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The effect of dimensions on the compaction properties of sodium chloride*

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Using a tablet press instrumented with strain gauges, anhydrous particulate sodium chloride was compressed to form compacts of different lengths in three dies of different diameters. For the limited range of dimensions applicable to most pharmaceutical tablets, there was a common linear relation between the applied compaction pressure and the force lost to the die wall per unit area of apparent die wall contact, during compression. Ejection forces were correlated using a similar expression. The mechanical strength of compacts was determined by diametrical compression. A relation was proposed to express the strength (F_c), of the compacts of different sizes in terms of the diametrical cross-sectional area at zero porosity ($D.L_o$), the relative volume (V_r) and the mean compaction pressure (P_m):

$$\left(\frac{F_c}{D.L_o}\right) . V_r = k.P_m - c, \text{ where k and c are constants.}$$

Previous investigators of the compaction behaviour of powders have prepared compacts of various lengths in dies of different diameters, and it is often difficult to compare the published results for compression forces, and physical and mechanical properties of the final compacts. The objective of the present investigation was to establish relations between these variables and the dimensions of the compacts produced.

As early as 1882, Forscheimer was aware that the pressure in a column of sand decreases with depth, due to friction between the particulate material and the container wall.

Shaxby & Evans (1923) derived an expression to describe the vertical pressure, P_z, at depth z in a system of non-coherent, particulate material. If gravitational effects are considered negligible in comparison with the applied pressure P_a, the relation becomes:

$$\frac{P_z}{P_a} = e^{-\left(\frac{2cz}{r}\right)} \quad \ldots \quad \ldots \quad \ldots \quad \ldots \quad (1)$$

where r is the radius of the cylindrical container, and c is a constant.

Numerous equations have been proposed which are modifications of the original expression quoted by Shaxby & Evans. For example, Unckel (1945) developed a relation in which the constant c, was equal to the product of the coefficient of friction at the container wall μ, and the stress ratio η (radial/axial stress). In Unckel's equation, P_z became the pressure transmitted through a compact, and the term "z" was equal to the *initial* length of the compact.

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In 1954, Duffield derived the first relation to account for changes in the bulk density of a compact during pressing,

$$\frac{P_b}{P_a} = V_r \cdot e^{-\left(\frac{2\mu\eta L}{r}\right)} \quad \ldots \quad (2)$$

in which P_b is the pressure transmitted through a compact of *actual* length L, and the relative volume V_r, is the ratio of the actual volume of the compact to the volume of solid material present.

The differences between these expressions are characteristic of the disagreements between results obtained for various materials by investigators such as Spencer, Gilmore & Wiley (1950), Ballhausen (1951), Sheinhartz, McCullough & Zambrow (1954) and Toor & Eagleton (1956).

Experimental results obtained by Squire (1947) for metal powders demonstrated that, for rectangular and cylindrical compacts, the density was indirectly proportional to the ratio of wall area to pressing area. Consequently, either a reduction in length for compacts of a certain diameter, or an increase in diameter for a constant length, produced an increase in density.

The mechanical properties of a powder compact may also be affected since they are determined to a great extent by the density. Unfortunately, Squire (1947) made no measurements of frictional effects, or of mechanical properties in this aspect of his investigations.

EXPERIMENTAL

Plane faced punches of 0·8, 1·0 and 1·2 cm diameter, were used in conjunction with a single-punch eccentric tablet press (Lehman). Each upper punch and the lower punch holder were instrumented in a manner similar to that of Shotton & Ganderton (1960), using Saunders Roe $\frac{1}{8}$-inch, linear foil resistance strain gauges.

Sodium chloride was selected as a pure, cubic-crystalline material which forms a coherent compact by direct compression. A batch of sodium chloride (B.P. quality) was screened to obtain a 30–40 mesh fraction, and fine particles were removed using an Alpine Air-Jet Sieve. The density of the characterized material (2·17 g cm^{-3}) was determined pycnometrically at 25° using *m*-xylene.

The weights of material for compression were selected to facilitate comparison of the present results with those of previous workers, including Shotton & Ganderton (1960), who employed a $\frac{1}{2}$-inch (diameter) die. Sufficient material was compressed in each die (Table 1) to produce compacts of the following length, L_0, at zero porosity:

1. $L_0 = 0{\cdot}4$ cm
2. $L_0 = 0{\cdot}4 \times \left(\frac{\text{diameter of respective die}}{\text{diameter of } \frac{1}{2}\text{-inch die}}\right)$ cm
3. $L_0 = 0{\cdot}4 \times \left(\frac{\text{cross-sectional area of respective die}}{\text{cross-sectional area of } \frac{1}{2}\text{-inch die}}\right)$ cm

Compression samples were weighed to $\pm 0{\cdot}5$ mg, dried for 1 h at 110° and stored over silica gel for 24 h before use. The ambient relative humidity was maintained at less than 20%, during the pressing operation, and at least five pressure levels were investigated for each diameter-length combination. Five compacts were prepared at each pressure and the die was cleaned before the preparation of each compact.

