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Powder flow studies IV. Tensile strength and orifice flow rate relationships of binary mixtures

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Summary

The tensile strength of consolidated powder beds of spray-dried lactose and binary mixtures of lactose containing different concentrations of glidants and/or lubricants was determined. The orifice flow rate of these powders was measured by choosing an appropriate orifice diameter. Powder mixtures containing up to 1% glidant resulted in general in a decrease in the tensile strength and an increase in the flow rate. The flow rate of powder mixtures containing simple glidants such as corn starch and microcrystalline cellulose at different concentrations was linearly related to the tensile strength. More complicated second- and third-degree relationships were obtained when tensile strength and flow rate of binary mixtures of lactose with different concentrations of magnesium stearate, stearic acid, colloidal silicone dioxide and talc were plotted. The major limitation of relating tensile strength to orifice flow rate is that the powder system should be sufficiently cohesive to produce measurable tensile strength of the consolidated powder, yet sufficiently free-flowing to produce a gravity flow under an unknown and changing consolidation state.

Introduction

The knowledge of the properties of powders consisting of relatively large cohesion-free particles is abundant. Most empirical (Beverloo et al., 1961; Rose and Tanaka, 1959) and semi-theoretical (Harmens, 1963; Brown, 1961) flow rate equations predict that the flow rate of free-flowing particles through an orifice by gravity increases slowly as the particle size decreases.

The rheological behavior of cohesive particles, the so-called sticky powders, is not very well understood. It is known that the orifice flow rate decreases as the particle size decreases due to the changing relative magnitude of dispersion forces and gravitational forces per particle as particle size decreases. This cohesive tendency coupled with frictional resistance (between powder particles or between hopper wall and powder particles) tends to oppose gravitational downward flow tendency. There is also very little known about the fundamental problem of the relationship between the measurable properties of individual particles such as size, shape, surface texture, surface fine structure, specific surface area and the collective physical properties of the powder mass. Some of these are used in understanding and predicting flowability of non-free-flowing powders and include the repose angle of powders (Train, 1958; Riley et al., 1978; Gold et al., 1966), the minimum orifice diameter for discharge and the rate of discharge through orifices (Gold et al., 1968; Jones and Pilpel, 1966; Harwood and Pilpel, 1969), tablet and capsule weight variation (Ho et al., 1977; Augsburger and Shangraw, 1966; Gold et al., 1968; Chowhan and Chow, 1980; Bagster and Crooks, 1978), consolidation ratio (Chowhan and Chow, 1981), tensile strength (Kocova and Pilpel, 1973, 1974; Verthalis and Pilpel, 1976, 1977; Nash et al., 1965; York, 1975; Crooks et al., 1977; Chowhan and Chow, 1981), and shear strength (Ho et al., 1977; York, 1975; Kocova and Pilpel, 1974; Crook et al., 1977; Pilpel and Walton, 1974; Marshal and Sixsmith, 1976; Kurihara and Ichikawa, 1973).

In the orifice flow of powders, both extrinsic and intrinsic factors play their role. The extrinsic factors relate to the hopper and bin geometry, such as the hopper angle, the hopper orifice diameter, the hopper shape, etc. The tensile and shear testing of powders were originally focused on extrinsic factors and were applied to the hopper and bin design (Jenike, 1964; Cheng et al., 1963). In recent years, the intrinsic factors relating interparticular forces of cohesive powders have been studied by tensile and shear cell measurements. These results were related to the flow of powders through some kind of a hopper device.

The increasing demand of handling non-free-flowing fine powders has necessitated improving the flowability of these bulk particulate solids. Because of the non-free-flowing nature of these powders, the evaluations of the effects of added excipients, generally classified as glidants, is carried out with a hopper by increasing the hopper orifice diameter to facilitate flow. Several other collective physical properties of the powder mass have been used to study and predict the effect of glidants on flowability of non-free-flowing powders. The angle of repose (Gold et al., 1966; Jones and Pilpel, 1966), compressibility (Peleg and Mannheim, 1973), bulk density (Jones, 1968; Nash et al., 1965), flow factor (York, 1975), tensile strength (Varthalis and Pilpel, 1977; Eaves and Jones, 1972) and bulk tensile strength (Nash et al., 1965) have been used.

This paper reports the results of the tensile strength and orifice flow rate of relatively free-flowing spray-dried lactose and binary mixtures containing lactose with different concentrations of glidants and/or lubricants. Powder mixtures containing up to 1% glidant resulted in general in a decrease in the tensile strength and an increase in the flow rate. Powder mixtures containing simple glidants such as corn starch and microcrystalline cellulose resulted in a linear relationship between tensile strength and flow rate. More complicated second- and third-degree relationships were obtained with powder mixtures containing different concentrations of lubricant and/or glidant.

