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The effect of particle shape on the mechanical properties of powders

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Summary

This paper investigates the effect of particle shape on the compressional properties of various batches of Starch 1500, sodium chloride, lactose and Emcompress. Measurements were made of their tensile strength, compressibility, elastic recovery, yield values and fragmentation propensities using a Dartec Universal Testing machine. For materials which consolidate by plastic deformation, e.g. Starch 1500 and sodium chloride, there is a large increase in compressibility, a large decrease in yield values and a small decrease in elastic recovery in going from regular to irregular particles. This explains the observed increase in tensile strength which is due to the increased area of contact between particles as they deform. For materials which consolidate by fragmentation, e.g. lactose and Emcompress, the shape of the particles has practically no effect on the above properties. However, the irregular particles are found to fracture more than the regular ones as shown by the values of Pf_B and fragmentation propensity.

Introduction

Although it is often assumed that the strengths of tablets increase with an increase in irregularity of the primary particles, only a few investigations have specifically discussed the relation between particle shape and tablet strength. This arises from the difficulty of obtaining samples of a particular material which have the same particle size but different particle shapes.

The effect of particle shape on tablet strength has been examined by Alderborn and co-workers (Alderborn and Nystrom, 1982; Alderborn et al., 1988). However, they only achieved variations in shape by milling coarse fractions of the materials and the resulting samples were not accurately classified. They concluded that the tablet strengths of plastically deforming materials were markedly affected by a change in particle shape, while for fragmenting materials they were independent of particle shape.

In this work, 90–150-μm sieve fractions of Starch 1500, lactose and Emcompress were shape sorted into regular and irregular particles using a Jeffrey Galion shape sorting table (Ridgway and Rupp, 1969). Irregular NaCl was produced from a coarse fraction of the regular material by milling and was then sieved. The compressibility and elastic recovery of the materials were measured with a

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Dartec Universal testing machine (Dartec Ltd.) and their tendency to fragment determined on a modified Rigden apparatus (Wong et al., 1988b).

Materials and Methods

The 90–150-μm sieve fractions of Starch 1500 (Colorcon Inc.), NaCl (Direct Supplies), lactose and Emcompress (both from Forum Chemicals) were dried and sorted into regular and irregular particles using a Jeffrey Galion shape sorting table (Ridgway and Rupp, 1969). Irregular NaCl was produced by milling a coarse fraction of the regular material and was sieved. The particle densities, ρ_s (g cm^{-3}), were determined with a Beckman air comparison pycnometer (Beckman Instruments, model 930), their surface areas, S_w (m^2 g^{-1}), being measured with a sorptometer (Flowsorb II 2300, Micrometrics, GA). The shape coefficients, α, of the samples were calculated from the expression:

$$\alpha = S_w \rho_s d_c + N \quad (1)$$

This is a modified Heywood expression (Nikolakakis and Pilpel, 1988) where N denotes the elongation ratio (= length, L/breadth, B) and d_c is the Heywood equivalent mean diameter (= $(LB)^{1/2}$). Relevant properties of the powder samples are listed in Table 1 and representative photomicrographs are shown in Fig. 1A–H.

