Particle-Size Effects in the Compression of Powders

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The compression of powders to form tablets, briquettes, pellets, etc. is a widespread practice which has developed largely as an art, with few investigations of other than a qualitative nature. This may perhaps be attributed to the relatively simple shapes which are commonly pressed and to the fact that many powdered materials compact readily and thus require only a minimum of experimentation to develop a satisfactory product. Lately however the potential advantages of metal, ceramic, and cermet products formed initially by powder compaction have resulted in more stringent requirements for uniformity of density in complex pressings, and these have emphasized the need for better understanding of the basic compression process.

Complete evaluation of the powder-pressing operation requires analysis of the pressure-volume relationships, including the effects of the size, shape, and surface condition of the powder particle. Also to be considered are the strength of the bond obtained and the effect of such macrofactors as die condition and the size and shape of the compact. The present paper explores pressure-volume relations in the compression of dry, nonmetallic powders, particle size being the primary variable.

PREVIOUS WORK

The literature on powder compression is not extensive, and most references deal with the nature of the bond produced (*1*, *2*) or with the distribution of density within the powder mass (*3*, *4*).

Walker (*5*) made the first accurate measurements of pressure-volume relationships with both metallic and nonmetallic powders and observed that the logarithm of pressure plotted vs. volume as a substantially straight line. Bal'shin (*6*) deduced a similar result by assuming that the frictional work dW expended in effecting an irreversible volume change dV in the powder is directly proportional to the pressure increase required:

$$dW = -PdV = idP \qquad (1)$$

This relation neglects any work performed in possible elastic compression of the powder mass. Despite its somewhat questionable basis Bal'shin integrated Equation (1) and substituted V_o for V to obtain the more generalized relation

$$\log p = -L\,V_o + b \qquad (2)$$

where L and b are constants. L, the absolute value of the slope of log P vs. V_o, was designated the *pressing modulus*. The expected linearity was demonstrated for metallic powders at low and intermediate pressures, but deviations occurred at high pressures. Subsequent work showed that L varies considerably with such factors as particle size and shape.

Several investigators have considered the compaction of powders from the standpoint of porosity reduction on pressure increase and have shown this reduction over low-pressure ranges to follow a relation of the type

$$\epsilon = \epsilon_o\,e^{-aP} \qquad (3)$$

Shapiro (*7*) considered this compaction to be analogous to the reduction in volume on isostatic compression of a solid, but Spencer, et al. (*8*), Konopicky (*9*), and Heuberger (*10*) merely employed Equation (3) as a convenient but empirical means of data correlation. Since porosity is calculated from the difference in bulk and solid densities, it is subject to the usual error of small differences, and wide deviations from the relation are observed at high pressures. Thus the relation is not as useful as Equation (2) for data correlation over wide pressure ranges.

Ballhausen (*11*) employed an empirical modification of Equation (3), replacing ϵ with the term $\epsilon/(1-\epsilon)$. This resulted in a better fit in the low-pressure region but did not affect the high-pressure deviation.

Smith (*12*) proposed still another empirical relation:

$$\rho_a - \rho_{a_o} = C\sqrt[3]{P} \qquad (4)$$

Again wide deviation from the relation occurs in the high-pressure region.

Little consideration has been given to the effect of particle size on pressure-volume relations in powder compaction. Duffield and Grootenhuis (*13*) made measurements on spherical cop-

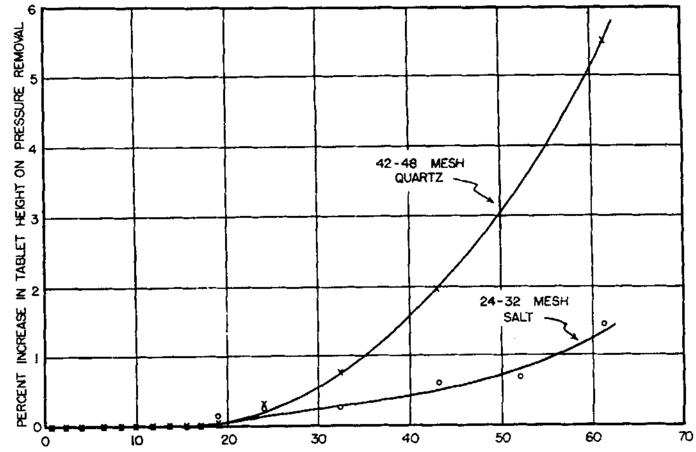

Fig. 1. Elastic recovery in compression of sodium chloride and quartz powders.

