

Studies on Compaction of Powders. III.¹⁾ Compression Behavior of Several Powders under Low Pressure

AKINOBU OTSUKA, KAZUMI DANJO, and HISAKAZU SUNADA

Faculty of Pharmacy, Meijo University²⁾

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The compression runs on several powders of different particle size and hardness were made in the relatively low pressure range where the fragmentation of particles did not take place. The porosity/pressure data were arranged on the assumption that the porosity at a given pressure can be directly related with the average axial force acting on each particle. An empirical equation containing the constants which are not affected by particle size was then obtained. It was confirmed that the compaction parameters obtained above are related to the pressure transmission ratio and hardness of the material being compressed.

Studies³⁾ on the relation between compression force and the apparent volume or the porosity of powder beds have been carried out for a long time. However, most of the empirical equations presented in literatures seem to be applicable only to the specific samples under the specific experimental conditions. Furthermore, it cannot be said that the physical meaning of the constants in those equations are satisfactorily clear. This is due to the difficulty of investigating the variables which affect the compaction behavior of powder particles, such as particle size, particle shape, particle density, hardness or frictional characteristics, independently.

Another complexity of powder compression is as follows. It has been believed that there are three basic mechanisms of compaction⁴⁾ during compression of powders, *viz.*, (1) packing of particles by sliding, (2) elastic and plastic deformation and (3) fragmentation. In practice, however, it may be almost impossible to distinguish the mechanism which predominates in any stage of compaction, because two or more ways of compaction occur either simultaneously or consecutively.

TABLE I. Powders Used

Material	Mean particle diameter, μ	Material	Mean particle diameter, μ
Carborundum (C)	45.2	Calcium carbonate (Ca)	13.7
	23.3		7.1*
	17.8		5.3*
Alumina (A)	16.2	Kaolin (K)	5.0*
	9.7		5.0
	7.6		3.9*
	6.8		29.5*
	5.5		13.0*
Quartz (Q)	22.2	Chloramphenicol (CP)	65.1
	19.3		17.3
	6.8		4.5

1) Part II: A. Otsuka, H. Sunada, K. Danjo, and M. Nakagaki, *Yakugaku Zasshi*, **90**, 244 (1970).

2) Location: *Tempaku-cho, Showa-ku, Nagoya*.

3) *e.g.* K. Kawakita and K.H. Ludde, *Powder Tech.*, **4**, 61 (1971).

4) W.A. Gray, "The Packing of Solid Particles," Chapman & Hall, 1968, p. 96.

The purpose of this work is to study the effect of some of the variables which affect the compression behavior of powders, especially the effect of particle size and hardness, in detail. To simplify discussions compression runs were made under the relatively low pressures where the fragmentation of particles did not take place.

Experimental

Samples used are commercial materials and are shown in Table I. Those marked * are samples classified by hydraulic elutriation method. The particle size distribution was measured by the sedimentation method and the arithmetical mean diameter on weight basis was obtained. The specific surface area of each sample was measured by the air permeability method. It was found that there was no difference in the surface area before and after compression. Hence, it was confirmed that the fragmentation of particles has not occurred during the compression process.

The apparatus shown in Fig. 1 was designed for compressing powders. This apparatus was set on the lower platen of the Shinkoh Universal testing instrument TOM-5000X which was equipped for the automatic recording of applied force and transmitted force. The instrument was operated at a downward crosshead motion of 0.5 mm/min and the change in height of a compact with pressure was determined from the crosshead travel recorded on a chart. The porosity of the compact was calculated from the diameter of the die and the height of the compact.