Compressibility and elastic recovery

The powders were compressed in a 10 mm

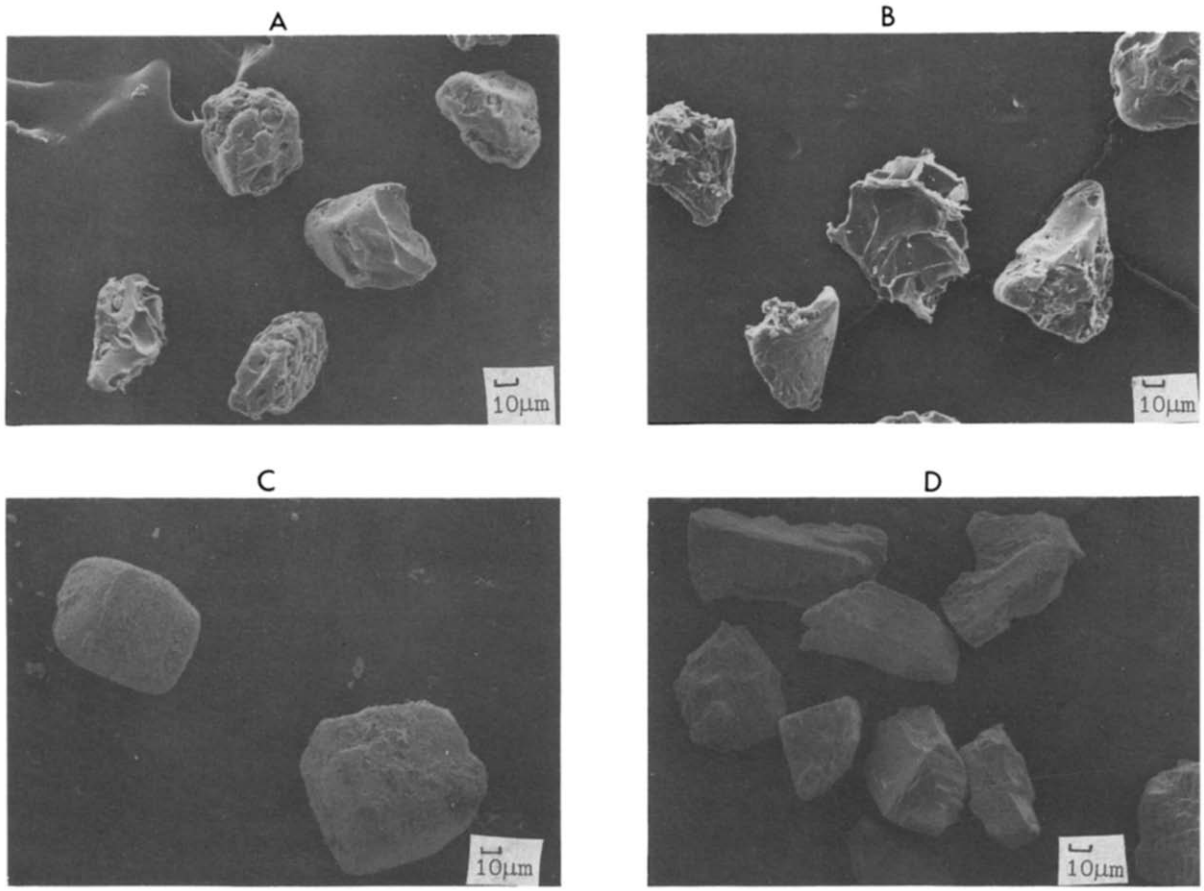

Fig. 1. (A–H). Photomicrographs of the materials used: (A) Starch 1500 1, (B) Starch 1500 12, (C) regular NaCl, (D) irregular NaCl, (E) lactose 1, (F) lactose 12, (G) Emcompress 1 and (H) Emcompress 12.