Materials and methods

Materials

The excipients used in this study were spray-dried lactose USP (Foremost, San Francisco, CA), magnesium stearate NF (Mallinckrodt Chemical Works, St. Louis, MO), stearic acid powder USP (Emery Industries, Cincinnati, OH), talc USP (J.T. Baker Chemicals, Phillipsburg, NJ), colloidal silicon dioxide NF (Aerosil 200, Degussa, Teterboro, NJ), microcrystalline cellulose NF (pH 102, FMC, Philadelphia, PA) and corn starch NF (Staley Manufacturing, Decatur, IL).

Mixing

The powders of each binary mixture were mixed by geometric dilution on a piece of glassine paper. The powder mixture was screened through a no. 40-mesh screen to ensure proper mixing and to avoid powder agglomeration due to compaction. The final mixing was done in a polyethylene bag. All powder mixing and tensile strength measurements were carried out at $23 \pm 1°C$ and $40 \pm 5\%$ relative humidity.

Tensile strength

The tensile strength of lactose and binary mixtures was determined by a cohetester (Hosokawa Micromeretics Laboratory, Osaka, Japan). The instrument consists of a main unit, an operational amplifier and an x–y recorder. The main unit included 4 major components; (a) a constant-speed motor for exerting a pulling force through a steel string to break the powder bed; (b) a cylindrical powder cell with one half fixed to the main unit and the other half movable by suspension onto a metal frame; (c) a strain gauge of 200 *g* force for the quantitative measurement of tensile stress required to break the powder bed; and (d) a linear, variable differential transformer for the measurement of powder bed displacement.

The operational amplifier contained a strain amplifier, displacement meter and an operational unit. The latter was designed to indicate only the actual load by subtracting the necessary blank load that varied with displacement.

Blank tests were run to ensure that there was no extra load between the two half-cells before the powder was loaded. An accurately weighed amount of the powder was charged evenly through the cell extension tube, and the compaction weight (0.3 kg) was inserted gently into this tube. After 20 s, an additional 2 kg weight was loaded in 4 parts with 20 s intervals between each loading. The weights and the cell extension tube were unloaded after 5 min. A steady-state condition in the powder bed was thus reached. The pulling string connecting the powder cell through a strain gauge was tightened under suitable tension before the clamp of the two half-cells was released. Then the constant-speed motor switch was turned on to pull the moving half-cell.

During the breaking process, the x–y recorder automatically recorded the tensile force profile over the total displacement of the two half powder beds. The peak value on the vertical axis of the tensile force–displacement curve gave the maximum tensile force of the powder. The area of breakage was normally 5 cm^2, provided the compaction weight descended to the proper position. Otherwise, the area was corrected for the powder bed thickness difference. The tensile strength of the powder bed was obtained by dividing the maximum tensile force with the area of powder bed breakage. Three determinations were made, and the mean and the standard deviation were calculated. Only results obtained from the tensile fracture where the powder bed broke into two halves along the joint of the two half-cells was recorded.

Powder flow

The flow of the powder and powder mixtures was determined by means of conical glass funnels. The upper funnel diameter was 10 cm with an orifice diameter of 1.62 cm. The lower funnel which was 1 mm below the orifice of the upper funnel had a diameter of 6.1 cm and an orifice diameter of 0.935 cm. The orifice of the upper funnel was blocked by means of a flat paper board and the powder or powder mixture loaded in the top funnel. Then the powder was allowed to flow through both funnels by removing the paper board. After 5 s of equilibration, a tared container was placed under the orifice of the lower funnel and powder collected for 9 s and accurately weighed.

Results and discussion

The tensile strength of powders and powder mixtures changes drastically with the consolidation pressure. Plots of the tensile strength versus consolidation pressure were linear (Chowhan and Yang, 1981) and the dimensionless slope of the lines was regarded as material constant, which is a measure of the increase in cohesive force between adjacent particles due to consolidation. The intercept indicated the cohesive forces in an unconsolidated powder bulk. For free-flowing powders, the intercept is zero. In flow rate studies, the consolidation state of the powder bed is changing. The consolidation state in a hopper depends on the size of the hopper and the history of the powder bed. Because of the complexity of the consolidation state of the powder bed in a powder flow situation, no attempt was made to mimic this consolidation state in tensile strength measurements. The tensile strength was measured at a fixed consolidation pressure (117 g/cm^2).

The results of the flow rate of powder mixtures containing different precentages of colloidal silicon dioxide are given in Fig. 1. There was a small increase in the flow rate when the concentration of the glidant in the powder mixture was 0.25%. Above 0.25% glidant concentration, the flow rate of the powder mixture decreased as the concentration of the glidant increased. The tensile strength data of the powder mixture (Fig. 1) shows a minimum at 0.25% colloidal silicon dioxide concentration. It was not possible to measure the tensile strength of powder mixtures containing more than 2% colloidal silicon dioxide. Under the experimental conditions of consolidation, mixtures of lactose containing more than 2% colloidal silicon dioxide were not sufficiently cohesive to form a consolidated powder bed. This is the limitation of the tensile strength determinations. The powders and powder mixtures should be sufficiently cohesive to form a consolidated powder bed, yet free-flowing to produce a gravity flow under an unknown and changing consolidation state.