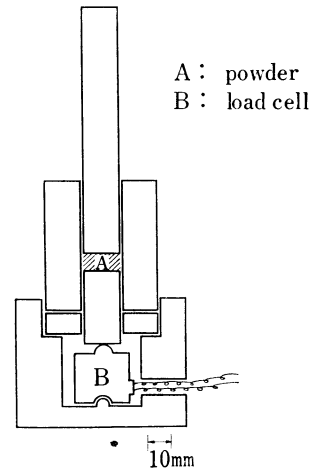


Fig. 1. Die Pressing Apparatus

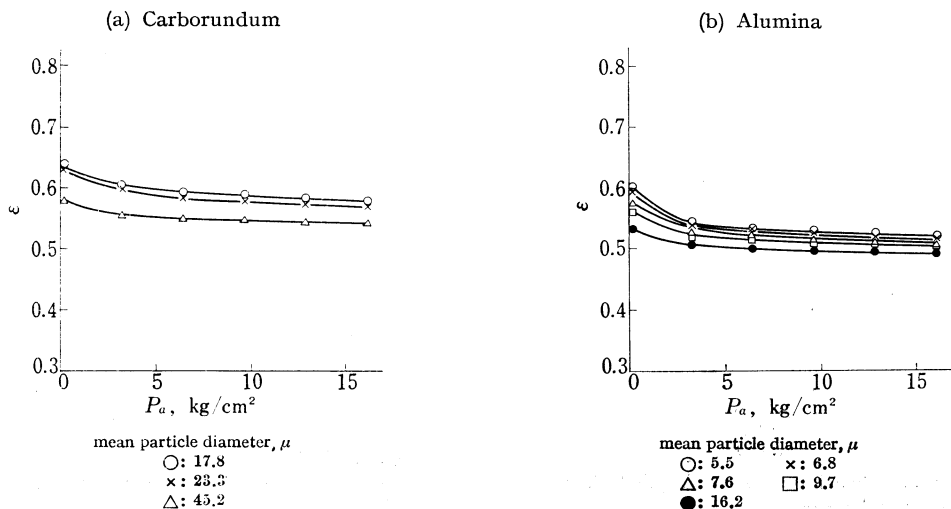
Result

1) Pressure Porosity Data

Fig. 2(a)—(g) shows the relation between applied pressure and porosity. In each case the initial height of a powder bed is 2.5 cm. As is generally known, porosity at a given pressure increases with decrease in mean particle size for each material.

2) Pressure Transmission Ratio

Fig. 3 shows the typical results of the applied pressure P_a and the axial pressure transmitted through the bed P_t for alumina. It can be seen that P_a and P_t are approximately in a proportional relation. Similar results were obtained for other samples used.



