The effect of the relationship between punch velocity and particle size on the compaction behaviour of materials with varying deformation mechanisms

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The effect of punch velocity over the range 0·033–300 mm s⁻¹ on the compaction properties of lactose, microcrystalline cellulose and a drug substance (a phthalazine derivative) for a range of particle sizes has been studied using the yield pressure derived from the Heckel relationship and a strain rate sensitivity index (SRS index), as the criteria to describe their behaviour. For lactose, a material which deforms by a mixed mechanism of particle fracture and plastic deformation at the contact points, the yield pressure increased and the SRS index decreased as particle size decreased, due to a reduction in the amount of fragmentation of the particles. For microcrystalline cellulose, a material which is known to deform plastically, the yield pressure and the SRS index were independent of particle size. For the phthalazine derivative the yield pressure increased as particle size decreased; however the SRS index reduced from 41% to zero, indicating that the deformation mechanism was changing from plastic flow to brittle behaviour. This decrease in the SRS index has been explained in terms of the relative amounts of strain-hardened material produced as milling severity increased, resulting in an increasing resistance to deformation and thus an apparent increase in brittle behaviour as particle size decreased.

The most widely used equation relating the relative density, D, of a powder bed during compaction to the applied pressure, P, is the Heckel equation (Heckel 1961a, b):

$$\ln [1/(1 - D)] = KP + A \qquad (1)$$

The slope of the straight line portion K, is generally expressed as a reciprocal and is referred to as the mean yield pressure P_y. From the value of A the intercept, the relative density D_A, can be calculated using equation (2).

$$D_A = 1 - e^{-A} \qquad (2)$$

The value of D_0, which is the relative density of the powder bed at the point when a measurable force is applied, has also been used to describe the initial rearrangement phase of densification (Roberts & Rowe 1985).

The effect of varying particle size of a material on the constants derived from the Heckel equation has been investigated by a number of workers. For plastically deforming materials (sodium chloride and potassium chloride) Hersey et al (1973) and Humbert-Droz et al (1982) reported that measured yield pressures, P_y, were independent of particle size. However for materials that are thought to deform by particle fragmentation (lactose and calcium carbonate), Hersey et al (1973) and York (1978) found that yield pressures increased with a reduction in particle size. Recently Roberts & Rowe (1985) extended the concept by investigating the effect of punch velocity on the mean yield pressure of a wide variety of materials and have shown that each material can be characterized by a strain rate sensitivity index.

The purpose of the present study is to investigate the relationship between punch velocity and mean particle size to obtain information on the brittle/ductile transition of a material as its particle size is varied.

MATERIALS AND METHODS

Materials
The three materials used were lactose monohydrate (Dairy Crest, Industrial Division, Milk Marketing Board, Thames Ditton, Surrey, UK)—an example of material that undergoes particle fragmentation during compaction; microcrystalline cellulose (Avicel—F.M.C. International, Food and Pharmaceutical Products, Little Island, Cork, Eire)—a material that undergoes plastic deformation, and a drug substance, a substituted phthalazine derivative.

A range of particle sizes for lactose and micro-crystalline cellulose were obtained by selecting suitable grades from the respective manufacturers as indicated in Table 1. Additionally a batch of Lactose 170 was fluid energy-milled (4″ microniser, F. W. Berk and Co. Ltd, London, UK) to obtain a smaller particle size than those supplied. To obtain a range of particle sizes of the phthalazine derivative, this was milled using a low energy swing hammer mill (Apex Construction Ltd, Gravesend, Kent, UK), a high energy swing hammer mill (Bramigk and Company, London, UK) and a fluid energy mill. The median particle sizes and specific surface areas of the materials were evaluated using an air jet sieve (Alpine Model 320 LS, Alpine Process Technology Ltd., Runcorn, Cheshire, UK) and an air permea-bility technique (Fischer Sub Sieve Sizer, Model 95, KEK Ltd, Macclesfield, Cheshire, UK), respec-tively: these are presented in Table 1.
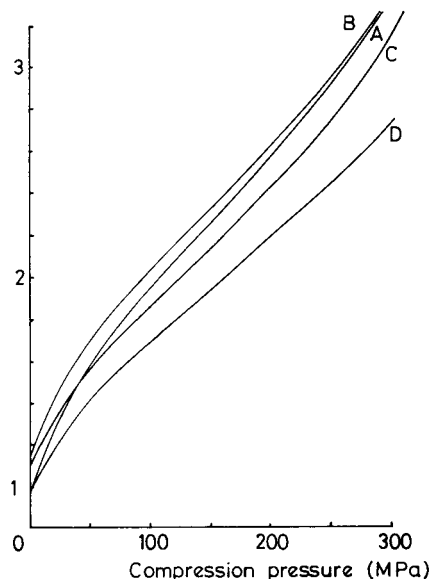

