

Effects of paraffin coatings on the shearing properties of lactose

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An annular shear cell has been employed to measure the shearing properties of lactose powder coated with up to 10^{-4} mol g^{-1} of powder of a series of paraffin hydrocarbons. The angle of internal friction, Δ , of the coated powders was independent of their packing density. The shearing properties of the powders depended on the concentration, the viscosity and thickness of the coating material.

The presence of coatings on the surfaces of the particles in a powder has been shown to have marked effects on its tensile strength (York & Pilpel 1973; Pilpel & Hephher 1977; Malamataris & Pilpel 1980; Irono & Pilpel 1982). These effects can be ultimately explained in terms of changes produced in van der Waals' and other forces that act between the particles.

We have previously shown (Irono & Pilpel 1982) that the tensile strength of lactose depended on the nature and amount of the coating and on the temperature employed during the experiments. The purpose of the present investigation has been to extend the work by measuring the changes produced in the shearing properties of lactose coated with the same paraffin hydrocarbons and tested at temperatures from -6 to $+50$ °C.

MATERIALS AND METHODS

The materials used were the same as in the previous investigation: lactose powder B.P. from Whey Products Ltd; light liquid paraffin B.P.C. and liquid paraffin B.P. from McCarthy Ltd; white soft paraffin B.P. and yellow soft paraffin B.P. from Evans Medical Ltd; paraffin wax B.P. from Hopkins and Williams Ltd; and diethyl ether (analytical reagent from Fisons).

Samples of lactose in the size range $10-30$ μm were coated as before with between 0.5 and 10.0×10^{-5} mol g^{-1} of the different paraffins by treatment with ethereal solutions containing the appropriate amount of paraffin.

Shear stress tests

The design of the annular shear cell has been described (Carr & Walker 1968; Kočova & Pilpel 1972) but it was modified for operation at both low

and elevated temperatures (York & Pilpel 1973), thus enabling controlled alterations to be made to the viscosities of the paraffin coatings on the lactose. The cell was either enclosed in a cooling chamber into which pre-cooled dry nitrogen gas was passed or in a heating chamber controlled by a Variac power unit, Fig. 1. The cell was maintained at the required temperature, ± 0.5 °C, for 30 min.

Different consolidating loads up to about 25 N were applied to obtain yield loci at different packing fractions and the procedure was repeated at a series of different temperatures. A fresh unsheared bed of powder was prepared for each consolidating load.

If the shear stress is assumed to have developed uniformly over the annular area of the shear cell, the total measured torque is given by (Carr & Walker 1968):

$$Q = \int_{r_2}^{r_1} \tau r^2 \pi dr$$

$$\text{or} \quad Q = 2\pi \tau (r_2^3 - r_1^3)/3 \quad (1)$$

where τ is the shear stress in Nm^{-2} , r_1 is the inner radius of the cell in metres, r_2 is the outer radius of the cell in metres.

Values of shear stress were calculated from the torque developed (Carr & Walker 1968) after initially calibrating the strain gauges with known torques, applied using a pulley system. The calibration was checked at the different temperatures of investigation.

RESULTS

Fig. 2a shows representative yield loci for lactose powder coated with 0.5×10^{-5} mol g^{-1} of light liquid paraffin at 20 °C plotted in the usual way with the normal stress σ_N , Nm^{-2} as the abscissa and the shear stress at failure τ , Nm^{-2} as the ordinate. The line joining the end points of the loci to the origin is the critical compaction yield locus and its slope is

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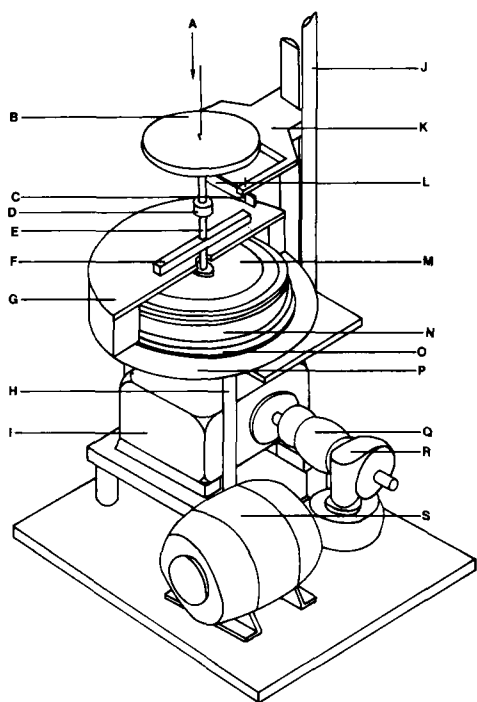


