

A Consideration of the Range and Strength of Interparticle Forces in Coated Lactose Powder

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Abstract—A study has been made of the range and strength of interparticle forces in lactose powder coated with increasing concentrations of paraffins at different temperatures. Marked changes occurred in these forces as the paraffin content was altered and the temperature increased from -10° to $+50^{\circ}\text{C}$. The strength and range of interparticle forces depended on the mechanical properties of the coating materials and the surface free energy of the coated powders.

The tensile strengths of dry powder beds are expressed in terms of the density and particle size distribution by assuming that the particles touch only at the tips of the asperities on their surfaces (Cheng 1968). The tensile strength, T , is given as:

$$T = h(t) A \quad (1)$$

Where A = total overall area of contact of particle pairs in the plane of the split surface, and $h(t)$ = interparticle force per unit area of contact, and t is the average interparticle separation expressed by,

$$t = t_0 - \frac{\bar{d}}{3} \left(\frac{P_f}{P_{fo}} - 1 \right) \quad (2)$$

$\bar{d}$ is the mean diameter of the particles, P_f the packing fraction, P_{fo} the packing fraction when $T=0$ and t_0 the effective range of interparticle forces.

t_0 depends on the size, surface free energy (Johnson et al 1971), hardness and elasticity of the particles and $h(t)$ is characterized by one energy or 'strength' parameter E_0 , one length or 'range' parameter t_0 and by other dimensionless parameters, that describe the simplest functional form of the associated potential function (Hakala 1967).

One method of altering the values of E_0 and t_0 without significantly altering the surface geometry, particle size and shape is to coat the particles with a relatively thin film of another material. In the present investigation which continues work on the properties of lactose powder (Pilpel & Hephher 1977; Malamataris & Pilpel 1980, 1982, Irono & Pilpel 1982 a,b; Sakr & Pilpel 1982, we have compared the values of E_0 and t_0 of samples at different temperatures before and after coating the particles with various paraffins. We hoped that it might be possible to establish some connection between the mechanical properties of the coatings, which depend on temperature, and the strength and range of the interparticle forces.

Materials and Methods

Details have been given elsewhere (Irono & Pilpel 1982 a,b; Igwilo & Pilpel 1983) of the lactose and of the methods used to coat the particles with different paraffins and then measure

the tensile strengths of the powder beds at different temperatures. The mechanical properties of the coating materials at these temperatures, measured on cast films using a micro-indentation apparatus (Pilpel & Britten 1979), were expressed in terms of their Brinell hardness (BH) and Young's modulus of elasticity, using the following equations (Ridgway et al 1970).

$$BH = \frac{W}{\frac{\pi D}{2} \left[D - \sqrt{D^2 - d^2} \right]} \quad (3)$$

$$\text{Modulus of elasticity} = \frac{0.268W}{\Delta h e \sqrt{h}} \quad (4)$$

where $\Delta h e$ is the elastic recovery after removal of load, W ; h , the depth to which the indenter penetrates under load, D the diameter of the spherical indenter and d the diameter of the impression made by the indenter on the film surface which is equal to $\sqrt{4hD}$. The results of the measurements are given in Table 1.

Surface free energy determination

The surface free energy of the coated powders, γ_s was calculated from the measurements of contact angles exhibited by water and methylene iodide (Wu & Brzozowski 1971) on the solid surfaces. Details of the contact angle measurements have been given elsewhere (Igwilo & Pilpel 1983). The basis for using the contact angle to determine the

Table 1. Variation of Brinell hardness and modulus of elasticity of paraffins with temperature.

Paraffins and Solidifying Melting Points ($^{\circ}\text{C}$)	Temperature ($^{\circ}\text{C}$)	Brinell Hardness BH ($\text{Nm}^{-2} \times 10^{-5}$)	Modulus of Elasticity ($\text{Nm}^{-2} \times 10^{-6}$)
1. Liquid paraffin (4)	-20 -10 -1	9.934 8.652 7.148	— — —
2. White soft paraffin (48)	-20 -10 0 10	10.036 8.922 8.609 8.122	5.252 4.126 2.911 —
3. Paraffin wax (52)	-20 -10 0 10	12.706 10.496 9.601 8.508	7.341 4.867 3.943 3.213

Table 2. The densities and surface free energy values for water and methylene iodide at 20°C.