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per powders, with screen fractions covering a 5:1 range in particle diameters. A significant effect was observed, but it was noted that similar effects could be obtained with a narrow-size fraction by varying the initial packing density.

In his treatise on fine particle technology Dalla Valle (*14*) introduced the concept of particle pressure diffusion. He proposed that small particles tend to diffuse into the voids of a packing of mixed sizes as pressure is applied. Assuming this diffusion to be a function of average particle size, compact density, and pressure, he obtained by dimensional analysis

$$\mathcal{D} = n\, d\sqrt{P\, g_c/\rho_a} \qquad (5)$$

At the time of his writing there were no suitable data for testing his proposal.

EXPERIMENTAL CONDITIONS

Pressure-volume relationships were studied in the conventional manner by simple die compaction of the powder, the assumption being made that the die diameter was constant and that volume change was directly indicated by height change in the powder mass. Limiting the initial powder fill height to a maximum of 1-die diam. insured that density variation in the compact due to wall friction would be negligible. Repeated striking of the die during the filling kept the initial packing density variation within ± 3%, with no trend in this variation with particle size.

APPARATUS

The compression runs were made in a laboratory press with an air chamber added to provide steady pressure. Two calibrated gauges covered the ranges from 0 to 2,000 and from 0 to 25,000 lb./sq.in. A simple untapered die and punch set for commercial tabletting machines was employed. The die had an inside diameter of 0.4070 in., and the punches were 0.4063 in. in diameter.

The lower punch was fixed in the die. Movement of the upper punch in the die was measured by two dial indicators attached to the press head and bearing on a plate attached to the die. The indicators were graduated in 0.0005-in. divisions and were read to the nearest 0.0001 in.

Elastic distortion of the press frame was read as a function of pressure with the die empty. This correction, which amounted to less than 6% of the compact height at the highest pressures used, was then subtracted from observed readings in powder compressions.

MATERIALS

Three crystalline powders were employed: sodium chloride, sucrose, and quartz representing easily deformable, intermediate, and essentially nondeformable crystalline materials. The salt powder was prepared by slow evaporation of a saturated aqueous solution, followed by drying, crushing, and screening of the crystal aggregates. Commercial granulated sucrose was simply dried and screened. The quartz was prepared by ball mill crushing of large single crystals of alpha silica, followed by washing with dilute hydrochloric acid, rinsing with water, drying, and screening.

Fourteen standard screens from 24 to 200 mesh were employed, which gave seven size fractions ranging from 0.0254 to 0.0032 in. The powder particles were all essentially equiaxed, and the size of the fraction was assumed equal to the average screen opening. With the close screen sizes the maximum nominal deviation from the average within a fraction was only ± 9%. A statistical microscopic measurement of 200 particles of the 24- to 28-mesh salt fraction gave a mean particle diameter of 0.0262 in. as compared with the nominal value of 0.0254 in.

PROCEDURE

A typical compression run was made by quickly transferring dry powder to the die cavity, inserting the upper punch, and then closing the press until contact was made with the powder, but without measurable pressure application. The indicators were zeroed, and the compression was carried out in a series of steps, each pressure being held until no further movement was observed on the indicators. This required no more than 1 min./point and generally much less. The maximum pressure applied was 61,200 lb./sq.in. When this level was reached, the tablet was removed from the die, weighed, and measured. In computing V_o the densities of salt, sucrose, and silica were taken as 2.163, 1.588, and 2.650 g./cc., respectively.

ELASTIC COMPRESSION EFFECTS

To measure the elastic compression of the powder, in the first experiments the pressure was almost totally released after each level was reached, and the indicators were read before proceeding to the next pressure level. It is apparent from Figure 1 that no elastic effect was present in salt or quartz up to 20,000 lb./sq. in. Beyond this pressure a gradual increase in elastic compression was observed which amounted to about 1.5% of the pressure-free volume at 61,200 lb./sq. in. for salt and to about 5.5% for quartz.