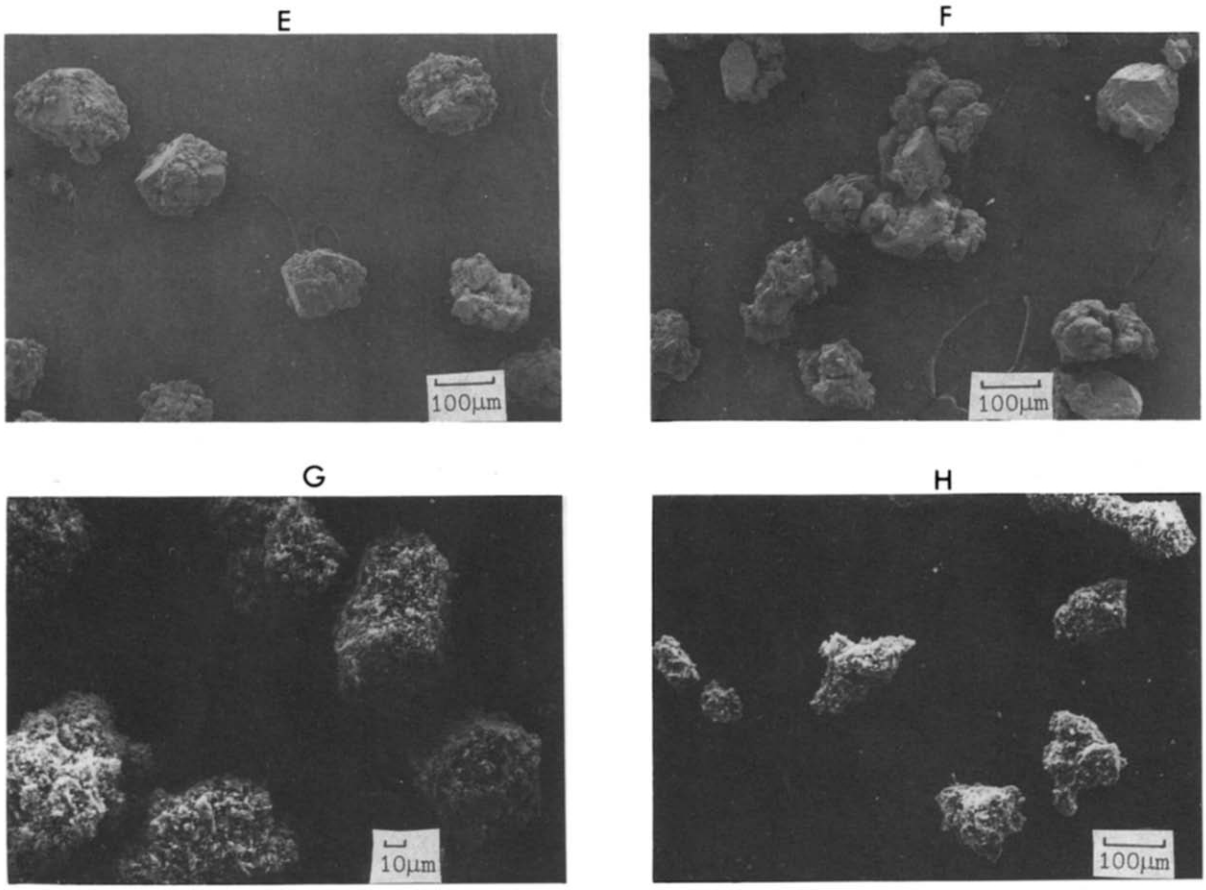

Fig. 1 (continued).

TABLE 1

The relevant properties of the materials

Materials and code		Particle diameter (d_e) (μm)	Elongation ratio (N)	Specific surface area ($cm^2\ g^{-1}$)	Particle density (ρ) ($g\ cm^{-3}$)	Modified Heywood shape coefficient (α)
Starch 1500	1	121	1.02	705	1.46	13.5
	12	127	1.16	913	1.46	18.2
NaCl	(regular)	129	1.01	235	2.17	7.5
	(irregular)	122	1.31	344	2.17	10.4
Lactose	1	109	1.02	828	1.52	14.8
	12	118	1.20	930	1.52	18.0
Emcompress	1	119	1.01	1440	2.35	41.3
	12	127	1.15	1554	2.35	47.7

NB: The numbers in column 1 denote the pot in which samples were collected in the shape sorter.

diameter flat-faced lubricated punch and die set using a Dartec Universal testing machine. Loads of 5, 10, 15 and 20 kN were applied at the rate of 1 kN per s, then held for 30 s while the movement of the upper punch was recorded using a digital storage oscilloscope (Nicolet 3091). The compressibility was defined by

$$C = \frac{(H_p - H_t)}{H_t} \times 100\% \quad (2)$$

where H_p and H_t represent the thickness of the tablet at maximum force and after holding for 30 s, respectively.

After ejection from the die, the tablets were stored in air-tight vials for 24 h and their final thickness H_o was measured. Their elastic recovery, ER, was calculated from

$$ER = \frac{H_o - H_t}{H_t} \times 100\% \quad (3)$$

The value of C will be influenced by a number of experimental variables including the rate of loading (Bangudu and Pilpel, 1985), the magnitude of the applied force, the length of time for which it is held (Bangudu and Pilpel, 1985), and the dimensions and state of the punches and die employed. As measured, the compressibility, C, is a combination of the plastic, elastic and viscoelastic deformation (Staniforth and Patel, 1989) of the materials. Preferably, it should be measured as the applied load is allowed to decay but this was not possible with the present equipment. Nevertheless, it seems reasonable to expect that C and ER,