Liquid	Density (g cm ⁻³)	γ_L^d (mJm ⁻²)	γ_L^p (mJm ⁻²)	γ_L (mJm ⁻²)
Water	1.000	23.3	48.9	72.2
Water saturated with lactose)	1.032	21.5	49.0	70.5
Methylene iodide	3.325	52.1	0	52.1

surface free energies of solids rests on the relationship between solid surface free energy per square centimetre, γ_s , liquid surface free energy per square centimetre, γ_L and their interfacial free energy per square centimetre, γ_{LS} as shown in the following equations (Zisman 1964; Adamson 1967; Wu 1971),

$$\gamma_s = \gamma_L \cos \theta + \gamma_{LS} \quad (5)$$

$$\gamma_{LS} = \gamma_L + \gamma_s - \frac{4\gamma_L^d \gamma_s^d}{\gamma_L^d + \gamma_s^d} - \frac{4\gamma_L^p \gamma_s^p}{\gamma_L^p + \gamma_s^p} \quad (6)$$

where γ_L^d and γ_L^p are the non-polar and polar components of the surface free energy of the liquid and their sum ($\gamma_L^d + \gamma_L^p$) is γ_L (The method for determining these two components has been described (Wu 1971) and their values are given in Table 2). γ_s^d and γ_s^p are the non-polar (dispersion) and polar components of the total surface free energy of the powder, ($\gamma_s^d + \gamma_s^p$) = γ_s , and the values are given in Table 3.

Results and Discussion

The values of the particle size parameters, expressed as effective mean diameter, $\bar{d}$, effective surface per particle (based on the area of the smaller particle in a pair) $\bar{s}$, and effective mean volume per particle, $\bar{v}$, were calculated using equations derived by Cheng (1968) and are 14 μm , 925 μm^2 and 16047 μm^3 , respectively.

The effects of paraffin content on the range and strength of interparticle forces are shown typically in Fig. 1 (a,b) and Fig. 2 (a,b), respectively, at two of the temperatures investigated. It is seen that both t_0 and E_0 initially increase, then decrease, and then increase again as the amount of coating material is increased up to about $5 \times 10^{-5} \text{ mol g}^{-1}$. The changes in the parameters depend on the physical properties

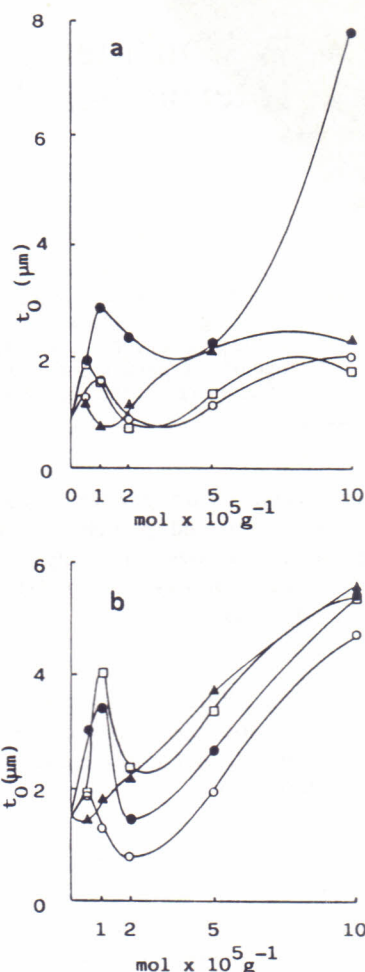


FIG. 1. (a,b) ' t_0 ' versus paraffin concentration at (a) -10°C (b) $+50^\circ\text{C}$. ● Light Liquid paraffin, ○ Liquid paraffin, ▲ White soft paraffin, □ Paraffin wax.

and melting points of the coatings employed and clearly in some of the experiments the coatings were in a liquid state and in others in a solid state. However, the state of the coatings seems to have less effect on the values of t_0 which are all between 1 and 5 μm than on the values of E_0 which increase about 10 fold as the temperature is raised from -10° to $+50^\circ\text{C}$.

E_0 , the strength of the interparticle forces, arises due to

Table 3. Surface free energies of coated lactose powder at 20°C.

Lactose + coating	Coating content mol g ⁻¹ × 10 ⁶	Non polar component of surface free energy γ_s^d (mJm ⁻²)	Polar component of surface free energy γ_s^p (mJm ⁻²)	Total surface free energy γ_s (mJm ⁻²)
Liquid paraffin	10.0	43.2	27.3	70.5
	20.0	46.1	24.6	70.8
	50.0	41.8	24.7	66.5
	100.0	41.6	24.2	65.8
White soft paraffin	10.0	38.9	21.6	60.5
	20.0	37.9	20.4	58.3
	50.0	34.9	19.8	54.7
	100.0	34.0	19.0	53.0
Paraffin wax	10.0	32.5	14.9	47.4
	20.0	29.5	13.5	43.0
	50.0	26.8	12.9	39.7
	100.0	23.7	12.5	36.2