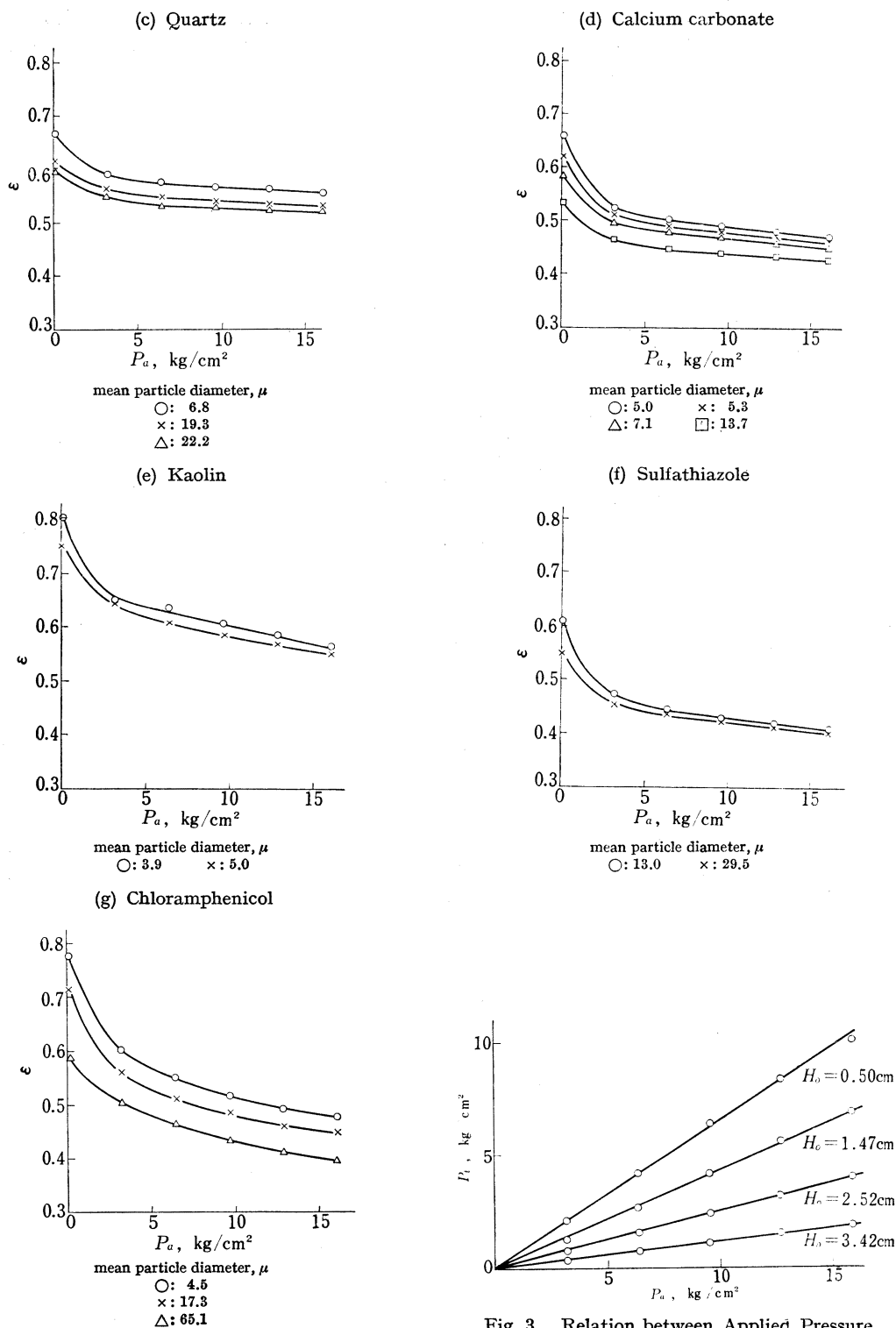
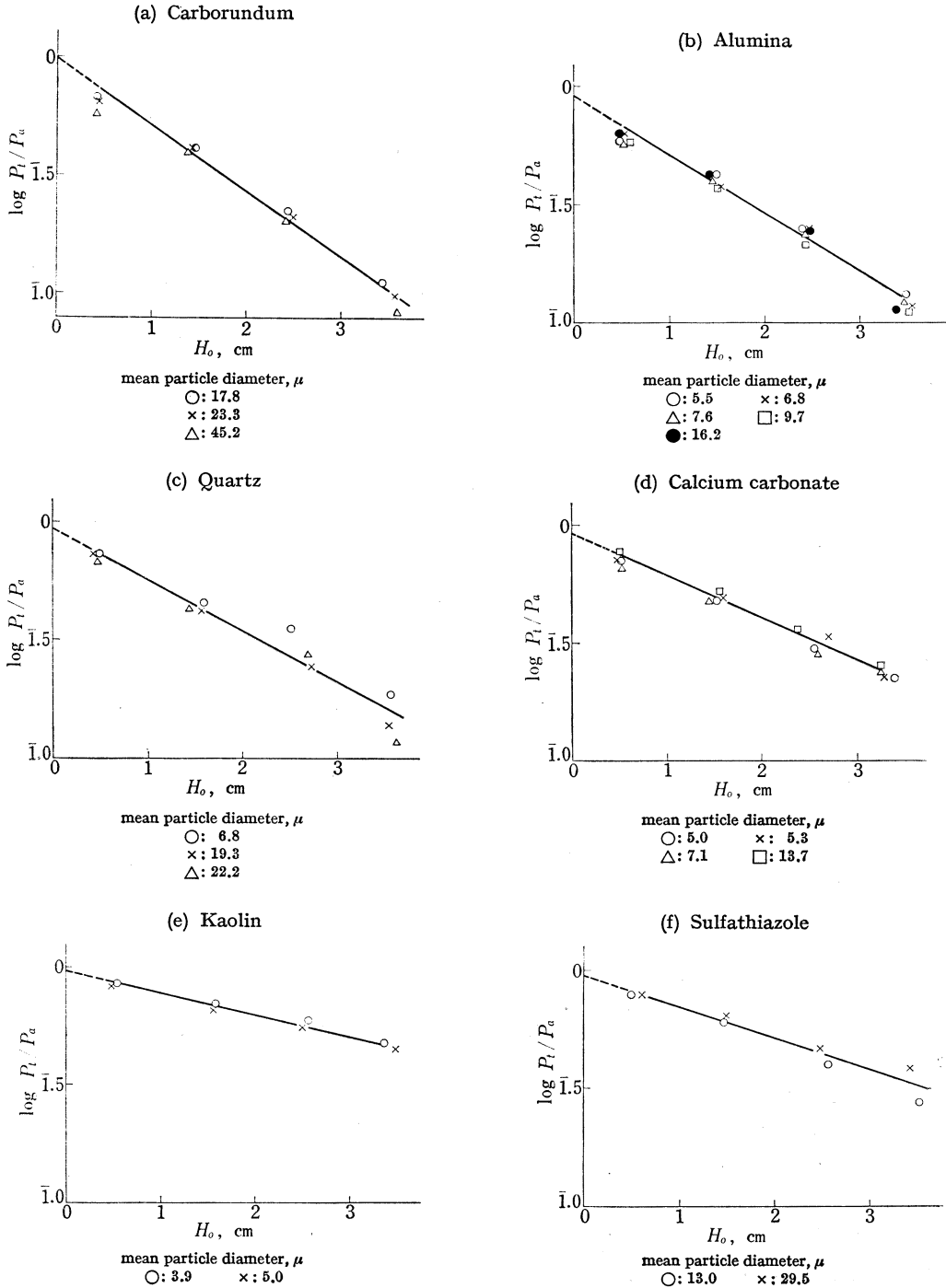