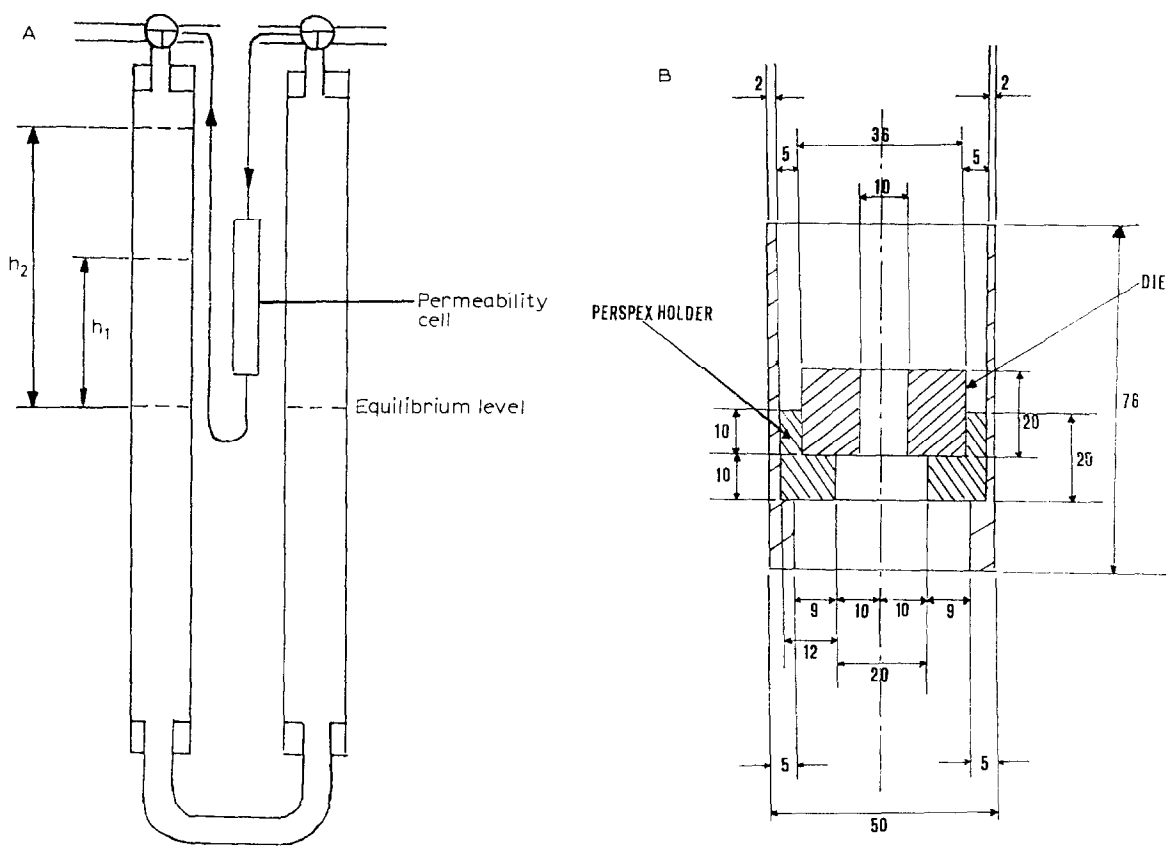

Fig. 2. (A) The Rigden apparatus. (B) The permeability cell. Scale, 1 : 1. All dimensions in mm.

though not the absolute quantities, provide some comparative measures of the changes that occur in the 'plastoelastic' properties of powders as the shapes of the particles are varied.

Diametral compression test

The diametral breaking strength of each tablet was determined using the Dartec machine in its stroke mode. The upper punch was driven at 1 mm s^{-1} onto the tablet which was positioned on its edge on the face of the lower punch. Only the results from clean symmetrical breaks are acceptable for calculating tensile strength, T (York and Pilpel, 1973) from

$$T = \frac{2P}{\pi D_t t_t} \tag{4}$$

where P = load needed to cause failure, D_t = tablet diameter and t_t = tablet thickness.

Fragmentation propensity

In a second series of experiments, the powders were compressed in the Dartec machine using a controlled stroke mode of 1 mm s^{-1} and the die containing the tablet was then removed and inserted into the modified holder of a standard Rigden apparatus (Fig. 2A and B). One hour after compression the permeability of the tablet was measured and its surface area was calculated using the Kozeny Carman equation (see Appendix).

Heckel parameters

For these studies the powders were compressed into tablets in the Dartec apparatus as before. The load and displacement of the upper punch were continuously recorded using a Vela data logger (Data Harvest). The data were then analysed using an Apple IIe computer to determine the various Heckel parameters (Khan et al., 1988).