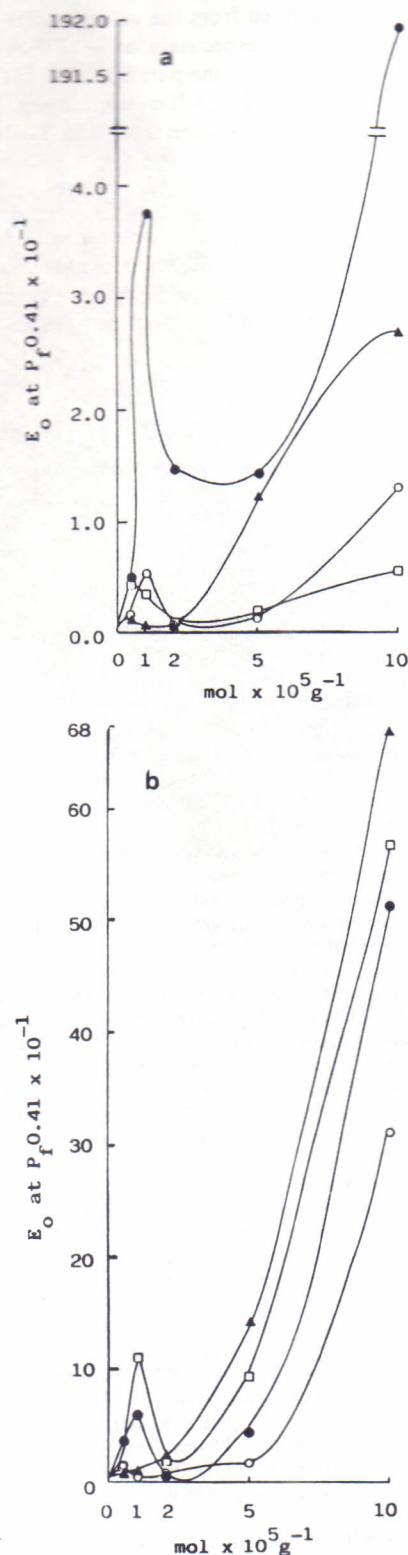


FIG. 2. (a,b) E_0 at $P_f 0.41$ versus paraffin concentration at (a) -10°C (b) $+50^\circ\text{C}$ (key as in Fig. 1)

frictional and Van der Waals' forces of attraction between the particles. The initial increase observed in E_0 as paraffin is added may be due to the adsorption of a discontinuous layer of paraffin on the surface of the powder particles leading to bond formation between paraffin molecules and lactose

particles, and bonds between paraffin-paraffin molecules, in addition to the lactose—lactose bonds already existing in the powder bed. This effect is similar to that produced by addition of very small amounts of moisture to powders (Cheng 1970).

Addition of more paraffin to the powders produced a decrease in E_0 probably due to the masking of Van der Waals' forces and reduction in frictional forces between the particles by the coatings. However, once a monolayer of the coating material had formed (Irono & Pilpel 1982a), further addition of paraffins led to the development of enhanced mechanical binding forces between the particles and additional attractive surface tension forces due to the presence of liquid films. This has been noted previously by Sakr & Pilpel (1982), when increasing amounts of a series of non-ionic surfactants was added to lactose powder.

Considering the special case of two rigid spherical particles in contact, denoted 1 and 2, Derjaguin (1961) showed that the adhesive strength, F , i.e. the forces required to separate them, is given as

$$F = \frac{3\pi R_1 R_2}{2(R_1 + R_2)} \sqrt{Y_{S1} \cdot Y_{S2}} \text{ J m}^{-1} \quad (7)$$

where R is the radius and Y_S is the surface free energy. But if the surfaces become deformed the adhesive force increases (Krupp & Sperling 1967) to

$$F = \frac{3\pi R_1 R_2}{2(R_1 + R_2)} \sqrt{Y_{S1} \cdot Y_{S2}} \frac{(1 + 3\pi X)}{Y} \cdot \sqrt{Y_{S1} \cdot Y_{S2}} \text{ J m}^{-1} \quad (8)$$

where Y is the yield pressure of the deformed layer and X is a numerical term which depends on its area, shape and thickness. Particles become deformed by being subjected to external forces greater than their yield pressure and since adsorption of liquid often causes the surfaces of particles to soften, the adhesive force increases. With particles whose surfaces do not soften, the liquid is absorbed on them in a layer several molecules thick, and this also has the effect of increasing the area of contact and the adhesive force between them. These effects may become pronounced at elevated temperatures and it seems probable that these mechanisms could also be contributing to the marked increases observed in E_0 as the paraffin content increased up to $10^{-4} \text{ mol g}^{-1}$ powder.