Fig. 2. Relation between Applied Pressure and Porosity

Fig. 3. Relation between Applied Pressure and Transmitted Pressure for Alumina: $d = 16.2 \mu$

The relation between the slope of the ratio P_t/P_a and the original height H_0 was examined and the results are given in Fig. 4(a)—(g). These figures show that the data can be expressed by the following empirical equation.

$$\log(P_t/P_a) = -KH_0 + \log C \tag{1}$$



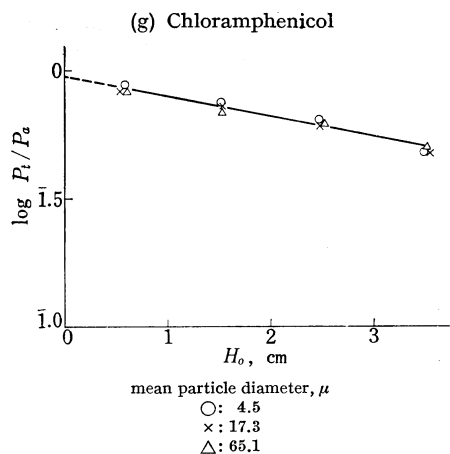


Fig. 4. Effect of Bed Height on Pressure Ratio

$$\log (P_1/P)/(V_r-1) = kd^n \quad (3)$$

where V_r is the relative volume and P_1 the extrapolated value at $V_r=1$. Varma and Venkateswarlu⁶ pointed out that particle diameter d , diameter of compact D and surface energy of material γ are the factors which affect porosity ϵ . From dimensional analysis they considered that ϵ is proportional to $(\gamma/Pd)^m(d/D)^n$. Both of above works have been carried out at considerably high pressures and it can therefore be assumed that particles have been subjected to complicated changes such as deformation or fragmentation. Also, in both works a particulate material has been treated as a mass. In this paper compaction phenomena are discussed from a microscopic viewpoint, that is, the mean axial force acting on each particle was taken into consideration in place of the total force on a bed.