Results and Discussion

Tensile strength

Graphs of log tablet tensile strength (log T) vs packing fraction are plotted for Starch 1500, NaCl, lactose and Emcompress of different shapes in

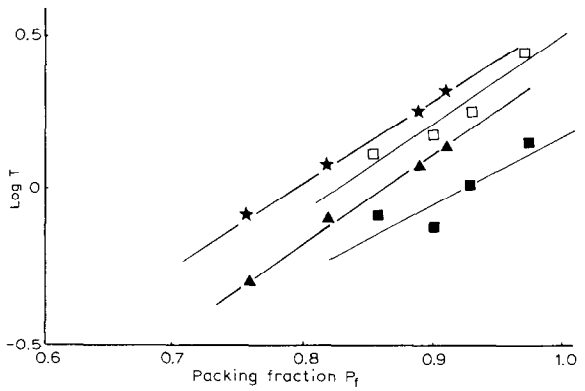

Fig. 3. Log T vs packing fraction (P_f) of Starch 1500 and NaCl tablets. (△) Starch 1500 1, (★) Starch 1500 12, (■) regular NaCl, (□) irregular NaCl.

Figs. 3 and 4 and obeyed the equation (York and Pilpel, 1973):

$$\log T = A_1 P_f + A_2 \tag{5}$$

Where A_1 and A_2 are constants depending upon the particular material. Table 2 gives the equations of the best-fitting straight lines and the correlation coefficients.

It is seen that at all packing fractions the tensile strengths of tablets made from irregular Starch 1500 and NaCl are much higher than those of regular fractions. However, for lactose and Emcompress the difference is much smaller. These

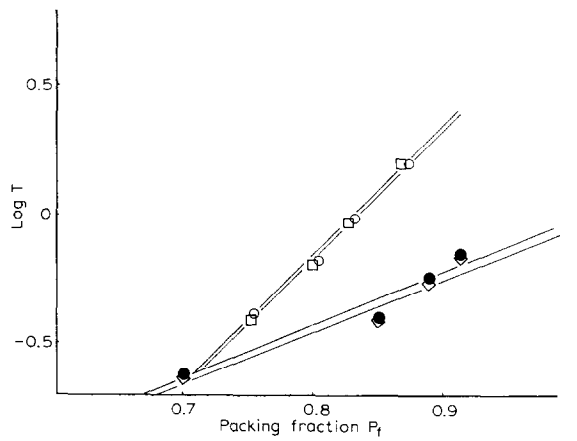

Fig. 4. Log T vs packing fraction (P_f) of lactose and Emcompress tablets. (◇) Lactose 1, (●) lactose 12, (□) Emcompress 1, (○) Emcompress 12.

TABLE 2

Relationship between tensile strength and packing fraction for the different materials

Sample		Equation for best-fitting line	Correlation coefficient
Starch 1500	1	$\log T = 2.885\ P_f - 2.480$	0.997
	12	$\log T = 2.654\ P_f - 2.097$	0.995
NaCl	(regular)	$\log T = 2.157\ P_f - 1.983$	0.800
	(irregular)	$\log T = 2.797\ P_f - 2.305$	0.927
Lactose	1	$\log T = 2.022\ P_f - 2.081$	0.853
	12	$\log T = 2.080\ P_f - 2.097$	0.887
Emcompress	1	$\log T = 4.970\ P_f - 4.163$	0.998
	12	$\log T = 4.909\ P_f - 4.094$	0.999

results agree well with those of Alderborn and Nystrom (1982) and are attributed to the different consolidation behaviour of the two groups of materials.

Starch 1500 and NaCl are thought to consolidate by plastic deformation (Wong et al., 1988a) and this occurs most extensively on the asperities and surface roughness of the irregular particles. It will be expected to produce an increase in the area of contact between these particles and therefore the tensile strength of tablets made from the irregular particles should be greater than from the regular ones. In addition, during the milling of the NaCl crystals, dislocations are produced in the crystal lattice which also increase the plastic deformability of the crystals (Milosovich, 1963; Hüttenrauch, 1988).

In contrast to the above, lactose and Emcompress seem to consolidate by fragmentation (Wong et al., 1988b). A new population of particles is thereby produced and these all tend to be irregular, irrespective of the shape of the original particles. This would account for the results in Fig. 4.

It is now interesting to establish whether the above explanations agree with the results actually obtained during the measurements of compressibility and elastic recovery of the four materials.