FIG. 1. Heckel plots for the four grades of lactose at a punch velocity of 3·33 mm s^{-1}. (A) Lactose 50, (B) Lactose reg., (C) Lactose 170, (D) Lactose 170 (fluid energy-milled).
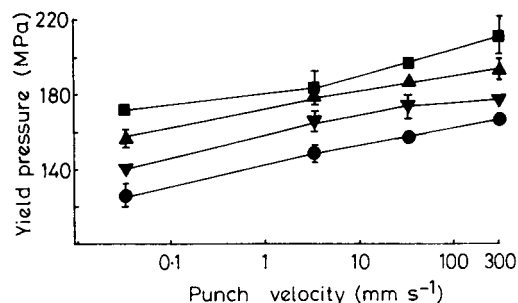
Table 1. Particle sizes, specific surface areas and true densities of the three materials.

Material	Median particle size μm	Specific surface area m^2 g^{-1}	True density g cm^{-3}
Lactose 50	105	0·065	1·529
Lactose REG	50	0·340	1·529
Lactose 170	21	0·610	1·530
Lactose 170 (FEM)	(3·1)	1·250	1·532
Avicel PH102	83	0·280	1·523
Avicel PH101	40	0·280	1·514
Avicel PH105	17·5	0·760	1·510
Phthalazine deriv. (LESH)	(26·8)	0·140	1·685
Phthalazine deriv. (HESH)	(5·3)	0·670	1·676
Phthalazine deriv. (FEM)	(1·3)	2·840	1·671

( ): Median particle size evaluated from specific surface area values.
(FEM): Fluid energy mill.
(LESH): Low energy swing hammer mill.
(HESH): High energy swing hammer mill.

FIG. 2. Yield pressure versus punch velocity for: ●, Lactose 50; ▼, Lactose reg.; ▲, Lactose 170; ■, Lactose 170 (fluid energy-milled).

The compression technique and data manipulation method were as previously described (Roberts & Rowe 1985).

The strain rate sensitivity of each material was calculated from the relative increase in the yield pressure, P_y, between the punch velocities of 0·033 and 300 mm s^{-1}, expressed as a percentage.

RESULTS AND DISCUSSION

The effect of particle size of lactose on both its compaction behaviour at a punch velocity of 3·33 mm s^{-1} and its sensitivity to punch velocity are shown in Figs 1 and 2. For the powder grades of lactose, an increase in the particle size resulted in an increase in the relative density for a given applied pressure (Fig. 1). This is probably due to the ability of the smaller particle size fraction to occupy the interparticle voids between the larger crystals. During compaction, fragmentation of the larger angular shaped crystals and filling of the remaining inter-stices lead to a further increase in the relative density of the compact as reported by McKenna & McCaf-ferty (1982). Furthermore, for the smaller particle size material, the increased frictional forces asso-ciated with particle sliding tend to restrict densifi-cation.

The Heckel plot for the crystalline grade, Lactose 50, does not conform to the expected trend. This is

Table 2. D_0 and D_A values (relative density) at varying punch velocities.