Taking N to be the number of particles existing in a certain horizontal cross section of a powder bed and W to be the total axial force at this cross section, the mean axial force on each particle \bar{w} is

$$\bar{w} = W/N \quad (4)$$

and N can be expressed by

$$N = \pi D^2/4A \quad (5)$$

where D is the diameter of a die and A the average area associated with each particle. A depends on the state of packing of particles and was evaluated by Rumpf,⁷ Otsuka and others¹¹ as

$$A = \pi d^2/6(1-\epsilon) \quad (6)$$

where d is the mean particle diameter. When the transmission of compression force through a powder bed is in accordance with Eq. (2), the volume average pressure \bar{P} can be calculated from the equation:

$$\bar{P} = (P_a - P_t)/\ln (P_a/P_t) \quad (7)$$

In terms of \bar{P} the mean total axial force at a cross section of the compact W is

$$W = \pi D^2 \bar{P}/4 \quad (8)$$

where K and C are constants. C is close to unity for every powder and consequently, Eq. (1) can be approximated as follows:

$$\log (P_t/P_a) = -KH_o \quad (2)$$

The value of K does not seem to be affected by particle size but differs with each material.

Discussion

The effect of particle size on the compression of several powder has been examined by Huffine and Bonilla.⁵ Their results were interpreted according to Balshin's equation and it was indicated that the compaction modulus increased with increasing size. The relation has been described by the expression:

5) C.L. Huffine and C.F. Bonilla, *A.I.Ch.E. Journal*, **8**, 490 (1962).

6) Y.B.G. Varma and D. Venkateswarlu, *Chem. Proc. Eng.*, **48**, 77 (1967).

7) H. Rumpf, *Staub*, **19**, 150 (1959).

Combining Equations (4), (5), (6), (7), and (8), the mean axial force on each particle \bar{w} may be represented by

$$\bar{w} = \pi d^2(P_a - P_t)/6(1 - \epsilon) \ln(P_a/P_t) \tag{9}$$

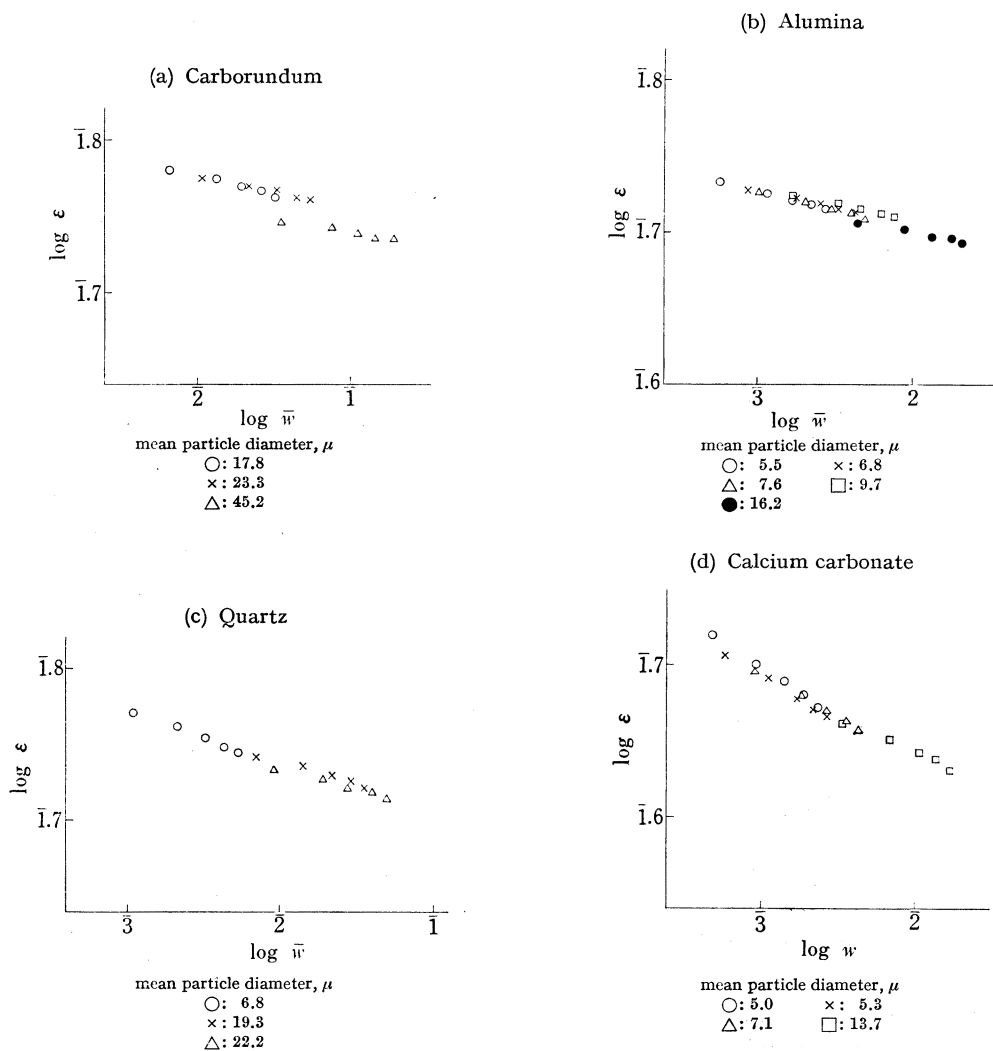
Then, one can calculate the value of \bar{w} using the experimental data of P_a , P_t , and ϵ .