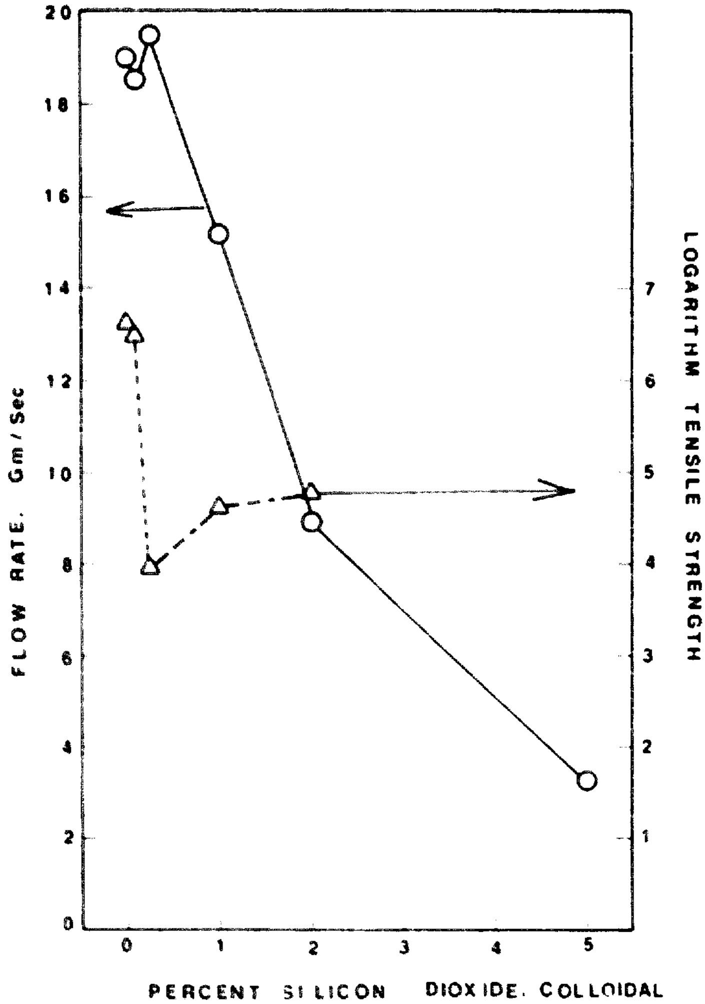

Fig. 1. Plots of the logarithm of tensile strength and flow rate of powder mixtures containing different concentrations of colloidal silicon dioxide.

A large decrease in the tensile strength of powder mixture containing 0.25% colloidal silicon dioxide is reflected in a small increase in the flow rate. Powder mixtures containing higher concentrations of colloidal silicon dioxide show an increase in the tensile strength and a decrease in the flow rate.

Fig. 2 gives the results of the flow rate and logarithm of tensile strength of powder mixtures containing different concentrations of stearic acid. The flow rate of the powder mixture containing 0.5% stearic acid was higher than the flow rate of lactose alone. Above this concentration, the flow rate decreased slowly to approximately the level of lactose. The tensile strength of powder mixtures containing more than 0.1% stearic acid was lower than the tensile strength of lactose showing a minimum at 1% stearic acid concentration in the mixture.

A large decrease in the tensile strength of powder mixtures containing 0.25% and 1.0% magnesium stearate did not result in an improvement in the flow rate (Fig. 3). The flow rate of the mixtures containing 1–5% magnesium stearate decreased as the concentration was increased. Magnesium stearate which could be termed a true