Compressibility and elastic recovery

Table 3 lists the percentages of compressibility (C) and elastic recovery (ER) of the materials measured under different forces, calculation of the values being performed using Eqns. 2 and 3. Since C decreases and ER increases with increasing force, as shown in Figs. 5 and 6, it was decided to define the total compressibility and the total elastic recovery as the areas under the appropriate plots between 5 and 20 kN (Table 3). Ignoring the very slight differences, it is seen (a) that the total elastic recovery of all the materials is hardly affected by the particle shape, (b) that essentially the same is true for the total compressibility of lactose and Emcompress but (c) that for Starch

TABLE 3

Values of C, ER, compressibility and total elastic recovery value for the various materials at different forces

Sample		Force (kN)								Total compressibility	Total elastic recovery
		5		10		15		20			
		C	ER	C	ER	C	ER	C	ER		
Starch 1500	1	5.50	2.52	2.80	4.23	1.15	6.60	0.51	7.62	65.0	85.5
	12	6.13	2.43	3.20	3.91	2.50	6.54	1.47	7.48	92.0	81.4
NaCl	(regular)	4.20	0.78	1.46	1.14	0.93	4.00	0.55	5.25	46.5	42.5
	(irregular)	4.90	0.60	2.20	1.10	1.70	3.26	1.20	4.80	64.6	39.4
Lactose	1	1.40	2.20	0.73	2.30	0.75	5.20	0.70	6.15	28.4	66.4
	12	1.50	2.15	0.75	2.20	0.80	5.15	0.73	5.90	29.1	66.4
Emcompress	1	1.17	1.79	0.72	2.29	0.66	4.91	0.31	5.52	19.0	57.2
	12	1.52	1.74	0.87	2.14	0.68	4.45	0.25	5.48	23.9	54.4

Values of C and ER are expressed as percentages, total compressibility and elastic recovery being given in kN%.

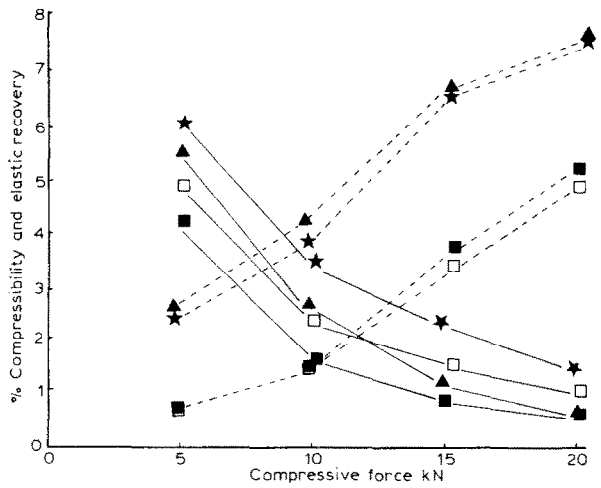

Fig. 5. Total compressibility (———) and elastic recovery (— — —) vs compression for Starch 1500 and NaCl tablets (symbols as in Fig. 3). Compression force is expressed in kN; compressibility and elastic recovery as percentages.

1500 and NaCl the total compressibility of the irregular particles is significantly greater than those for regular ones. It therefore seems that the total compressibility is a more sensitive indicator than the total elastic recovery for differentiating between the effects of particle shape on the compressional properties of powders, particularly those such as Starch 1500 and NaCl which, as mentioned above, consolidate by plastic deformation. For other powders like lactose and Emcompress, the work so far has shown that their particle shape has practically no effect on their tensile strength (Fig. 4), compressibility and elastic recovery (Table 3). It was therefore considered of interest to determine whether their particle shape might affect the extent to which they fragment under load. This aspect is dealt with in the next two sections.