Materials	Punch velocity mms^{-1}							
	0·033		3·33		33·3		300	
	D_0	D_A	D_0	D_A	D_0	D_A	D_0	D_A
Lactose 50	0·650	0·754	0·608	0·716	0·602	0·706	0·594	0·695
Lactose reg.	0·707	0·768	0·680	0·752	0·667	0·748	0·650	0·737
Lactose 170	0·686	0·756	0·656	0·730	0·642	0·720	0·621	0·703
Lactose 170 (FEM)	0·659	0·748	0·613	0·691	0·611	0·694	0·589	0·690
Avicel PH102	0·442	0·508	0·387	0·495	0·396	0·469	0·333	0·432
Avicel PH101	0·450	0·495	0·400	0·465	0·397	0·452	0·353	0·425
Avicel PH105	0·486	0·529	0·436	0·480	0·452	0·477	0·396	0·458
Phthalazine deriv. (LESH)	0·763	0·804	0·723	0·780	0·721	0·783	0·703	0·793
Phthalazine deriv. (HESH)	0·720	0·772	0·702	0·752	0·680	0·733	0·666	0·732
Phthalazine deriv. (FEM)	0·643	0·698	0·613	0·644	0·608	0·659	0·596	0·651

For details of the abbreviations in brackets, refer to Table 1.

due to the more uniformly sized angular crystals of this particular grade, which, because of its crystal shape and lack of finer sized material, will tend to have a lower initial packing density as reflected in the low value of D_0 (Table 2). Thus the compaction process begins at a lower relative density resulting in the Heckel plot being displaced down the y-axis (Fig. 1). The rate of densification for this grade is greater than for the other grades (Fig. 1), probably due to the high stress concentrations at contact points, as a consequence of the angularity of the crystals, from which cracks can easily develop. Thus fractured particles are able to fill the interparticle void spaces in a progressive manner as the fracturing process proceeds during the application of a compression force.

For lactose the yield pressure P_y increased (Fig. 2) as the particle size decreased at any punch velocity. This supports the findings of Hersey et al (1973) who suggested that, as particle size decreases, the stress necessary to cause fracture of particles along cracks increases as predicted from crack theory (Griffiths 1920). As the particle size of lactose decreases the initial particle fragmentation mechanism responsible for filling voids and increasing relative density is reduced, as is evident from the decrease in the strain rate sensitivities—32·4, 24·8, 21·3 and 21·0% for Lactose 50, Lactose Reg, Lactose 170 and Lactose fluid energy-milled, respectively.

It is apparent, therefore, that for lactose the transition from a brittle crystal, due to the presence of fissures, to purely ductile material occurs at a median particle size of approximately 20 μm. Lactose probably densifies by a mixed mechanism involving fracture and plastic flow at the contact points as previously reported by Roberts & Rowe (1985).

For microcrystalline cellulose, the initial packing density D_0 decreases (Table 2) as particle size increases. This indicates that the irregularity of the particles increases with particle size, as is also evident from photomicrographs presented by Shangraw et al (1981), who reported an increase in aggregation for Avicel PH102 when compared with Avicel PH101. Microcrystalline cellulose has been reported to consolidate predominantly by plastic flow (David & Augsburger 1977) due to the numerous slip planes and dislocations present in its structure (Lamberson & Raynor 1976).

For microcrystalline cellulose, the Heckel plots (Fig. 3) and hence the mean yield pressure (Fig. 4),
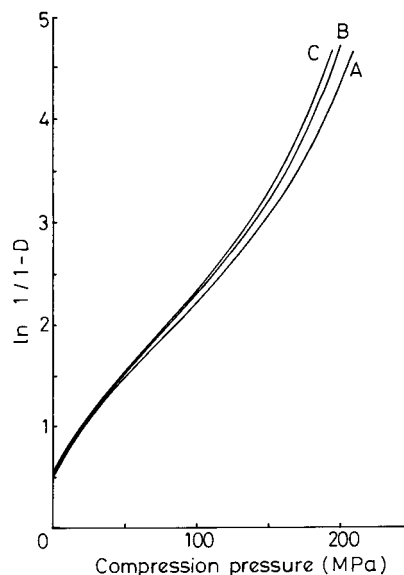