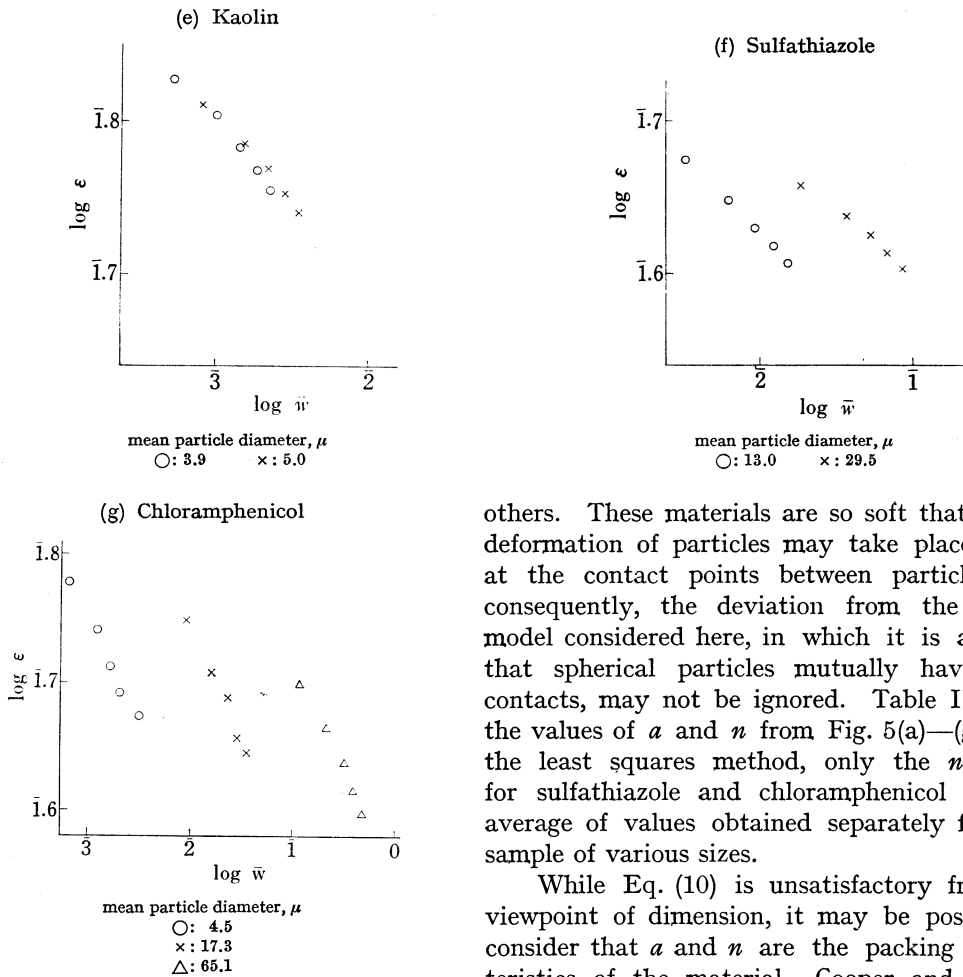
Assuming that the average force on one contact of a particle with another is proportional to \bar{w} , it is likely that the value of \bar{w} is directly related to the porosity of a powder compact. In Fig. 5(a)—(g) the arranged data of Fig. 2(a)—(g) are plotted with $\log \bar{w}$ as abscissa and $\log \epsilon$ as ordinate. All the data, except sulfathiazole and chloramphenicol, can be represented by a single line for each material and the following empirical equation is then obtained:

$$\epsilon = a\bar{w}^{-n} \tag{10}$$

where a and n are constants. As the effect of particle size is included in the value of \bar{w} constants k and n are independent of particle size.

In the cases of sulfathiazole and chloramphenicol the data cannot be represented by a single line. This may be ascribed to the difference in hardness between these materials and



Fig. 5. $\log \epsilon$ against $\log \bar{w}$

others. These materials are so soft that plastic deformation of particles may take place easily at the contact points between particles and consequently, the deviation from the simple model considered here, in which it is assumed that spherical particles mutually have point contacts, may not be ignored. Table II shows the values of a and n from Fig. 5(a)—(g) using the least squares method, only the n values for sulfathiazole and chloramphenicol are the average of values obtained separately for each sample of various sizes.

While Eq. (10) is unsatisfactory from the viewpoint of dimension, it may be possible to consider that a and n are the packing characteristics of the material. Cooper and Eaton⁸⁾ discussed the effect of hardness on the compression of alumina, magnesia, silica and calcite.

For metal powder compaction, Heckel⁹⁾ pointed out that plastic deformation is particularly of importance and he discussed the relation between the compaction parameter and the yield strength of compacted material. Varma and Venkateswarlu⁶⁾ reported that compaction parameter m was related to the coefficient of internal friction of the material.

TABLE II. a and n for Each Powder

Material	a	n
Carborundum	0.514	0.0338
Alumina	0.450	0.0246
Quartz	0.474	0.0313
Calcium carbonate	0.341	0.0540
Kaolin	0.303	0.1066
Sulfathiazole	—	0.0929
Chloramphenicol	—	0.1669

8) A.R. Cooper Jr. and L.E. Eaton, *J. Amer. Ceram. Soc.*, **45**, 97 (1962).9) R.W. Heckel, *Trans. Met. Soc. A. I. M. E.*, **221**, 671 (1961).

Fig. 6 shows n as a function of Mohs hardness and it is found that the softer the material the greater is the decrease in volume for a given pressure increase. This result is of interest since it is expected that plastic deformation of particles plays an important role even in the compression under low pressure. In the case of centrifugal compaction,¹⁰⁾ however, it is difficult to find a certain relation between the compaction parameter n ¹¹⁾ and hardness as shown also in Fig. 6 with points x. It might be suggested that the predominating mechanism of compaction differs with each procedure.

Measurements of pressure transmission through a powder bed have been described Jansen, Shaxby and others.¹²⁾ The principal assumptions made in their studies are the following:

- (1) there is no stress distribution in radial directions;
- (2) the ratio of horizontal pressure to vertical pressure is constant;
- (3) bulk density is independent of depth.