Fig. 1. Modified annular shear cell. A = counterpoise (via pulley). B = weight platform. C = restraining pins. D = Tufnel block. E = vertical steel rod. F = steel bar (used in calibrating strain gauges). G = cooling/heating chamber. H = support for cooling/heating chamber (with one half removed). I = gear box. J = counterpoise pulley support. K = restraining bracket. L = strain bar. M = lid of cell. N = base of cell. O = turntable. P = platform for cooling/heating chamber. Q = coupling. R = reduction gear. S = constant speed motor.

denoted δ' . Another parameter of a powder which can be derived from this plot is the cohesion (C) which is a measure of the attractive forces between particles and depends upon the packing fraction achieved during consolidation.

Fig. 2b shows the corresponding modified yield loci for the lactose powder coated with 0.5×10^{-5} mol g⁻¹ of light liquid paraffin at 20 °C which were obtained by plotting the shear stress at failure against the compound normal stress ($\sigma_N + T$), Nm⁻². T is the tensile strength of the powder at the same packing fraction obtained separately using a tensile tester (Irono & Pilpel 1982). The line joining the end points of the modified loci to the origin is the internal friction yield locus and its slope, Δ , is the angle of internal friction of the material for sustained yield. Δ was found to be independent of the packing fraction.

The technique employed for estimating values of

cohesion was to convert the powder yield loci equation (Ashton et al 1965):

$$(\tau/C)^n = \frac{\sigma_N + T}{T} \quad (2)$$

where τ is the shear stress at failure, n is the shear index, C is the cohesion, σ_N is the normal stress and T is the tensile strength, into its logarithmic form:

$$\text{Log } \tau = K + \frac{1}{n} \text{log} (\sigma_N + T)$$

where $K = \text{log} (C/T^{1/n})$

and plot the logarithm of shear stress at failure versus the logarithm of the compound normal stress. Typical yield loci for lactose powder coated with 0.5×10^{-5} mol g⁻¹ of light liquid paraffin at 20 °C, plotted in this manner are shown in Fig. 3. The best fitting straight line through the points was calculated by regression analysis and the value of cohesion, C, was calculated from the intercept K.

The values of cohesion for all the samples at all the

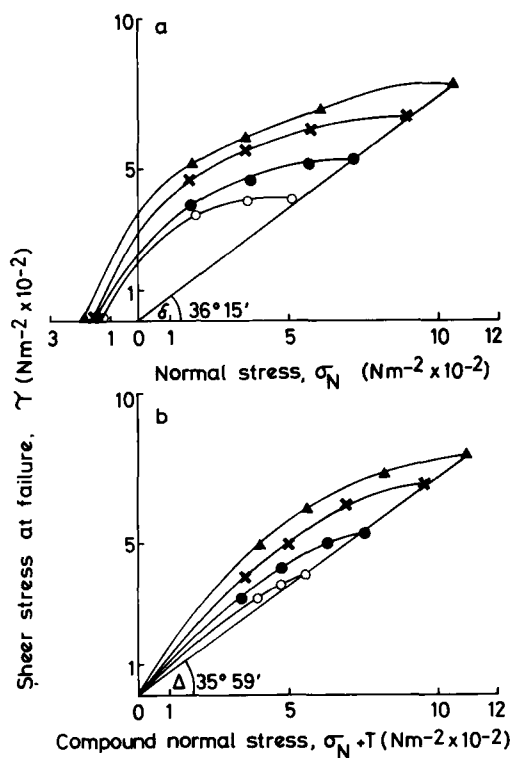


Fig. 2. (a) Yield loci for lactose powder coated with 0.5×10^{-5} mol g⁻¹ of light liquid paraffin at 20 °C. P_f values: \blacktriangle 0.346, \times 0.282, \bullet 0.279, \circ 0.261. (b) Yield loci plotted on axes of compound normal stress and shear stress at failure for lactose powder coated with 0.5×10^{-5} mol g⁻¹ of light liquid paraffin at 20 °C. P_f values: \blacktriangle 0.346, \times 0.282, \bullet 0.279, \circ 0.261.