PARTICLE SIZE EFFECTS

Compression runs on salt, sucrose, and quartz for each of the particle size

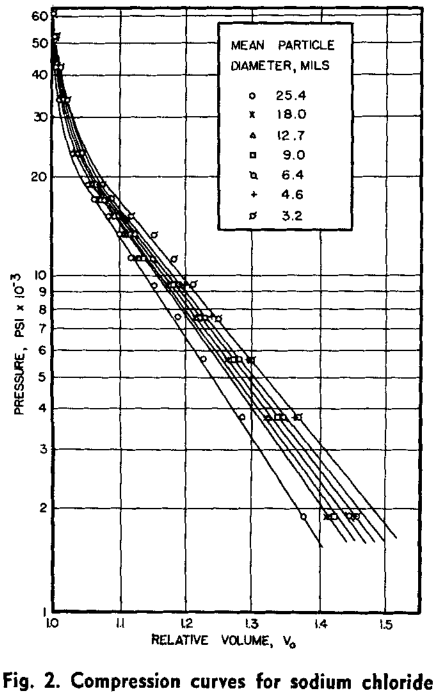

Fig. 2. Compression curves for sodium chloride powders.

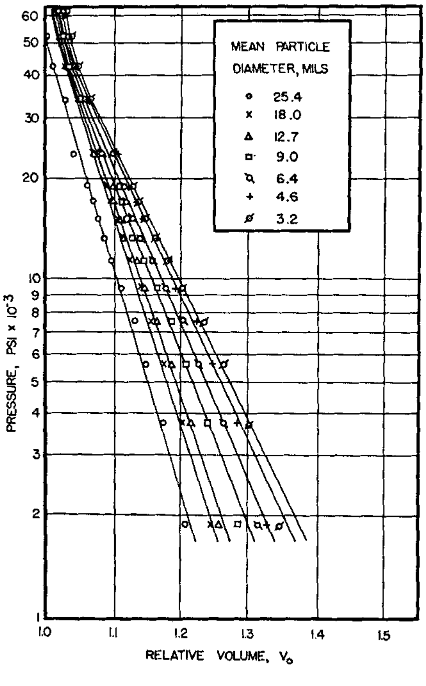

Fig. 3. Compression curves for sucrose powders.

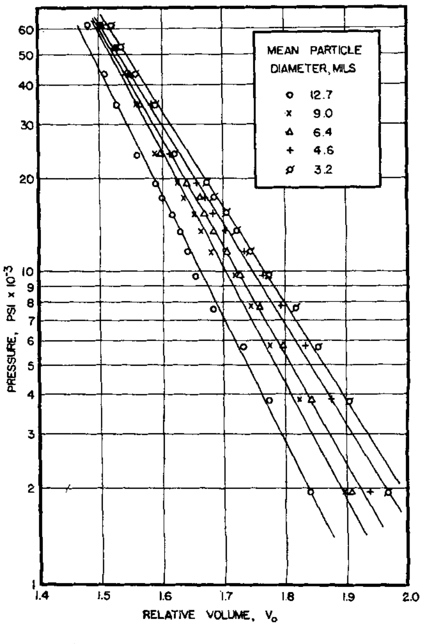

Fig. 4. Compression curves for quartz powders.

fractions are presented in Figures 2, 3, and 4. The curves shown were determined by least squares. Salt deviates from linearity appreciably, but sucrose and quartz follow the Bal'shin relation reasonably well. The voids in salt fall to zero at the highest pressures and almost to zero for sucrose. For quartz extrapolation of the curves indicates that 2,000,000 to 5,000,000 lb./sq.in. would be required for such compaction. In each case the result of increase in particle size is to increase the slope or pressing modulus. This indicates that the initial volume and the decrease in volume for a given pressure increase are greater for small particles than for large ones.

Figure 5 shows the slopes of the previous curves as an exponential function of the particle size. The slope or pressing modulus is expressed as log $(P_1/P)/(V_o - 1)$, where P_1 is the extrapolated intercept at the $V_o = 1$ axis. The effect of particle size for each material can evidently be expressed fairly closely as

$$\log (P_1/P)/V_o - 1) = k\, d^a \qquad (6)$$

Table 1. Values of Constants k and a for Various Materials

Material	k	a
Sodium chloride	4.5	0.11
Sucrose	18.4	0.26
Quartz	8.5	0.18

The constants k and a are shown in Table 1 and are different for each material. They are presumably related to the mechanical properties of the material, particularly its elastic modulus and yield strength. The constants in Table 1 however do not seem to vary in a manner consistent with the relative properties of the three materials. In addition the properties of these materials generally exhibit considerable scatter. Thus no correlation between the constants and the properties is presently evident. Further evaluation with powdered metals having well-defined properties is indicated.