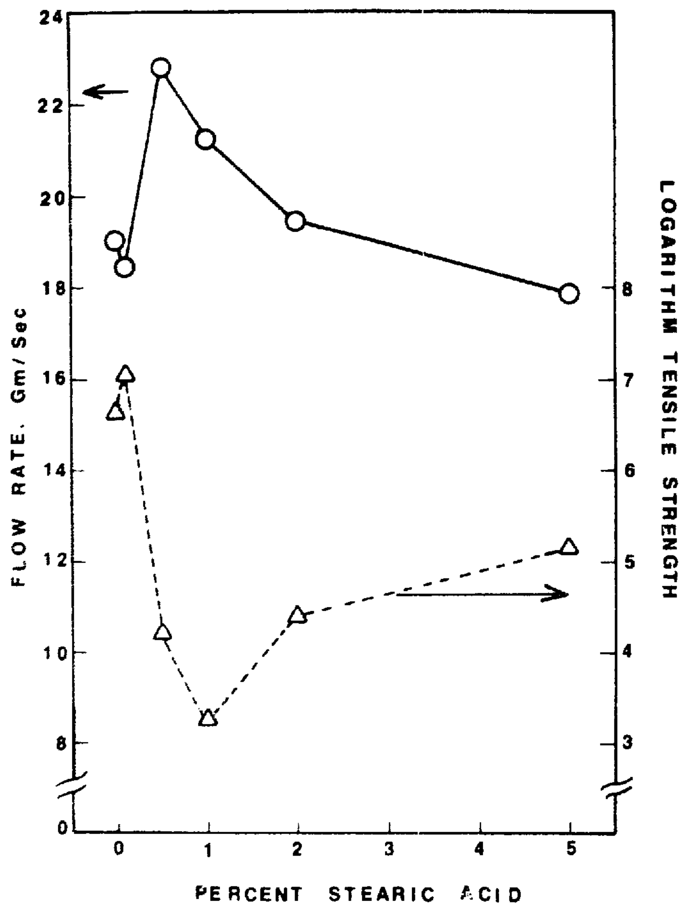

Fig. 2. Plots of the logarithm of tensile strength and flow rate of powder mixtures containing different concentrations of stearic acid.

lubricant did not improve but rather retarded powder flow above 0.25% concentration. This is due to the poor glidant properties of magnesium stearate. The tensile strength of the powder mixture increased as the concentration of the magnesium stearate in the mixture was increased above 1%. The tensile strength of the powder mixture containing 5% magnesium stearate was lower than the tensile strength of lactose.

The results of the flow rate and the logarithm of tensile strength of the powder mixtures containing different percentages of talc are given in Fig. 4. These results indicate an increase in the flow rate and a corresponding decrease in the tensile strength of powder mixtures containing 0.25% talc. Moderately effective lubricant properties and excellent anti-adherent properties of talc account for an increase in flow rate at lower concentrations. Powder mixtures containing higher concentrations of talc showed a decrease in the flow rate without a corresponding increase in the tensile strength.

An increase in the flow rate and a corresponding decrease in the tensile strength of powder mixtures was observed (Fig. 5) when 0.25% and 1% corn starch were mixed with lactose. The powder flow rate decreased and the tensile strength

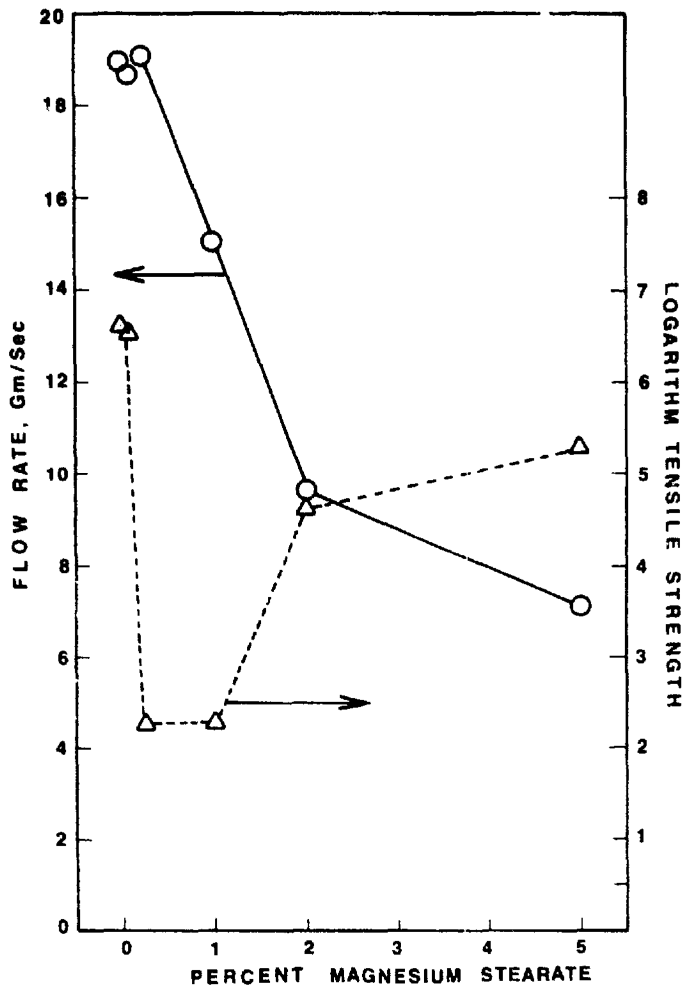

Fig. 3. Plots of the logarithm of tensile strength and flow rate of powder mixtures containing different concentrations of magnesium stearate.

increased when powder mixtures containing more than 1% corn starch were tested.

Fig. ⌄ gives the results of the flow rate and logarithm of tensile strength of powder mixtures containing microcrystalline cellulose. The flow rate increased and the tensile strength decreased as the concentration of microcrystalline cellulose in the powder mixture was increased to 1%. Powder mixtures containing higher concentrations of microcrystalline cellulose showed a decrease in the flow rate and an increase in the tensile strength.