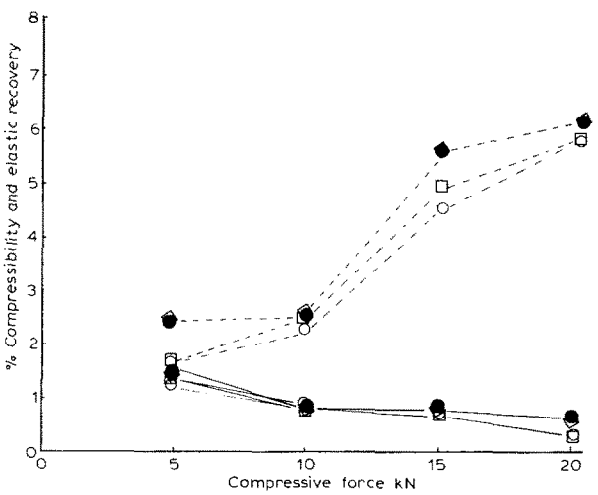

Fig. 6. Total compressibility (———) and elastic recovery (— — —) vs compression force for lactose and Emcompress tablets (symbols as in Fig. 4).

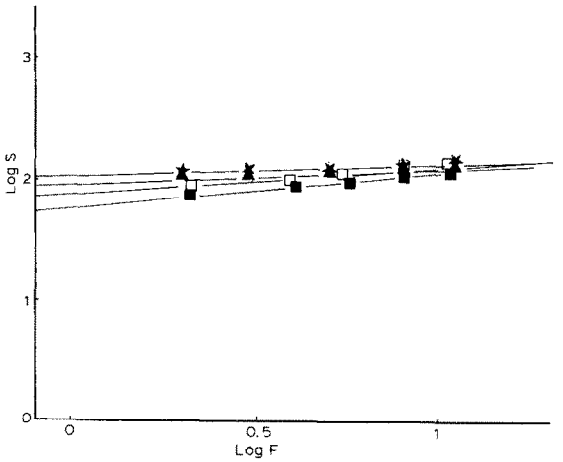

Fig. 7. Log S vs log F for Starch 1500 and NaCl tablets (symbols as in other figures).

Fragmentation propensity

The effects of compaction force F on the surface areas S of the tablets measured by gas permeability on all the materials are depicted in Figs. 7 and 8. Plotting log S vs log F produced straight lines fitted through the experimental points by regressional analysis. The increase in surface area with increase in force for all the materials indicates fragmentation of particles during compaction and the slopes of the lines provide a quantitative measure of the fragmentation propensity.

The fragmentation propensities of all the materials are listed in Table 4. As might have been expected the fragmentation propensities of lactose and Emcompress are much higher than those of Starch 1500 and NaCl (although it may be noted that the latter two do exhibit some fragmentation during compaction even though the predominant mechanism is plastic).

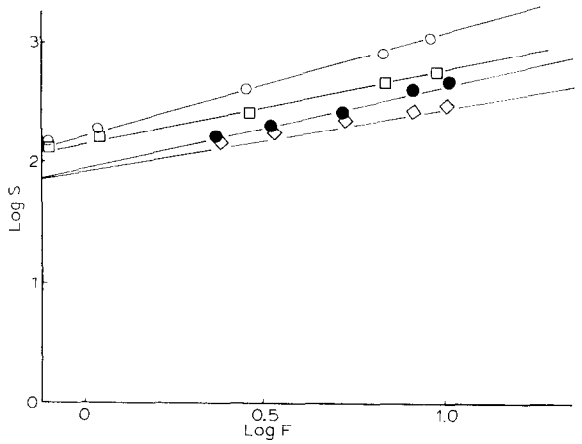

Fig. 8. Log S vs log F for lactose and Emcompress tablets (symbols as in other figures).

The shape of the particles has practically no effect on the fragmentation propensities of Starch 1500 and NaCl whereas it exerts quite a large effect on those of lactose and Emcompress indicating that their irregular particles are more brittle than regular ones.

By studying the changes in relative density of the powders during consolidation and employing the Heckel equation (Heckel, 1961a,b), one can obtain additional information on whether a particular powder is consolidating by fragmentation or by plastic deformation. This is dealt with in the following section.

TABLE 4

Fragmentation propensities of the different materials

Sample		Fragmentation propensities (m^{-1} kN^{-1})
Starch 1500	1	0.136
	12	0.128
NaCl	(regular)	0.303
	(irregular)	0.287
Lactose	1	0.507
	12	0.680
Emcompress	1	0.603
	12	0.869

Heckel plots

Table 5 gives the various parameters derived from the Heckel plots of $\ln\{1/(1-P_f)\}$ vs compression force. Starch 1500 and NaCl are characterised by low yield and Pf_B values (densification from particle movement and rearrangement). It is also seen that for these materials the yield values of the irregular fractions are lower than those of the regular ones. Therefore, the irregular materials are more plastic than the regular ones, which supports the earlier results on tensile strength and compressibility. As expected, the values of Pf_0 which reflect the initial packing density are higher for the regular materials. There is, however, scarcely any noticeable difference in the Pf_B values of the regular and irregular materials.

