which exists between 391 and 625 kg/cm², small voids in the bed of powder seems to be filled by plastic deformation of particles in appearance. In the range above the critical pressure, since the strain of particle must become much larger, many fragments or cracks are produced and these small fragments move to fill up voids very closely, so that the values of the porosity of powder bed and the pressure–transmission-ratio approach to zero and 100%, respectively.

(2) In the case of lactose powder, small voids in the bed of powder are mainly filled by fragmentation of particles, for lactose particles behave as the brittle bodies, and the thickness of powder bed and the pressure–transmission-ratio result to stop the changes after about five hours of compressing time in spite of considerable amounts of void left still.