MECHANISMS OF POWDER COMPACTION

It may be postulated that the behavior of a mass of particles under pressure will be largely controlled by the number and nature of the contacts between particles in the mass. For a given packing of spherical particles of diameter d in a circular die of diameter D there is an average area A associated with each particle in a layer transverse to the direction of punch travel. This area would be proportional to d^2:

$$A = m\, d^2 \qquad (7)$$

For uniform regular packing m is readily evaluated. For example it is unity for the simple cubic packing and 0.866 for the hexagonal closest packing. As a first approximation it may be assumed that m varies inversely with the packing density:

$$m = m'/\rho_a \qquad (8)$$

Thus the number of particles N in a full layer is

$$N = (\pi D^2/4)/(m'\, d^2/\rho_a) \qquad (9)$$

In terms of the pressure P the total axial force on the compact is

$$F = \pi D^2 P/4 \qquad (10)$$

and the idealized axial force f on each particle is

$$f = F/N = m'\, P d^2/\rho_a \qquad (11)$$

Different packings, presence of nonspherical particles, uncertain number and direction of contacts, and other factors will affect this result, but in general the average axial force per contact may be considered proportional to Pd^2/ρ_a.

The force exerted on the contacts between particles at a given die pressure is therefore considerably less per contact for the small particles than for the large. However the decrease in volume of the mass for a given pressure increase is greater for the small particles. The decrease in volume on applying pressure to a powder mass may take place in several ways:

1. The particles may slide over each other without appreciable deformation to new equilibrium positions.
2. The particles may deform elastically at and around the contact points.
3. The particles may deform plastically.
4. The particles may deform by fracture and breakage.

The mechanism which predominates in any case will depend primarily on the material of which the particles are composed, but it is probable that in most cases a combination of two or more of the above possibilities will occur, either simultaneously or consecutively. Thus the initial phase for any material will most probably be the first of the above actions. Eventually however, as pressure continues to increase, the possibilities for this slippage decrease, both because there are no more places for the particles to go and because the cohesive forces developed at the contact points become too great for the particles to slide against them. After this another mechanism will begin to predominate.

The volume change accompanying slippage may well be greater than for any subsequent action, because it involves movement of the particle as a whole and not merely a change within the particle. Accordingly the pressing modulus log $(P_1/P)/(V_o - 1)$ would be lowest for a material undergoing compression by slippage and shifting.

If one neglects wall friction, the total volume change accompanying slippage to the same force per contact might be the same for a packing of large particles as for small ones, since there would be expected a greater volume change per slip for the large particles but a correspondingly smaller number of slips in a unit volume. The pressure at which each slip is completed however would be considerably higher for the finer particles. Consequently the slippage phase of compression of the large particles at low pressures has been largely supplanted by the subsequent phases, and the pressing modulus is correspondingly high. For the small particles slippage remains predominant to a much higher pressure, and the pressing modulus is low.

Dalla Valle's theory of particle pressure diffusion (*14*) need not be limited to the movement of small particles. Rather it would appear that all particles, large or small, shift and slide to

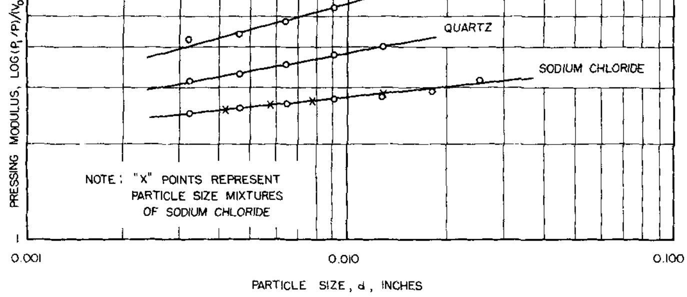

Fig. 5. Pressing modulus particle size relation for sized powders and powder mixtures.