To evaluate the influence of die wall "contamination", the experiments with a 1·2 cm (diameter) die were repeated. At each pressure level, the die wall was conditioned by compaction of two samples of material before the compression forces were measured for five subsequent samples.

Resulting compacts were weighed to ±0·1 mg and the dimensions were measured to ±0·005 mm. The diametrical crushing strength was then determined using the apparatus of Shotton & Ganderton (1960). A strict time schedule of experimentation ensured that a constant time interval of 10 min elapsed between the compression and testing of each compact.

RESULTS

The data presented in this section relate the forces of compression to the properties of the compacts produced. In the subsequent discussion, these results are correlated and expressions are derived from them.

The results in Figs 1 and 2 show that both the force lost to the die wall, F_d, and the force required to initiate movement of the compact during ejection, F_e, were increased by a reduction in die diameter, and in most cases by an increase in the

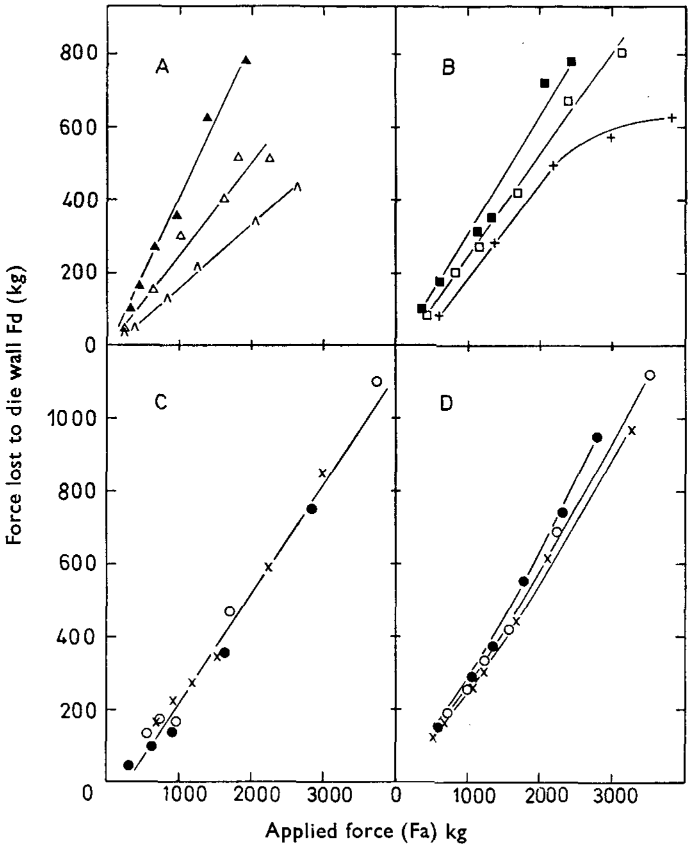

FIG. 1. Effect of die diameter and compact length on the relation between force lost to the die wall and applied force. Length at zero porosity, L_0: ▲, 0·40 cm; △, 0·25 cm; ∧, 0·16 cm; ■, 0·40 cm; □, 0·32 cm; +, 0·25 cm; ●, 0·40 cm; ○, 0·38 cm; ×, 0·36 cm. A. 0·8 cm (diam.) clean die.; B, 1·0 cm (diam.) clean die; C, 1·2 cm (diam.) clean die; D, 1·2 cm (diam.) conditioned die.

length of a compact. A linear relation between F_d and the applied force was observed for all compacts prepared in a clean 0·8 cm and 1·2 cm (diameter) die, and for all except the shortest compact pressed in a clean 1·0 cm die (Fig. 1). In the latter case, F_d approached a constant value above 2000 kg applied force. However, for the conditioned 1·2 cm die, an increase in slope occurred with increasing applied force over the range of conditions investigated (Fig. 1D).