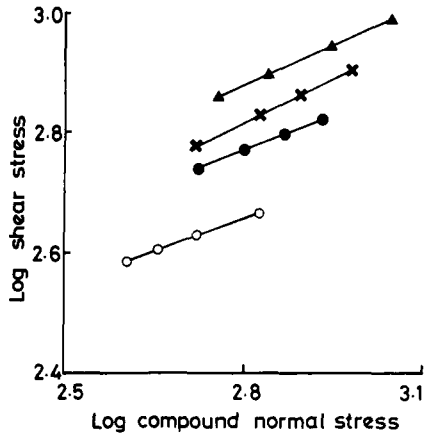


FIG. 3. Log of shear stress at failure versus log of compound normal stress for lactose powder coated with 0.5×10^{-5} mol g^{-1} of light liquid paraffin at 20 °C. P_f values: ▲ 0.346, X 0.282, ● 0.279, ○ 0.261.

temperatures investigated (corresponding to particular viscosities of the coating films) were reasonably found to fit the general equation:

$$\text{Log } C = A_1 P_f + B_1 \quad (3)$$

with a correlation coefficient >0.95 . A_1 and B_1 were constants which depended on the nature and amount of the paraffin used and on its viscosity at the temperature employed (all the samples had been subjected to the same consolidation pressures during the experiments in order to eliminate the effects of

pressure on viscosity). P_f is the packing fraction achieved during consolidation which in most cases ranged between 0.25 and 0.45. Equivalent consolidating conditions at different temperatures produced beds of different packing fraction. But it is essential to compare the values of cohesion and tensile strength at equivalent packing fractions hence the values of cohesion at different temperatures at a fixed P_f 0.41 (the same as previously used when considering tensile strengths (Irono & Pilpel 1982)) were determined. Fig. 4 (a, b) is representative of how the cohesion at this fixed P_f of 0.41 varied with paraffin content at 35 and 50 °C. (The corresponding viscosities of the paraffin coatings at these temperatures (see Irono & Pilpel 1982) are given in the legends to the graphs.) The curve for yellow soft paraffin was omitted to avoid overlapping with white soft paraffin. Fig. 5 (a, b) shows representative plots of how the change in angle of internal friction from no paraffin coating to the series of paraffin coatings used in the experiments varied with the paraffin content at 35 and 50 °C.

DISCUSSION

It is evident from Figs 2a and 3 that the samples investigated satisfactorily obey the equation 2 (Ashton et al 1965).

This equation makes use of the compound normal stress ($\sigma_N + T$) (as shown in Fig. 2b) instead of σ_N , for deriving information about the failure properties

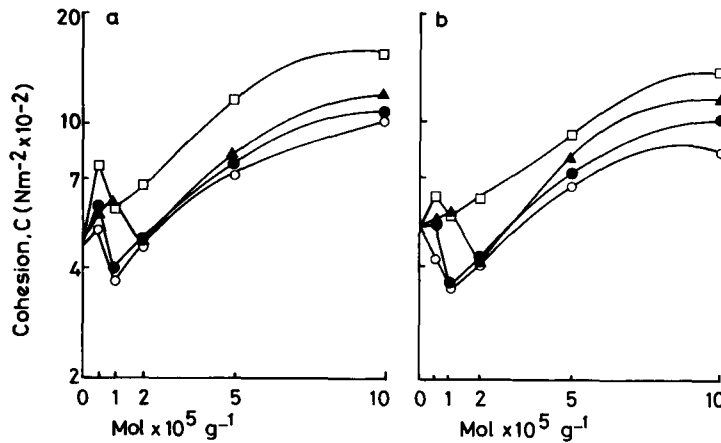


FIG. 4. Cohesion (Nm^{-2}) at P_f 0.41 versus concentration of paraffin at (a) 35 °C (b) 50 °C.

Key	Assumed viscosity of coating (cP)	
	(a)	(b)
● light liquid paraffin	24	17
○ liquid paraffin	27	19
▲ white soft paraffin	53	36
□ paraffin wax	66	44

of a powder. The advantage of this modified yield locus is that it allows for the tensile strength of the powder, as a component of the total normal stress during shearing. It is only at the end point of the modified yield locus that all the work being done by the shearing force is being spent in overcoming the forces of internal friction within the powder (Williams & Birks 1967). At all other points work is being done against both internal and external forces due to changes in the volume of the powder bed. The angle made by the line passing through the origin, on which the end points of the modified yield loci lie, and the abscissa represents the angle of internal friction, Δ . It is defined as a measure of the forces of internal friction between the particles when the bed of powder fails in shear without a change in volume. Williams & Birks (1967) suggested that this provides a better parameter for assessing the dynamic frictional properties. In the present work, the change in angle of internal friction from its value when there was no paraffin coating on the lactose to its value when a particular amount of coating had been applied has been plotted in Figs 5a, b.