In contrast to the above, lactose and Emcompress are characterised by high Pf_B and yield values. For these materials, there is hardly any difference in the yield values of the regular and irregular powders. This agrees well with the previous findings concerning the tensile strength and compressibility. The Pf_0 values of the regular materials are again higher than those of the irregular ones. It is also interesting to note here that the Pf_B values of the irregular materials are noticeably higher than those of the regular ones. This indicates that the irregular lactose and Emcompress undergo more rearrangement and slippage as a result of being more brittle, as shown by their fragmentation propensities.

TABLE 5

Values of the various Heckel parameters

Sample		Pf_0	Pf_B	Yield values (MP_a)
Starch 1500	1	0.437	0.035	42.0
	12	0.400	0.041	34.0
NaCl	(regular)	0.466	0.065	83.0
	(irregular)	0.442	0.069	72.3
Lactose	1	0.563	0.112	154
	12	0.410	0.173	150
Emcompress	1	0.338	0.342	264
	12	0.225	0.445	261

Conclusions

From this work it can be concluded that:
(1) For materials which consolidate by plastic deformations, e.g. Starch 1500 and NaCl, there is a large increase in compressibility, a large decrease in yield value and a small decrease in elastic recovery in going from regular to irregular particles. This explains the observed increase in tablet tensile strength which is due to the increased area of contact between particles as they deform.
(2) For materials which consolidate by fragmentation, e.g. lactose and Emcompress, the shape of the particles has practically no effect on the above properties. However, the irregular particles are found to fracture to a greater extent than the regular ones as shown by the values of Pf_B and fragmentation propensity.

Appendix

Calculation of the specific surface area of the tablets

The specific surface area of the tablets was calculated with the Carman-Kozeny equation corrected for slip flow. When the top of the powder bed is open to the atmosphere, the Carman-Kozeny equation for laminar flow is

$$S_k = 2A_f\epsilon^3 g\rho_L t'/ \left(5(1-\epsilon)^2\eta L a_m\{[(2V_c/a_m B) + 1]\ln[\{h_2(2B-h_1)/h_1(2B-h_2)\} - 4\ln\{(2B-h_1)/(2B-h_2)\}]\}\right) \quad (6)$$

The contribution of molecular slip to the surface area is

$$S_M = \frac{81 S_K^2 \eta(1-\epsilon)\sqrt{T_p}}{P\epsilon} \quad (7)$$

In this work, V_c was small and the flow of air is arrested well before equilibrium conditions are reached and therefore Eqn. 6 approximates to

$$S_K = \sqrt{\left\{\frac{2A_f\epsilon^3 g\rho_L t'}{5(1-\epsilon)^2\eta L a_m \ln\left(\frac{h_2}{h_1}\right)}\right\}} \quad (8)$$

The total surface area can then be calculated by combining Eqns. 7 and 8:

$$S_V = \frac{S_M}{2} + \sqrt{\frac{S_M^2}{4} + S_K^2} \quad (9)$$

Glossary

Symbol	Meaning
S_K	effective permeability volume specific surface of a powder assuming only viscous flow occurs in the determination
S_M	effective permeability volume specific surface of a powder assuming slip flow occurs in the determination
S_V	effective permeability volume specific surface of a powder
η	the viscosity of air at its temperature (1.045 g cm^{-3})
ϵ	porosity of a powder bed
T_p	temperature at the time of determination
ρ	particle density
A_f	cross-sectional area of a bed of powder perpendicular to the direction of flow
g	acceleration due to gravity
ρ_L	density of a manometric liquid during the determination at the selected operating temperature (1.045 g cm^{-3})
t'	time taken for the manometric liquid level to pass between the upper and lower timing marks
L	height of the powder bed
a_m	manometric cross-sectional area of a constant volume apparatus (4.54 cm^2)
h_1, h_2	manometric heights corresponding to the first and second timing marks on a constant volume apparatus
V_c	volume of a constant volume apparatus between the manometer equilibrium level and the bottom of the powder bed
B	atmospheric pressure at the time of the determination as indicated by a height of manometer fluid

Acknowledgement

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