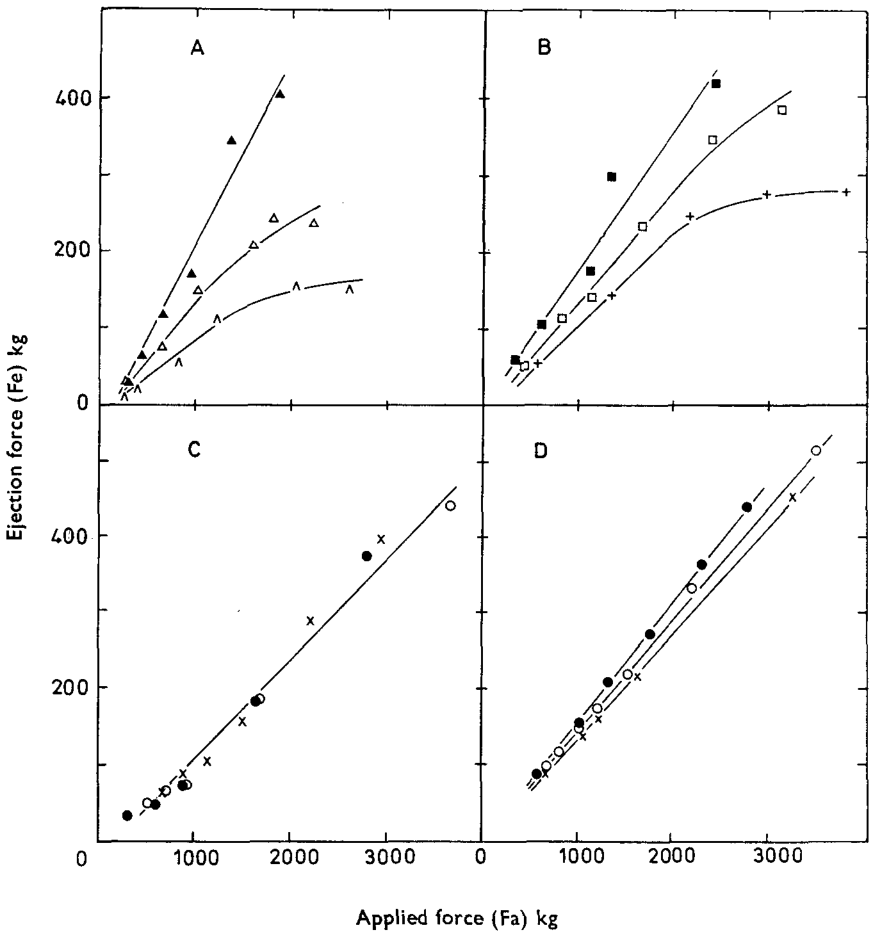

FIG. 2. Effect of die diameter and compact length on the relation between ejection force and applied force. Length at zero porosity, L_0: ▲, 0·40 cm; △, 0·25 cm; ∧, 0·16 cm; ■, 0·40 cm; □, 0·32 cm; +, 0·25 cm; ●, 0·40 cm; ○, 0·38 cm; ×, 0·36 cm. A–D as Fig. 1.

When the shorter compacts were pressed in the 0·8 and 1·0 cm dies, the ejection force approached a maximum value as the applied force increased (Fig. 2A and B).

Conditioning of the die wall produced an increase in the magnitude of F_d and F_e (Figs 1D and 2D), and the deviation of replicate values of F_d from a mean value was shown to be less than for samples pressed in a clean die (Table 2).

Table 1. *The weight, and length at zero porosity, L_o, of samples compressed to evaluate the effect of dimensions on the properties of a compact*

Die diameter (cm)	L_o (cm)	Compression weight (g)
1·2	1. 0·400	0·982
	2. 0·378	0·928
	3. 0·357	0·876
1·0	1. 0·400	0·682
	2. 0·315	0·537
	3. 0·248	0·423
0·8	1. 0·400	0·436
	2. 0·252	0·275
	3. 0·159	0·173

Compression of material in a die of large diameter produced a lower relative volume, V_r, than in a smaller diameter die (Fig. 3A). An increase in length apparently did not affect the relative volume of compacts pressed in the 0·8 and 1·0 cm clean dies, or in a 1·2 cm conditioned die (Fig. 3B). However, the longest compact prepared in a 1·2 cm clean die at low applied pressure possessed a low relative volume compared with the shorter compacts.

At each mean compaction pressure, P_m, an increase in the length of a compact produced an increase in the diametrical crushing strength (Fig. 4). Similarly, for compacts of equal length at zero porosity, an increase in diameter was associated with an increase in strength. Compacts pressed in a clean die were stronger than those prepared in a conditioned die.