When the force acting on the top of the bed is much larger than the weight of the powder the pressure transmission ratio P/P_u at depth H can be expressed by

$$P/P_u = \exp(-4\beta\mu H/D) \tag{11}$$

where β is the ratio of the horizontal pressure to the vertical pressure, μ the coefficient of wall friction and D is the diameter of the circular cross section. Spencer and others¹³⁾ also tried to derive a theoretical equation for pressure transmission using several assumptions and obtained an equation which is essentially the same as Eq. (11). Bessho and others¹⁴⁾ applied

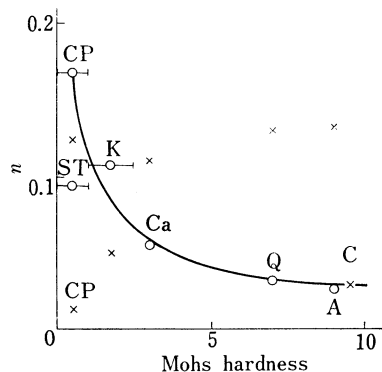


Fig. 6. Relation between Mohs Hardness and Constant n in Eq. (10)

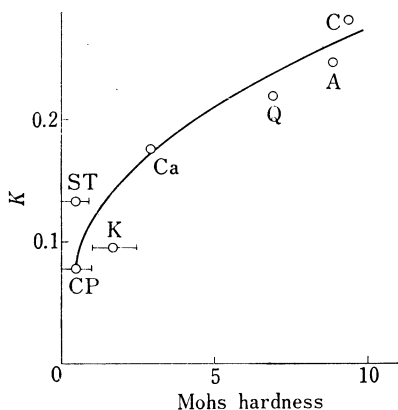


Fig. 7. Relation between Mohs Hardness and Constant K in Eq. (2)

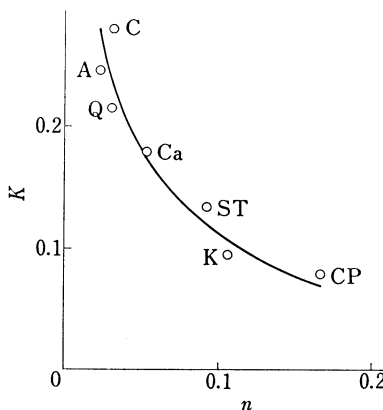


Fig. 8. Relation between n and K

- 10) A. Otsuka, H. Sunada, and K. Danjo, *Yakugaku Zasshi*, **89**, 1013 (1969).
- 11) In the case of centrifugal compaction the relation between the average force on each particle and the porosity can be expressed as: $\epsilon = bF^{-n'} + 0.26$
- 12) J.H. Shaxby and J.C. Evans, *Trans. Faraday Soc.*, **19**, 60 (1923).
- 13) R.S. Spencer, G.D. Gilmore, and R.M. Wiley, *J. Appl. Phys.*, **21**, 527 (1950).
- 14) S. Bessho, S. Tomita, and S. Ito, *Yakugaku Zasshi*, **89**, 599 (1969).

experimental data of various substances to this theoretical equation and discussed β and μ .

The empirical Eq. (1) obtained by the authors is identical to Eq. (11) when $C=1$ and $4\beta\mu/2.3D=K$ were taken. Since the value of C is close to unity and D is constant throughout the experiments, it may therefore be said that the present data approximately satisfy Eq. (11). However, it has been known from numerous studies that the assumptions discussed above are not necessarily applicable to general powders, so that the constant K cannot be expressed by the simple form mentioned above. In the present situation, therefore, it may be significant to find out factors which affect the value of K . First, it can be seen from Fig. 4 that K value is independent of particle size for any particular material. Next, Fig. 7 shows the correlation of K with hardness. Ridgway and others¹⁵⁾ examined the effect of crystal hardness on radial pressure at the wall of a tableting die and clarified that the pressure on the die wall increased as the hardness value decreased. Higuchi and others¹⁶⁾ also described that softer substances seemed to give higher die wall pressures. Our results here are in agreement with those of the above authors, while there are differences in the experimental method and the experimental condition.

Fig. 8 shows the relation between K and n . This correlation is to be expected since it was found already that both K and n are closely related to hardness.

15) K. Ridgway, J. Glasby, and P.H. Rosser, *J. Pharm. Pharmac.*, **21**, Suppl., 24S (1969).

16) T. Higuchi, T. Shimamoto, S.P. Eriksen, and T. Yashiki, *J. Pharm. Sci.*, **54**, 111 (1965).