FIG. 3. Heckel plots for the three grades of microcrystalline cellulose at a punch velocity of 3·33 mm s^{-1}: (A) Avicel PH102, (B) Avicel PH101, (C) Avicel PH105.
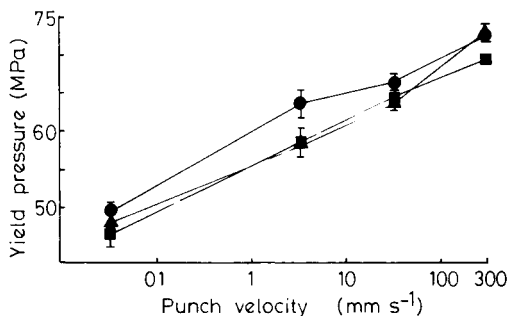
FIG. 4. Yield pressure versus punch velocity for: ●, Avicel PH102; ■, Avicel PH101; ▲, Avicel PH105.
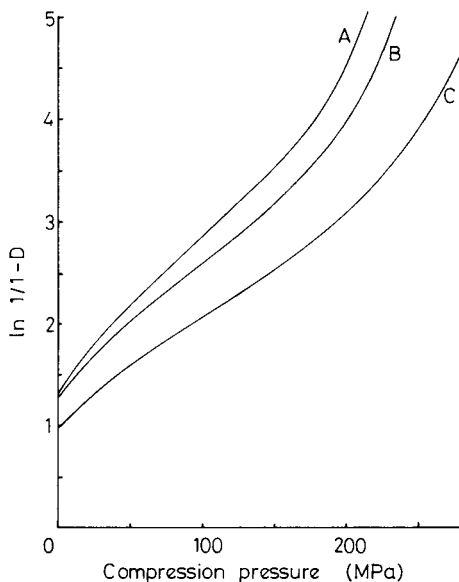

FIG. 5. Heckel plots for the phthalazine derivative at a punch velocity of $3 \cdot 33$ mm s^{-1}, milled using the following; (A) a low energy hammer mill, (B) a high energy hammer mill, (C) a fluid energy mill.

are independent of particle size. This was expected for a material that deforms by a mechanism involving plastic flow, relying purely on dislocations, and supports the findings of McKenna & McCafferty (1982). However, some differences are discernible in that there is a slight decrease in the relative densities and a decrease in the strain rate sensitivities with increasing particle size, ($52 \cdot 4\%$ for Avicel PH105, $50 \cdot 6\%$ for Avicel PH101 and $48 \cdot 3\%$ for Avicel PH102). Thus, as aggregate size increases, densification will include an additional initial fragmentation of the particles, tending to restrict the main deformation mechanism of plastic flow, and lowering the strain rate sensitivity.

The compaction of the phthalazine derivative shows similarities to that of lactose in that an increase in particle size resulted in an increase in the apparent relative density (Fig. 5). This suggests that the phthalazine derivative densifies by a mechanism involving fracture of the initial particles. The decrease in relative density as particle size decreases at any applied pressure (Fig. 5) is associated with an increase in the cohesiveness and friction of the particles. These effects would tend to restrict particle sliding and rearrangement and thus reduce densification.

The effect of punch velocity on the compaction of the different particle sizes of the phthalazine derivative is significantly different from the other two materials (Fig. 6). As particle size decreases, the slope of yield pressure versus punch velocity decreases, to the extent that for the very fine material its value approaches zero. This indicates that the mechanism of compaction is changing from plastic deformation to brittle fracture as reflected in a reduction in the strain rate sensitivity values of $40 \cdot 9\%$ for low energy swing hammer-milled, $24 \cdot 4\%$ for high energy swing hammer-milled and 0% for fluid energy-milled materials.

However, it would be expected that as particle size decreased the brittleness of particles would be reduced because of the lower probability of cracks being present in the crystal structure. This contradicts the proposed deformation mechanism, implying that other processes must be involved. It has been reported that increasing milling severity, as well as reducing particle size, increases the disorder of the crystal lattice (Pearce & Lewis 1972) and thus increases the surface energy of the material (Huttenrauch 1983). For a material that plastically deforms, milling will increase the dislocation density (Suryanarayanan & Mitchell 1985) and thus increase its strain-hardening, resulting in an increased resistance to deformation (Jetzer et al 1983). The large decrease in the strain rate sensitivity index and the reduction in the slope of yield pressure versus punch velocity (Fig. 6) with decreasing particle size, can thus be explained in terms of the relative amounts of work-hardened material produced during milling. Therefore, the fluid energy-milled material must have a higher proportion of strain-hardened crystals because it behaves as though it was a brittle material (zero strain rate sensitivity).