Table 2. *Variation in force lost to the die wall for a "clean" and a "conditioned" 1·2 cm (diameter) die*

Weight of compact (g)	Clean die			Conditioned die		
	F_a	F_d	σ	F_a	F_d	σ
0·982	326	45	12·2	584	156	1·5
	611	94	16·2	1043	289	3·2
	883	133	25·7	1340	371	2·1
	1655	355	7·1	1789	555	3·0
	2852	752	11·2	2326	745	1·1
	—	—	—	2796	952	1·5
0·928	538	134	3·6	716	190	4·3
	725	155	14·3	1043	258	4·9
	945	164	14·7	1250	330	2·8
	1716	468	11·8	1562	424	3·4
	3734	1102	9·3	2227	690	3·3
	—	—	—	3533	1123	2·5
0·876	693	153	9·9	520	121	5·5
	916	215	9·9	1063	260	3·1
	1176	273	15·4	1232	310	2·7
	1533	342	15·1	1668	446	2·8
	2244	589	12·5	2125	615	0·8
	2999	857	6·8	3258	967	5·2

σ = standard deviation as % of mean of five replicate values of F_d.

A linear relation between crushing strength and mean compaction pressure was valid for every compact at low pressures. At approximately 2,000 kg cm^{-2} P_m, a deviation from the linear relation occurred, and the strength of compacts tended to a maximum value, or even decreased for the shortest compacts in the smaller dies.

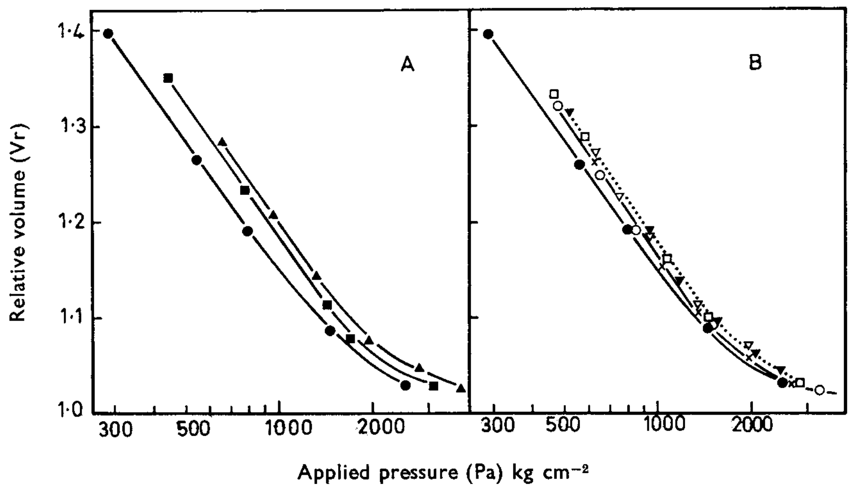

FIG. 3. Relation between relative volume and the applied pressure. A. Effect of diameter on compacts of 0·4 cm length (L_0) at zero porosity. Diameter: *clean die*: ●, 1·2 cm; ■, 1·0 cm; ▲, 0·8 cm.

B. Effect of length on compacts of 1·2 cm diameter. Length at zero porosity (L_0); *clean die*: ●, 0·40 cm; ○, 0·38 cm; ×, 0·36 cm; and *conditioned die* (dotted line): ▼, 0·40 cm; ▽, 0·38 cm; □, 0·36 cm;

DISCUSSION

Compression

In Fig. 5, the experimental results are plotted according to the relation (2) proposed by Duffield (1954), which may be written:

$$\ln \left(\frac{V_r}{R}\right) = 4\mu\eta \left(\frac{L}{D}\right) \quad \ldots \quad (3)$$

where R is the punch force ratio (F_b/F_a), and D is the diameter of the die.

The unsatisfactory correlation of the results in terms of this expression (correlation coefficient: 0·87) may be explained as follows. Contrary to the assumptions of previous authors, including Spencer & others (1950) and Sheinhartz & others (1954), the product $\mu\eta$ is not necessarily constant. From results obtained by compaction of granular polymers, Train & Hersey (1962) concluded that as the applied force increases, the stress ratio, η, also increases, whereas the coefficient of friction at the die wall, μ, decreases as indicated by the friction results of Pascoe & Tabor (1956). In addition, the exponential equation, originally proposed by Shaxby & Evans (1923), assumes that the vertical pressure at a certain depth is uniform across a horizontal section of a column of powder. It has been demonstrated more recently that compaction produces a radial, as well as axial, pressure distribution (Kamm, Steinberg & Wulff, 1947; Train, 1956).