The results given in Figs. 1–6 indicate that, in general, powder mixtures containing lower concentrations of glidants resulted in lower tensile strength and a higher flow rate. When flow rate and tensile strength data were plotted for powder mixtures containing different concentrations of glidants/lubricants, two types of curves were obtained. Fig. 7 shows a simple linear relationship between flow rate and tensile strength of binary powder mixtures containing starch or microcrystalline cellulose at different concentrations. Corn starch and microcrystalline cellulose are generally

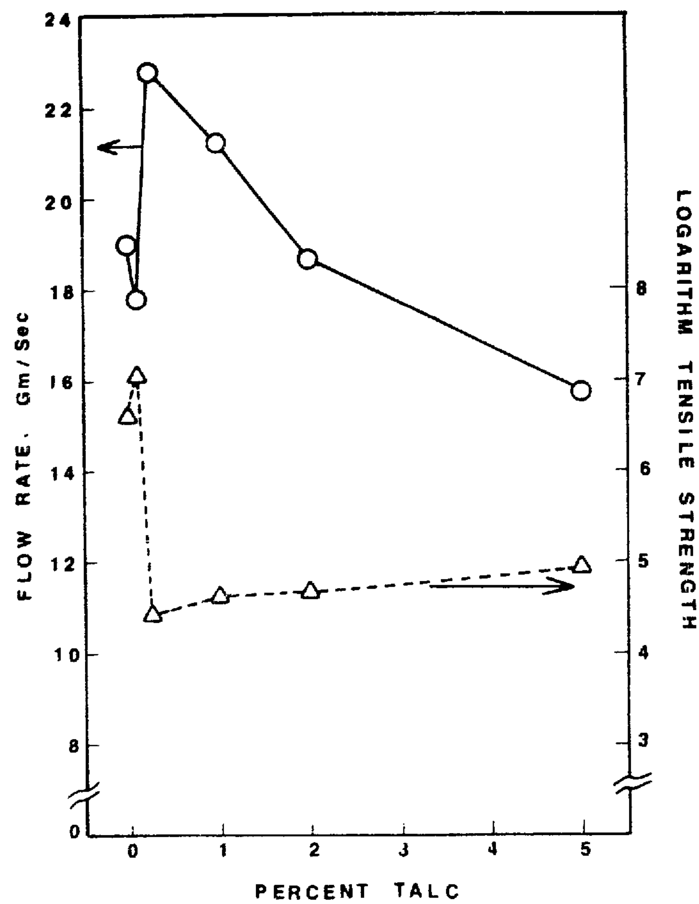

Fig. 4. Plots of the logarithm of tensile strength and flow rate of powder mixtures containing different concentrations of talc.

regarded as simple glidants. Although the mechanism by which glidants work in improving flow rate is not very well understood, it is generally believed that glidants improve powder flow by filling void spaces between particles and by reducing interparticulate cohesive forces. If for a simple glidant, this mechanism was operating, it is perhaps reasonable to expect a linear relationship between flow rate and tensile strength.

More complicated second-degree curves were obtained when tensile strength and flow rate of binary mixtures of lactose with magnesium stearate, colloidal silicon dioxide and talc were plotted (Fig. 8) showing a simple minimum. The plots of the powder mixtures containing stearic acid show a third-degree relationship between tensile strength and flow rate. The second- and the third-degree relationships of these powder mixtures are perhaps due to the combination of glidant, lubricant and antiadherent properties of these excipients. Lubricants reduce friction at the interface by providing a film that should prevent solid–solid contact. The glidants, on the other hand, increase bulk density by filling voids in the host particles and reduce interparticular cohesion. The magnitude of the lubricant and/or glidant action on