It can be seen that the patterns of the curves in Figs 4 and 5 are similar to the previously plotted graphs of tensile strengths (Irons & Pilpel 1982). This is as expected since coating modifies the forces in the powder bed and hence the tensile strength, cohesion and angle of internal friction of the powder. Like the tensile strength, C and Δ are determined by the magnitude and range of the forces that act between neighbouring particles in the bed. The initial increase in cohesion and change in angle of internal friction may be explained by the formation of a discontinuous film of paraffin on the lactose particles which becomes deformed when the particles are subjected to external forces greater than their yield pressure thus increasing the net 'cohesive' forces between the particles. But as more paraffin is added, the surface asperities of the lactose particles become continuously coated with the paraffin film which reduces the coefficient of friction between the particles under the influence of shearing forces, making it easier for them to slide past each other (Bowden & Tabor 1954, 1964). Less work is being done against both the internal and external frictional

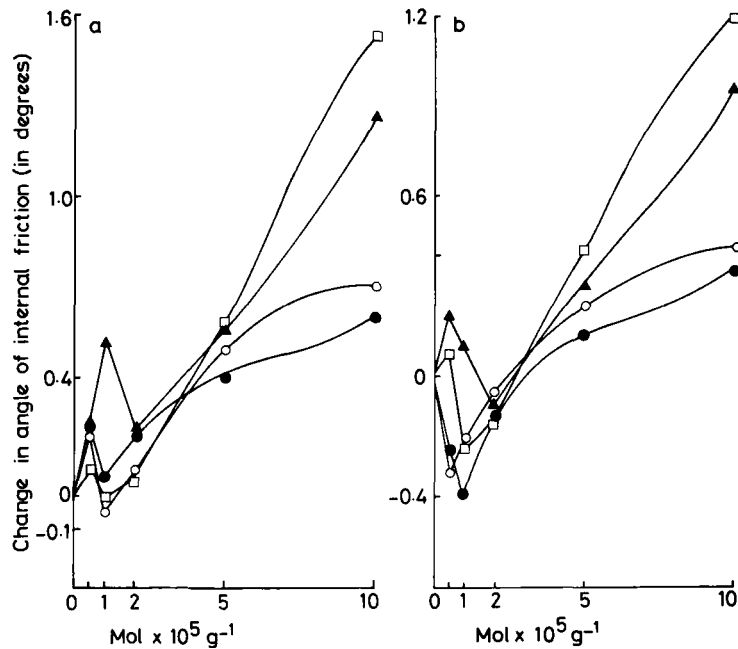


FIG. 5. Change in angle of internal friction versus concentration of paraffin at (a) 35 °C (b) 50 °C.

	Assumed viscosity of coating $c(P)$.	
	(a)	(b)
● light liquid paraffin	24	17
○ liquid paraffin	27	19
▲ white soft paraffin	53	36
□ paraffin wax	66	44

forces of the powder bed. The concentration range, $0.5-2.0 \times 10^{-5}$ mole of paraffin per g of lactose, that produced the minima in cohesion and angle of internal friction has been calculated to correspond to layers between 1 and 5 molecules thick (see Irono & Pilpel 1982).

Addition of more paraffin leads to the formation of thicker coatings with liquid or semi liquid paraffin forming bridges between the lactose particles. The true area of contact between the particles and the 'cohesive' forces between them are greatly increased and more work has to be done by the shearing force in overcoming friction within the powder. This produced the rises observed in cohesion and Δ observed after the minima.

Again the overall increase in cohesion and change in angle of internal friction observed in Figs 4 and 5 (and in the graphs in Irono & Pilpel 1982) as the melting/solidifying point of the coatings was increased may be explained by the fact that when pressure is applied in the shear cell, particles of powder in the plane of shear are brought into contact through the outermost asperities (coated in this case)

on their surfaces. Under the combined influence of pressure and shearing the asperities become deformed and those in the solid state may finally melt. This has been confirmed experimentally for a variety of materials (Jayasinghe et al 1969; Jayasinghe & Pilpel 1970) and the amount of actual melting that occurs becomes appreciable when the ambient temperature has been raised to within about 20 °C of the melting point of the coating material (Pilpel & Britten 1979). Consequently the areas of true contact between the particles increase with the thickness and viscosity of the melted coating producing the increases observed in cohesion and Δ as the viscosity of the coating material is increased.