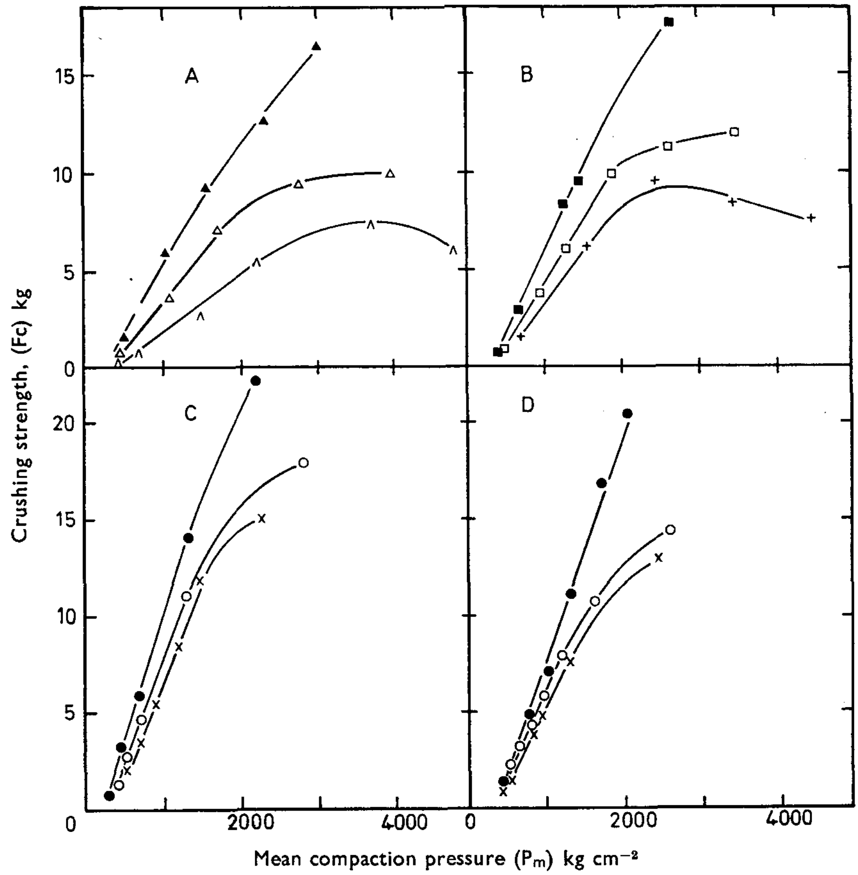

FIG. 4. Effect of diameter and length on the relation between crushing strength and mean compaction pressure. Length at zero porosity, L_0: ▲, 0·40 cm; △, 0·25 cm; ∧, 0·16 cm; ■, 0·40 cm; □, 0·32 cm; +, 0·25 cm; ●, 0·40 cm; ○, 0·38 cm; ×, 0·36 cm. A–D as Fig. 1.

In accordance with the friction theory of Bowden & Tabor (1954), Hersey (1960) proposed that the force lost to the die wall is proportional to the true area of contact, A_t, between the compact and the die wall, provided the shear strength of the compressed material remains constant:

$$F_d = S \cdot A_t \qquad (4)$$

where the shear strength, S, of the junction between the material and the die wall is normally considered to be the shear strength of the weakest material, viz. the compressed material.

Hersey's equation explains the increase in F_d with an increase in the length of a compact pressed in the clean 0·8 and 1·0 cm dies (Fig. 1A and B) and in the conditioned 1·2 cm die (Fig. 1D). However, during the compression of material in a clean 1·2 cm die, the variation in replicate values of F_d was greater than the variation for a conditioned die at the same applied force (Table 2). Despite the precautions taken to standardize techniques, removal of the die for cleaning after each compression will cause a variation in the relative position of the punches and die, and differences in the surface condition of the die wall. Since there are only small differences in the lengths, L_o, of the compacts prepared in the 1·2 cm die, a lack of precision in the values of F_d may obscure a relation between F_d and the length of a compact (Fig. 1C).

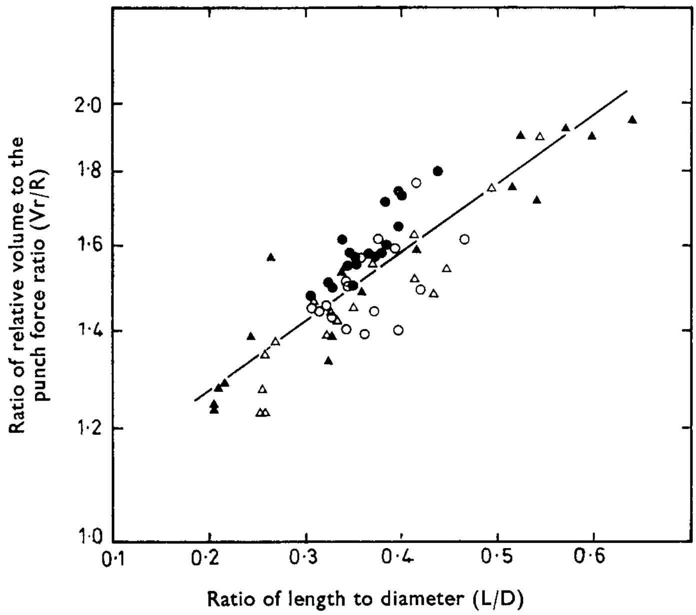

FIG. 5. Experimental results plotted according to the relation proposed by Duffield (1954). ▲, 0·8 cm (diameter) clean die; △, 1·0 cm (diameter) clean die; ○, 1·2 cm (diameter) clean die; ●, 1·2 cm (diameter) conditioned die.