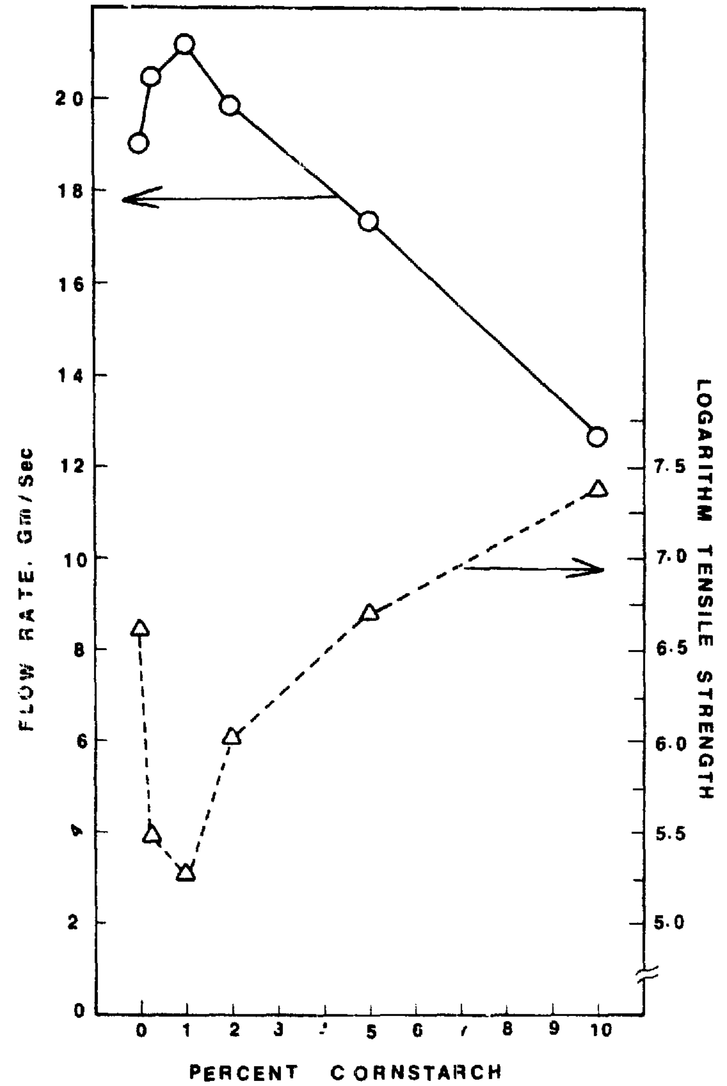

Fig. 5. Plots of the logarithm of the tensile strength and flow rate of powder mixtures containing different concentrations of corn starch.

the host particles would depend on the concentration of the lubricant/glidant used in the binary mixtures and the relative lubrication and glidant properties of a given excipient. In addition to the basic differences in the material properties of the added excipient, as the composition of the mixture is altered, changes in the range and magnitude of interaction forces that operate between particles will occur (Cheng et al., 1968; Kocova and Pilpel, 1973). The forces which could be included in this category are, Van der Waal, ionic, valency, lattice,frictional and capillary.

In a previous study (Chowhan and Yang, 1981), drug–lactose mixtures containing 10% starch and 0.5% magnesium stearate were used to study the relationship between tensile strength and flow rate. The only changes made in the formulations were the concentration of the drug and the concentration of the lactose. The concentration of the glidant and lubricant were fixed. This could explain the earlier

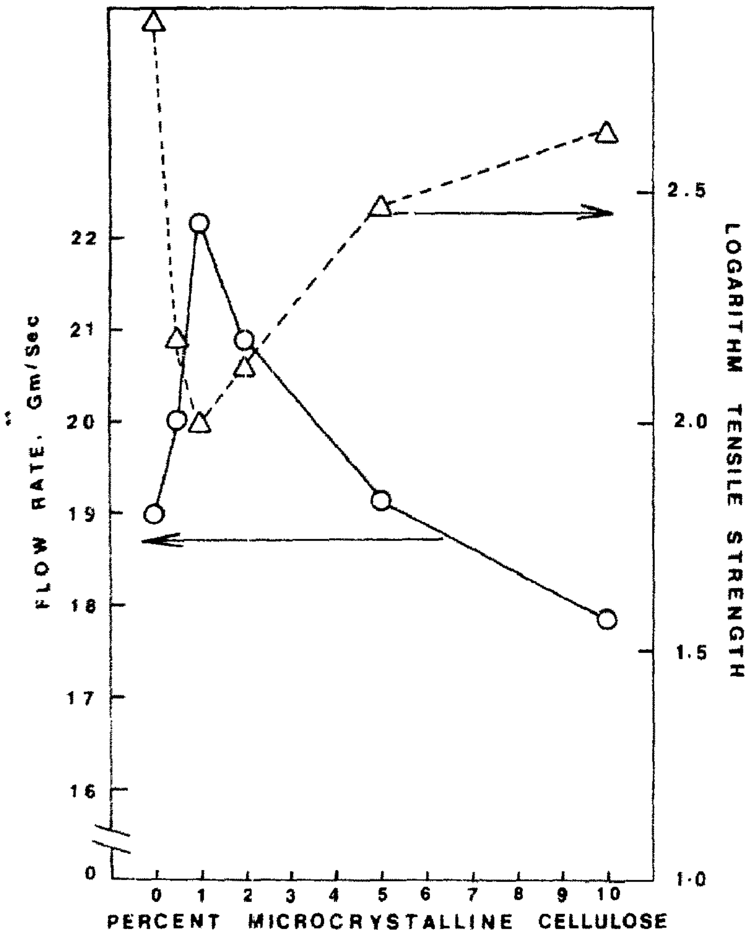

Fig. 6. Plots of the logarithm of tensile strength and flow rate of powder mixtures containing different concentrations of microcrystalline cellulose.