Lactose showed no signs of strain hardening, although it was milled under similar conditions to that used for the phthalazine derivative. Strain hardening occurs when the dislocation density
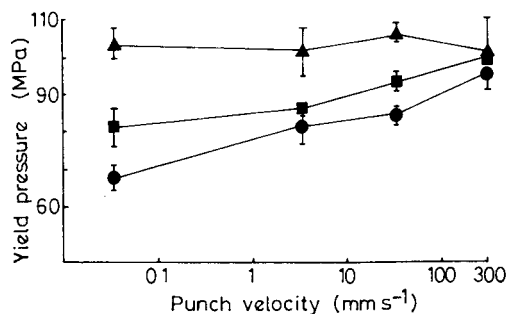
FIG. 6. Yield pressure versus punch velocity for the phthalazine derivative; ●, low energy hammer-milled; ■, high energy hammer-milled; ▲, fluid energy-milled.

reaches a critical point and the dislocations form tangled networks due to the intersection of different slip planes (Hayden et al 1965). The stress necessary to cause dislocation motion during milling will depend on the deformation properties of the material. The yield pressure reflects these differences; for the phthalazine derivative (low energy-milled) and lactose 170, the yield pressures were 81 MPa and 178 MPa, respectively (measured at a punch velocity of 3·33 mms⁻¹). Therefore, for lactose the energy provided during milling is not enough to provide extensive dislocation motion and entanglement. Finally at a punch velocity of 300 mm s⁻¹, particle size effects for the phthalazine derivative are diminished because of the similarities in the yield pressures (Fig. 6). Therefore, rate effects are the most significant parameter affecting the compressibility of this material.

In conclusion it can be seen that, as well as providing information on the deformation properties of materials in terms of the yield pressure, a study of the effects of punch velocity adds a further dimension to our understanding of the compaction process.

In a previous study (Roberts & Rowe 1985), we demonstrated that the strain rate sensitivity index could be used to rank materials in terms of their brittle and ductile behaviour. In this study we have extended the concept by showing the importance of not only the particle size of the material but also the way in which mechanical pretreatment can effect material properties.

REFERENCES

David, S. T., Augsburger, L. L. (1977) J. Pharm. Sci. 66: 155–159

Griffiths, A. A. (1920) Phil. Trans. R. Soc. Lond. Ser. A. 221: 163–172

Hayden, H. W., Moffatt, W. G., Wulff, J. (1965) The Structure and Properties of Materials Volume III Mechanical Behaviour. J. Wiley & Sons, New York, p. 123

Heckel, R. W. (1961a) Trans. Metall. Soc. A.I.M.E. 221: 671–675

Heckel, R. W. (1961b) Ibid. 221: 1001–1008

Hersey, J. A., Cole, E. T., Rees, J. E. (1973) Proceedings of the First International Conference on the Compaction and Consolidation of Particulate Matter, Brighton, October 1972, Powder Advisory Centre London, pp 165–172

Humbert-Droz, P., Mordier, D., Doelker, E. (1982) Pharm. Acta Helv. 57: 136–143

Huttenrauch, R. (1983) Pharm. Ind. 45: 435–440

Jetzer, W., Leuenberger, H., Sucker, H. (1983) Pharm. Tech. 7 (4): 33–39

Lamberson, R. L., Raynor, G. E. (1976) Man. Chem. Aerosol News April: 55–61

McKenna, A., McCafferty, D. F. (1982) J. Pharm. Pharmacol. 34: 347–351

Pearce, C. E., Lewis, D. (1972) J. Catalysis 26: 318–325

Roberts, R. J., Rowe, R. C. (1985) J. Pharm. Pharmacol. 37: 377–384

Shangraw, R. F., Wallace, J. W., Bowers, F. M. (1981) Pharm. Tech. 5 (9): 69–78

Suryanarayanan, R., Mitchell, A. G. (1985) Int. J. Pharm. 24: 1–17

York, P. (1978) J. Pharm. Pharmacol. 30: 6–10