The graph shown in Fig. 6 was obtained by regression analysis of sixty-six co-ordinates, and a correlation coefficient of 0·99 was determined. Thus, for compacts of different lengths compressed in the three dies at applied pressures of up to 4,000 kg cm^{-2}, good correlation of the results was provided by an expression of the form:

$$\left(\frac{F_d}{A}\right) = k_1 \left(\frac{F_a}{A_p}\right) - c_1 \quad \ldots \quad (5)$$

where $k_1 = 0{\cdot}224 \pm 0{\cdot}009$ (95% confidence limits), $c_1 = 51{\cdot}5$, A is the apparent area of contact at the die wall, and A_p is the punch face area.

The validity of this relation indicates that, for the range of dimensions studied, the total force lost to the die wall, F_d, may be considered to increase linearly with increasing distance from the plane of applied force. It is important to note that as the transmitted axial force, F_b, approaches zero in a compact of sufficient length, this linear approximation will become invalid, and equation (5) will not be applicable. However, during the compaction of pharmaceutical tablets it is essential that the force transmitted through the compact should be sufficient to produce adequate consolidation and bonding in regions of the compact remote from the plane of applied force. If the total force lost to the die wall, F_d, is of sufficient magnitude that the transmitted force approaches zero, the powder compact will be unsuitable since pharmaceutical tablets are not subjected to further treatment such as the sintering of metal powder compacts or the firing of ceramics. Accordingly, within the range of conditions appropriate to pharmaceutical powder compaction the type of relation quoted in equation (5) is apparently valid.

Although the force lost to the die wall is greater in a conditioned die, consolidation of the compact is inhibited by the increased frictional resistance. Consequently, equation (5) was also valid for compression in a conditioned die. However, the equation did not correlate results obtained for material which had been exposed to ambient conditions of greater than 66% relative humidity. This effect may be explained by the lubricant effect of liquid films which reduces the friction at interparticulate junctions and at the die wall boundary (Shotton & Rees, 1966).

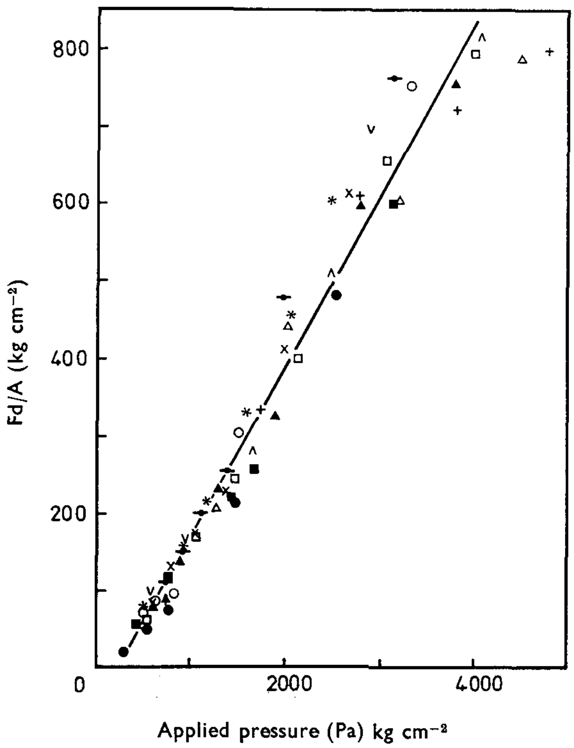

FIG. 6. The force lost per unit area of apparent die wall contact as a function of applied pressure. Length at zero porosity, L_0 (cm):

▲, 0·40; △, 0·25; Λ, 0·16 — 0·8 cm (diameter) clean die;
●, 0·40; ○, 0·38; ×, 0·36 — 1·2 cm (diameter) clean die;
■, 0·40; □, 0·32; +, 0·25 — 1·0 cm (diameter) clean die
*, 0·40; -●-, 0·38; V, 0·36 — 1·2 cm (diameter) conditioned die

Ejection

At a high compaction pressure, compression of the molecular lattice of crystalline material will occur, and subsequent removal of the applied load may permit elastic recovery of the crystal lattice. The residual force on the lower punch due to the die wall friction, and also the force required to eject the compact, will then approach constant values as the applied force increases (Fig. 2A and B).