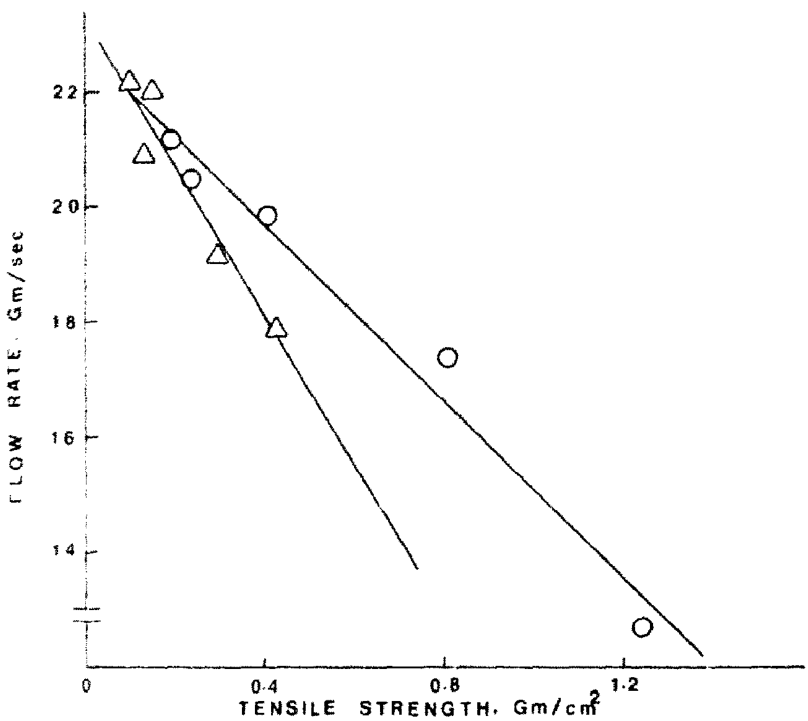

Fig. 7. Flow rate versus tensile strength of powder mixtures. Key: ○, corn starch; △, microcrystalline cellulose.

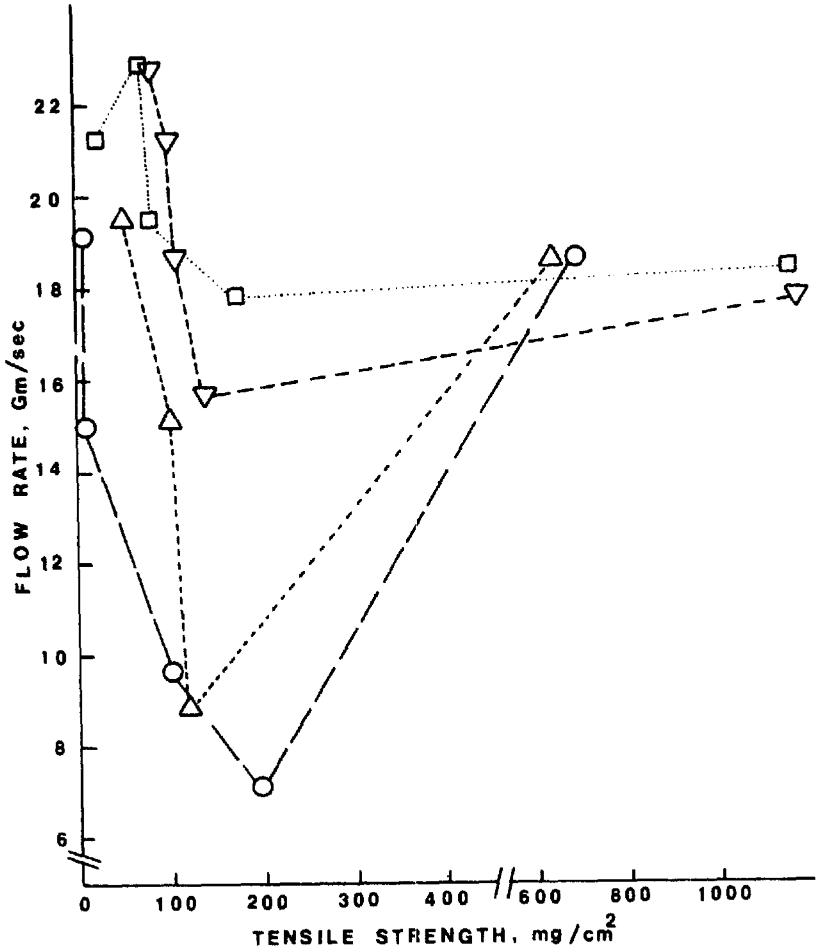

Fig. 8. Flow rate versus tensile strength of powder mixtures. Key: ○, magnesium stearate; △, silicon dioxide; ▽, talc; □, stearic acid.

results showing a linear relationship between the logarithm of tensile strength and the logarithm of flow rate.

In conclusion, it should be emphasized that the powder systems are complex and the relationship between tensile strength and orifice flow rate seen in these studies and the ones reported earlier (Varthalis and Pilder, 1977; Eaves and Jones, 1972) are applicable to only limited cases.

References

Augsburger, L.L. and Shangraw, R.F., Effect of glidants in tableting. J. Pharm. Sci., 55 (1966) 418–423.

Bagster, D.F. and Crooks, M.J., Evaluation of some properties of powder mixtures. Drug Dev. Ind. Pharm., 4 (5) (1978) 413–425.