The increase observed in E_0 between -10° and $+50^\circ\text{C}$ may be due primarily to the state of the coatings. At -10°C the coatings are generally in a solid state (this is seen from the values of the mean solidifying/melting point of all the paraffins given by Irono & Pilpel 1982a) and the pressure applied during the tensile tests does not produce appreciable melting of the coatings on the surfaces of the particles. However, at $+50^\circ\text{C}$, under the combined influence of temperature and pressure, the paraffin coatings melt (this is similar to melting of asperities reported by York & Pilpel 1972 and Pilpel & Britten 1979) with subsequent formation of molten bonds between the neighbouring particles, the strength of which increased with increase in the assumed viscosities of the coatings (Irono & Pilpel 1982b). These deduced viscosities in turn depend on the physico-mechanical properties of the paraffin coatings. This would agree with the observation of Bowden & Bastow (1931) that the

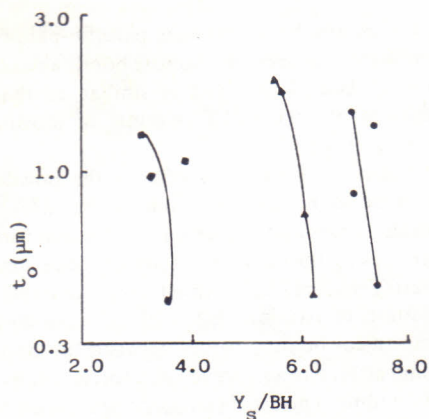


FIG. 3. t_o versus Y_s/BH for coated lactose at -20°C (coating content: (1.0, 2.0, 5.0 and 10.0) $10^{-3} \text{ mol g}^{-1}$ of paraffin coatings). ● Liquid paraffin, ▲ White soft paraffin, ■ Paraffin wax.

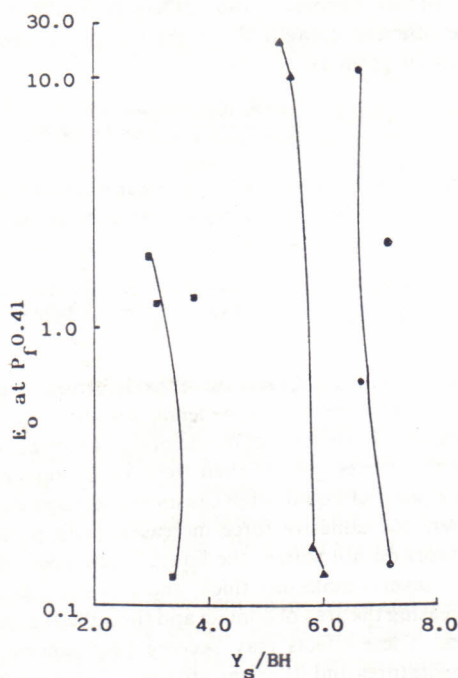


FIG. 4. E_o at $P_f 0.41$ versus Y_s/BH for coated lactose at -20°C (coating content: (1.0, 2.0, 5.0 and 10.0) $10^{-3} \text{ mol g}^{-1}$ of paraffin coatings) (key as in Fig. 3).

adhesion between surfaces is increased by interposing a viscous liquid between them.

From the above discussion, it seems logical to expect some connection between the range and magnitude of the interparticle forces and the mechanical properties of the coating material*. Considering equations 7 and 8 and assuming that the yield pressure Y of the coatings is a function of the reciprocal of Brinell hardness (Malamataris & Pilpel 1982), a correlation might be expected between t_o , E_o and Y_s/BH since the sizes of the particles have not been noticeably altered by the presence of the relatively thin film of coatings.

Figs 3, and 4 show the typical plots of t_o vs Y_s/BH and E_o

vs Y_s/BH (derived from the values of BH and Y_s given in Tables 1 and 3), respectively, at -20°C . (It was possible to obtain BH values of the paraffins only at the low temperatures, 0° , -10° , -20°C). It is seen that an approximate linear relationship exists between t_o , E_o and Y_s/BH .

Conclusion

The paraffin coatings alter the range and strength of the interparticle forces that act between the surfaces of the coated and uncoated particles. A correlation has been established between t_o , E_o and the mechanical properties of the paraffin coatings.

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* It is seen from Fig. 1 (a,b) and Fig. 2 (a,b) that different values of t_o and E_o are obtained using different types (but the same amount) of the paraffins for coating the powders.