As shown in Fig. 7, a reasonable correlation of the present results for compacts of different sizes was provided by the expression:

$$\left(\frac{F_e}{A}\right) = k_2 \left(\frac{F_a}{A_p}\right) - c_2 \quad \ldots \quad (6)$$

where $k_2 = 0{\cdot}102 \pm 0{\cdot}007$ (95% confidence limits), and $c_2 = 12{\cdot}5$. For experimental values of F_e at applied pressures of less than 4,000 kg cm^{-2}, the significance of the regression line is defined by a correlation coefficient of 0·97.

A comparison of equations (5) and (6) shows that for anhydrous sodium chloride, the relation between F_d and F_e was linear to 4,000 kg cm^{-2} P_a, and the force lost to the die wall was almost exactly equal to twice the ejection force.

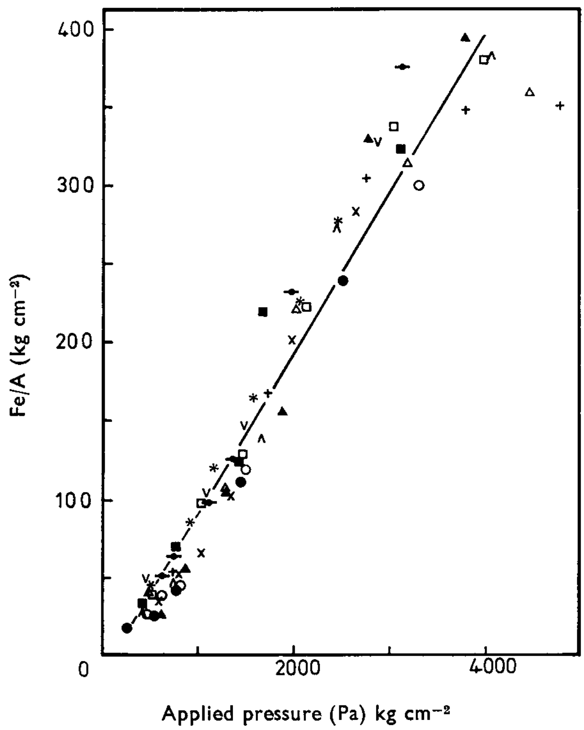

FIG. 7. The ejection force per unit area of apparent die wall contact as a function of applied pressure. Symbols for length at zero porosity as Fig. 6.

Relative volume

At low pressures, there was an exponential relation between the relative volume and the applied pressure (Fig. 3) as reported by Walker (1923) and Bal'shin (1938). This relation cannot be valid when the relative volume approaches unity, and the experimental results suggest that at high pressure, the relative volume decreases with applied pressure according to a power relation.

For compacts of equal length at zero porosity, L_0, a reduction in diameter produced an increase in the relative volume (Fig. 3A). Kamm, Steinberg & Wulff (1947) have shown that die wall friction exerts a retarding effect on the flow of powder during compaction and, as a result, greater consolidation occurs near the axis of a powder compact than at the periphery. It is apparent that the retarding effect of the die wall will influence a larger proportion of the total volume of a small compact than one of larger diameter. Shear failure and consolidation of material in contact with the die wall produces a dense "skin" at the periphery of a compact. However, owing to the small thickness, this boundary layer will have little effect on the relative volume of the total compact.

For the limited range of dimensions studied, the present results showed no evidence of an increase in relative volume with the length of a compact. The unexpected low relative volume recorded for the longest compacts in a 1·2 cm diameter clean die appears to substantiate a similar observation by Bal'shin (1938). He reported that as the length of a tungsten powder compact was increased, there was an initial decrease in the relative volume, before subsequent increase. These observations appear to contradict the results of Walker (1923) and Seelig & Wulff (1946), who demonstrated an increase in the relative volume of a compact with increasing length, but the present results may be rationalized as follows. Boundary effects at the lower punch may restrict local consolidation at the base of a powder compact, and this would significantly increase the relative volume of a short compact. As the depth of material increases, the restraining effect of the plane punch surface may become insignificant, thus reducing the relative volume in a longer compact. A similar effect would occur if the diagonal stress components described by Train (1956) "converge" below the face of the lower punch during compaction of short compacts. As the length of the compact increases, the high density zone observed by Train in the mid-lower-centre of a compact would develop. On the basis of this assumption, an increase in the length of a compact would produce an initial decrease in the relative volume to a minimum value as the high density zone was formed. However, as shown by Seelig & Wulff (1946), a further increase in the length would be expected to increase the relative volume, as the axial stress decreases through the compact.