To investigate if the yield locus parameters of the coated powder show the same correlation observed between the tensile strengths of the powder and the thickness and viscosity of the coating layer, Tables 1 (a, b) and 2(a, b) were drawn up in a similar manner to that described previously. It is seen that the values of cohesion and values of angle of internal friction, Δ of the powders seen in the vertical columns of Tables 1 (a, b) and 2(a, b) respectively were very similar

Table 1. Cohesion at fixed number of layers and fixed viscosity of coating.

(a) Paraffin coatings that are liquid at room temperature

Viscosity (cP)	20			30			60		
	Temp °C	Cohesion (Nm ⁻²)		Temp °C	Cohesion (Nm ⁻²)		Temp °C	Cohesion (Nm ⁻²)	
		for 3 layer coating	for 11 layer coating		for 3 layer coating	for 11 layer coating		for 3 layer coating	for 11 layer coating
Paraffins									
Light liquid	43	430	754	23	571	896	-6	693	997
Liquid	50	429	754	30	570	896	0	690	996

(b) Paraffin coatings that are solid at room temperature

Viscosity (cP)	45			60			80		
	Temp °C	Cohesion (Nm ⁻²)		Temp °C	Cohesion (Nm ⁻²)		Temp °C	Cohesion (Nm ⁻²)	
		for 3 layer coating	for 11 layer coating		for 3 layer coating	for 11 layer coating		for 3 layer coating	for 11 layer coating
Paraffins									
Yellow soft	39	552	865	27	630	897	16	710	1010
White soft	42	552	865	31	630	898	20	712	1012
Paraffin wax	48	550	864	38	632	899	29	712	1010

Table 2. Angle of internal friction, Δ , (in degrees) at fixed number of layers and fixed viscosity of coating.

(a) Paraffin coatings that are liquid at room temperature.

Viscosity (cP)	20			30			60		
	Temp °C	Δ (degrees)		Temp °C	Δ (degrees)		Temp °C	Δ (degrees)	
		for 3 layer coating	for 11 layer coating		for 3 layer coating	for 11 layer coating		for 3 layer coating	for 11 layer coating
Paraffins									
Light liquid	43	35.51	35.96	23	35.99	36.04	-6	36.05	36.20
Liquid	50	35.50	35.96	30	35.98	36.04	0	36.05	36.21

(b) Paraffin coatings that are solid at room temperature

Viscosity (cP)	45			60			80		
	Temp °C	Δ (degrees)		Temp °C	Δ (degrees)		Temp °C	Δ (degrees)	
		for 3 layer coating	for 11 layer coating		for 3 layer coating	for 11 layer coating		for 3 layer coating	for 11 layer coating
Paraffins									
Yellow soft	39	35.61	36.00	27	36.02	36.11	16	36.15	36.34
White soft	42	35.61	36.00	31	36.02	36.10	20	36.13	36.34
Paraffin wax	48	35.60	36.00	38	36.02	36.10	29	36.13	36.34

when the paraffin coatings had the same thickness and were of the same assumed viscosity due to temperature.

Conclusions

The failure properties of lactose powder coated with increasing amounts of a series of paraffins depend on the concentration, thickness and viscosity of the coating material.

The angle of internal friction, Δ , does not vary with the packing fraction indicating 'simple' behaviour for lactose powder coated with up to 10^{-4} mole of paraffin g^{-1} of powder.

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REFERENCES

- Ashton, M. D., Cheng, D. C. H., Valentin, F. H. H. (1965) *Rheol. Acta* 4: 206-218
- Bowden, F. P., Tabor, D. (1954, 1964) *Friction and Lubrication of Solids*; Parts 1 and 2, Clarendon Press, Oxford
- Carr, J. F., Walker, D. M. (1968) *Powd. Technol.* 1: 369-373
- Irono, C. I., Pilpel, N. (1982) in the press
- Jayasinghe, S. S., Pilpel, N. (1970) *J. Inst. Fuel* 43: 51-55
- Jayasinghe, S. S., Pilpel, N., Harwood, C. F. (1969) *Mater. Sci. Eng.* 5: 287-294
- Kočova, S., Pilpel, N. (1972) *Powd. Technol.* 5: 329-343
- Malamataris, S., Pilpel, N. (1980) *Ibid.* 26: 205-211
- Pilpel, N., Britten, J. R. (1979) *Ibid.* 22: 33-44
- Pilpel, N., Hephher, M. N. (1977) *Mater. Sci. Eng.* 27: 89-95
- Williams, J. C., Birks, A. H. (1967) *Powd. Technol.* 1: 199-206
- York, P., Pilpel, N. (1972) *Mater. Sci. Eng.* 9: 281-291
- York, P., Pilpel, N. (1973) *Ibid.* 12: 295-304