Beverloo, W.A., Leniger, H.A. and Velde, J., Vande, The flow of granular solids through orifices. Chem. Eng. Sci., 15 (1961) 260–269.

Brown, R.L., Minimum energy theorem for flow of dry granules through apertures. Nature (Lond.) 191 (1961) 458–461.

Cheng, D.C.H., Farley, R. and Valentin, F.H.H., The effect of particle size and interparticle forces on flow properties of powders. I. Chem. E. Symposium Series, 29 (1968) 19–24.

Chowhan, Z.T. and Chow Y.P., Powder flow studies I. Powder consolidation ratio and its relationship to capsule-filling-weight variation. Int. J. Pharm., 4 (1980) 317–326.

Chowhan, Z.T. and Chow, Y.P., Powder Flow Studies III. Tensile strength, consolidation ratio, flow rate and capsule-filling-weight variation relationships. J. Pharm. Sci., 70 (1981).

Crooks, M.J., Ho. R. and Bagster, D.F., Tensile and shear testing of some pharmaceutical powders. Drug Dev. Ind. Pharm., 3(4) (1977) 291–308.

Eves, T. and Jones, T., Effect of moisture on tensile strength of bulk solids I: Sodium chloride and effect of particle size. J. Pharm. Sci., 61 (1972) 256–261.

Gold, G., Duvall, R.N., Palermo, B.T. and Slater, J.G., Powder flow studies II. Effect of glidants on flow rate and angle of repose. J. Pharm. Sci., 55 (1966) 1291–1295.

Gold, G., Duvall, R.N., Palermo, B.T. and Slater, J.G., Powder flow studies IV. Uniformity of flow: instrumentation and applications. J. Pharm. Sci., 57 (1968) 2153–2157.

Harmens, A., Flow of granular material through horizontal apertures. Chem. Eng. Sci., 18 (1963) 297–306.

Harwood, C.F. and Pilpel, N., The flow of granular solids through circular orifices. J. Pharm. Pharmacol., 21 (1969) 721–730.

Ho, R., Bagster, D.F. and Crooks, M.J., Flow studies on directly compressible tablet vehicles. Drug Dev. Ind. Pharm., 3(5) (1977) 475–487.

Jenike, A.W., Storage and flow of solids. Bulletin of the University of Utah, 53(26) (1964) 1–167.

Jones, T.M. and Pilpel, N., The flow of granular magnesia. J. Pharm. Pharmacol., 18 (1966) 429–442.

Jones, T.M., Mechanism of flow improvement by the addition of fine particles to bulk solids. J. Pharm. Sci., 57 (1968) 2015–2016.

Jones, T.M., The effect of glidant addition on the flowability of bulk particulate solids. J. Cosmet. Chem., 21 (1970) 483–500.

Kocova, S. and Pilpel, N., The tensile properties of mixtures of cohesive powders. Powder Technol., 7 (1973) 51–67.

Kocova, S. and Pilpel, N., Shearing and tensile tests on mixtures of pharmaceutical powders. J. Pharm. Pharmacol., 26 (1974) 11P–15P.

Kurihara, K. and Ichikawa, I., Studies on flowability of powders and interparticle cohesion. Chem. Pharm. Bull., 21 (2) (1973) 394–403.

Marshall, K. and Sixsmith, The flow properties of microcrystalline cellulose. J. Pharm. Pharmacol., 28 (1976) 770–771.

Nash, J.H., Leiter, G.G. and Johnson, A.P., Effects of antiagglomerant agents on physical properties of finely divided solids. Ind. Eng. Chem., 4(2) (1965) 140–145.

Peleg, M. and Mannheim, C.H., Effect of conditioners on flow properties of powdered surcrose. Powder Technol., 7 (1973) 45–50.

Pilpel, N. and Walton, C.A., The effect of particle size and shape on the flow and failure properties of procaine penicillin powder. J. Pharm. Pharmacol., 26 Suppl. (1974) 1P–10P.

Riley, G.S., Mann, S. and Jesse, R.O., Angle of repose of cohesive powders; International Conference of Powder Technology and Pharmacy 1–9, Sponsored by Powder Advisory Center, London, U.K.

Rose, H.E. and Tanaka, Rate of discharge of granular materials from bins and hoppers. The Engineer, 208 (1959) 465–469.

Train, D., Some aspects of the property of angle of repose of powders. J. Pharm. Pharmacol., 10 (1958) 127T–135T.

Verthalis, S. and Pilpel, N., Anomalies in some properties of powder mixtures. J. Pharm. Pharmacol., 28 (1976) 415–419.

Verthalis, S. and Pilpel, N., The action of colloidal silicone dioxide as a glidant for lactose, paracetamol, oxytetracycline and their mixtures. J. Pharm. Pharmacol., 29 (1977) 37–40.

York, P., Application of powder failure testing equipment in assessing effect of glidants on flowability of cohesive pharmaceutical powders. J. Pharm. Sci., 64 (7) (1975) 1216–1221.