Strength

The increase in crushing strength with an increase in the dimensions of a compact (Fig. 4) suggested that an expression of the strength in terms of the diametrical cross-sectional area of a compact might permit correlation of the results.

For all compacts prepared in the clean dies at a mean compaction pressure P_m, not exceeding 2,000 kg cm^{-2}, a common linear relation was obtained between P_m and the crushing strength per unit diametrical cross-sectional area at zero porosity (F_c/DL_o). However, due to the higher die wall friction, the compacts pressed in a conditioned die were less dense, and therefore possessed a lower crushing strength than those prepared in a clean die at the same pressure. For this reason, the values of F_c/DL_o for compacts prepared in a conditioned die failed to correlate with the results for clean dies, but the introduction of a term to account for the differences in density provided a common relation of the form:

$$\left(\frac{F_c}{DL_o}\right) \cdot V_r = k_3 \cdot P_m - c_3 \qquad \ldots \quad \ldots \quad \ldots \quad (7)$$

where $k_3 = 0{\cdot}021 \pm 0{\cdot}001$ (95% confidence limits), and $c_3 = 4{\cdot}9$. This expression was valid to 2,000 kg cm^{-2}, P_m (correlation coefficient: 0·97), as shown in Fig. 8.

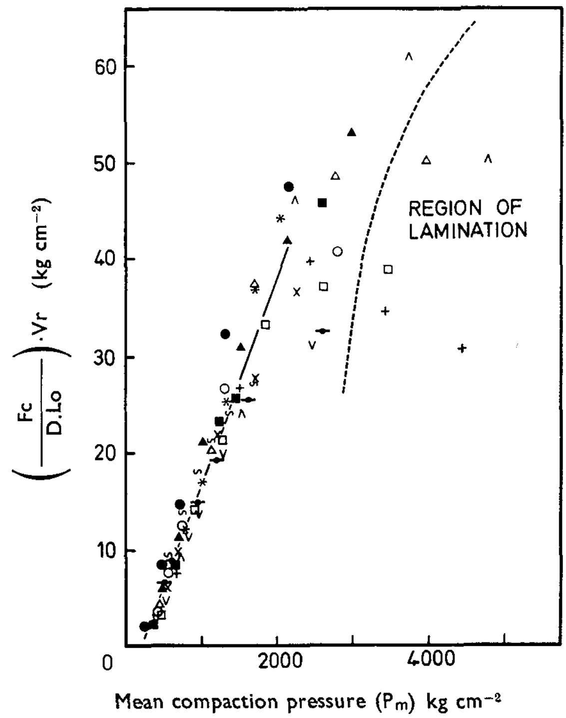

FIG. 8. The product of crushing "stress" and relative volume as a function of mean compaction pressure. Symbols for length at zero porosity as Fig. 6, ~: values derived from experimental results of Shotton & Ganderton (1960).

The correlation of results by the term F_c/DL_0 suggests that the diametrical cross-sectional area at zero porosity, (DL_0), is proportional to the true area of inter-particulate bonding in the diametrical plane of the compact. For the sodium chloride compacts prepared in this investigation, the term F_c/DL_0 may therefore be considered as a "stress". However, in the presence of a lubricant film or any other factor which inhibits bonding between contiguous crystals, the term F_c/DL_0 will not represent a true stress, and may not provide correlation of the results for compacts of different sizes.

The results for crushing strength obtained by Shotton & Ganderton (1960), for compacts of sodium chloride (30–40 mesh size) prepared in a $\frac{1}{2}$ inch diameter die, were converted to values of $(F_c/DL_0)V_r$. As shown in Fig. 8, the derived values were in close agreement with the proposed relation.

As the void space in a compact is reduced at high pressure, the crushing "stress" will theoretically approach a maximum value which is a property of the solid material. However, the co-ordinates in Fig. 8 enclosed by the interrupted line represent those compacts which showed visual evidence of lamination and "capping" following ejection from the die. At high pressures, the deviation from a linear relation was relatively large for short compacts, and for compacts in which the die wall contact area was large in relation to the volume of the compact. It is reasonable to conclude that the decrease in strength of these compacts was due to flaws resulting from the large frictional resistance at the die wall during compaction.

For compacts pressed in clean dies, the dependence of crushing strength on the length at zero porosity, L_0, and not the actual length, L, implies that shear failure and subsequent consolidation of an annulus at the die wall does not produce a significant increase in strength. This evidence confirms that the shearing effect at the die wall boundary was extremely localized and produced only a thin layer of densely packed material.

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