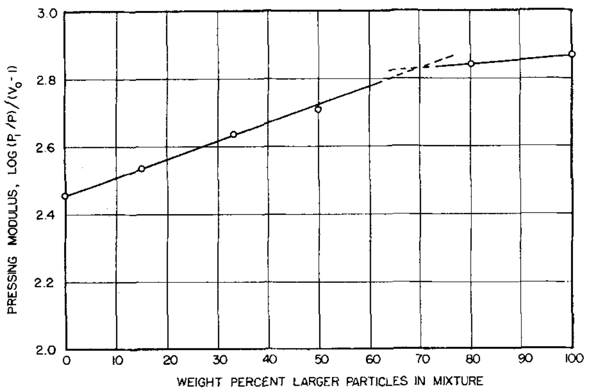

Fig. 6. Pressing moduli of mixed particle sizes of sodium chloride as function of percentage of larger particles in mixture.

some degree when pressure is applied in directions which reduce the porosity. This action may thus be regarded as a diffusion of matter into void space, with a diffusivity defined by Equation (5). Comparison of Equation (*11*) with Equation (5) shows that this diffusivity is in fact expected to be proportional to the square root of the average force per contact in the powder mass.

MIXED PARTICLE SIZES

Powders of uniform particle size are seldom used in commercial pressing operations, and the foregoing analysis was therefore tested for the compression of mixed sizes. A series of four compressions was made with mixtures of the 0.0180- and 0.0032-in. salt powders. Mixtures containing 15, 33, 50, and 80% by weight of the larger powder were used.

Results of these compressions showed slightly better conformance to the Bal'shin relation than did the single size fractions. The pressing moduli, log $(P_1/P)/V_o - 1)$, are plotted vs. percentage of the larger powder in Figure 6. The modulus increases regularly with addition of the large particles until the mixture becomes saturated at around 60 to 70% and shows little further change. This changeover can be interpreted as indicating the establishment of a continuous packing of the large particles. The small particles would fit loosely in the interstices of such a packing and would exert little influence on its behavior.

Shown in Figure 5 are the pressing moduli of the mixtures plotted vs. the geometric mean particle diameter d_{avg} defined as:

$$\log d_{avg} = \Sigma(w \log d)/\Sigma w \quad (12)$$

The values for the mixtures fall acceptably close to the line established for the closely sized fractions and thus Equation (6) can be used with this mean diameter to characterize the pressure-volume relations of mixed sizes.

CONCLUSION

The results of this study indicate that the compression of nonmetallic powders of equiaxed particles can be acceptably characterized by the Bal'shin relation and that the pressing modulus so defined is a simple exponential function of the particle size. The volume change accompanying a given pressure increase is greater for small particles than for large. This can be explained on the basis of the pressure level at which particle slippage ceases as a compaction mechanism and is supplanted by particle deformation, because this level is higher for small particles than for large. Mixed particle sizes behave in similar fashion and within the range studied can be simply characterized by use of the geometric mean particle size.

ACKNOWLEDGMENT

This paper is based on portions of the first author's doctoral dissertation, Columbia University, New York, 1953. This dissertation, which contains complete original data, is available from University Microfilms, Ann Arbor, Michigan, as Publication No. 6637 (Library of Congress Card No. MicA54-112).

NOTATION

a = exponential constant [Equation (6)]
b = integration constant [Equation (2)]
C = compressibility factor [Equation (4)]
D = die diameter, in.
$\mathfrak{D}$ = diffusivity, sq.in./sec.
d = particle diameter, in.
d_{avg} = geometric mean particle diameter, in.
e = natural logarithm base
F = axial force on powder in die, lb.-force
f = axial force on single particle, lb.-force
g_c = gravitational constant, (in./sec.^{2}) (lb.-mass/lb.-force)
i = proportionality constant [Equation (1)]
k = proportionality constant [Equation (6)]
L = pressing modulus [Equation (2)]
m = proportionality constant [Equation (7)]
N = number of particles in layer normal to direction of pressing
n = proportionality constant [Equation (5)]
P = pressure, lb.-force/sq.in.
V = volume of powder, cu.in.
V_o = relative volume, ratio of total volume to volume of void-free solid
W = frictional work of irreversible compression, in.lb.-force
w = weight of powder fraction of given size, lb.-mass

Greek Letters

α = exponential constant [Equation (3)]
ϵ = porosity, volume fraction of voids
ϵ_o = porosity of uncompressed packing
ρ_a = bulk or apparent density, lb.-mass/cu.in.
ρ_{a_o} = bulk density of uncompressed packing, lb.-mass/cu.in.

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Manuscript received October 28, 1960; revision received January 17, 1962; paper accepted January 18, 1962.