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The application of capillary rheometry to the extrusion of wet powder masses

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Summary

Extrusion is currently used in the pharmaceutical industry in the production of spherical granules by spheronisation. The range of materials suitable for extrusion/spheronisation is extremely narrow, suggesting a specific rheological requirement for the process. It is the purpose of this paper to outline the well-established theory of capillary rheometry, initially developed for polymer systems, and to apply this theory to the extrusion of a wet powder mix used in the spheronisation process.

Introduction

Extrusion, a process by which materials can be formed into structures of controlled cross-section by forcing them from large diameter reservoirs through small diameter orifices (dies), is currently used in the pharmaceutical industry in the production of spherical granules by spheronisation (Reynolds, 1970). The range of materials suitable for extrusion/spheronisation is extremely narrow, suggesting a specific rheological requirement for the process. Unfortunately, the two parameters necessary to characterize the flow of a material through a die, i.e. the force required and the velocity of throughput, cannot be directly measured on commercial extruders and accurate data can only be achieved using simple ram extruders (Ovenston and Benbow, 1968). Ideally the information gained must be able to characterize a material independently of the extrusion system used. Unfortunately, there has been a tendency in the literature to concentrate on the actual operation variables rather than defining the rheological characteristics of materials, since the two are often difficult to separate.

In this paper an attempt has been made to characterize the rheological behaviour of a wet powder mass used in the spheronisation process by the application of the theory of capillary rheometry.

Theory

(I) Wall shear stress τ_w

Bagley (1957) has shown the shear stress at the die wall τ_w in a ram extruder with a die length L

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and radius R can be determined from the equation:

$$\tau_w = \frac{(P_T - P_0)R}{2L} \quad (1)$$

where P_T is the total pressure drop in the extruder, and P_0 an additional finite pressure loss between the pressure measurement and the true entry pressure into the capillary.

The additional finite pressure loss can be evaluated by determining the total pressure drop at different die lengths at fixed volumetric output and die diameter, and plotting these values against the length-to-radius ratio of the die. This enables both this finite pressure loss, which is usually described by the term 'upstream pressure loss', and the wall shear stress to be evaluated as the intercept and slope of the plot, respectively.

(II) Upstream pressure loss

Extrusion of metals, where die lubrication is used to minimize the wall shear stress provides a system in which the pressure required for extrusion will be equivalent to the upstream pressure as defined above. Alexander and Lengyl (1971) reported that the total pressure drop in aluminium extrusion has been found experimentally to be proportional to the logarithmic ratio of the cross-sectional area of the barrel to the die.

$$P_T = \tau_m \cdot \ln(A_0/A) \quad (2)$$

where τ_m is a constant associated with the resistance of a metal to deformation, A_0 is the cross-sectional area of the barrel, A is the cross-sectional area of the die and A_0/A is the reduction ratio.

Benbow (1971) used this relationship to determine an approximate yield value for a paste, independently of the velocity of throughput in a ram extruder.

Eqn. 2 takes no account of any additional pressure losses that could result from the frictional losses upstream of the die entrance and the pressure losses in the region unassociated with the convergence of flow (the redundant loss). Eqn. 2 was modified to take account of these possible errors according to Alexander and Lengyl (1971) to:

$$P_T = A_m - B_m \cdot \ln(A_0/A) \quad (3)$$

where A_m is the pressure loss in the 'redundant' zone and B_m the pressure loss in the 'convergent' zone.

Inoue et al. (1981) have reported that for a polymer system the extrusion pressure through a single-holed die at a single velocity of throughput was a linear function of the reduction ratio (rather than logarithmic), suggesting that the relationship between the upstream pressure loss and the reduction ratio is material-dependent.

When a material is forced through a ram extruder, it is subjected to a complex deformation, and the simple approach of metal extrusion cannot be used to evaluate yield values independently of the process conditions for all types of material. It is doubtful whether any single component of the finite pressure loss described by the upstream pressure loss can be differentiated by ram extrusion without additional rheological information gained from alternative rheometers such as oscillating systems (Bullivant and Jones, 1981).

(III) Wall shear rate

The flow of fluids whose properties are independent of time in the die of a ram extruder may be described in the form:

$$\frac{4Q}{\pi R^3} = \frac{4}{\tau_w^3} \cdot \int_0^{\tau_w} f(\tau)\tau^2 \, d\tau \quad (4)$$

where Q is the volumetric throughput, assuming that the velocity at the die wall is zero (Wilkinson, 1980).

It can be seen from Eqn. 4 that the functional relationship connecting the wall shear rate expressed as $4Q/\pi R^3$ and the wall shear stress will be independent of the radius of the die. Hence the value of the wall shear stress (determined from Eqn. 1), when expressed as a function of the shear rate, should lie on the same line independent of die diameter. However, in both polymer systems (Lupton and Regester, 1965) and concentrated suspensions (Berghaus, 1957) die radius dependency has been reported. Provided that the possibilities of time dependency and non-laminar flow are eliminated then the separation of these curves can be interpreted as evidence of anomalous flow behaviour, or effective slip near the die wall. An expression to take account of this change in flow behaviour close to the wall of the die was derived by Oldroyd (1949), who assumed that the rate of shear close to the wall of the die would not be a function of the wall shear stress alone. Therefore a further term $S(\tau_w)$ which will take account of the effective slip at the die wall was substituted into Eqn. 4 as follows:

$$\frac{4Q}{\pi R^3} = \frac{S(\tau_w)}{R} + \frac{4}{\tau_w^3}\int_0^{\tau_w} \tau^2 f(\tau)\,d\tau \quad (5)$$

By rearranging Eqn. 5 yields:

$$\frac{4Q}{\pi R^3}\tau_w = \frac{Z(\tau_w)}{R} + \phi(\tau_w) \quad (6)$$

where

$$Z(\tau_w) = \frac{S(\tau_w)}{\tau_w} \quad (7)$$

and

$$\phi(\tau_w) = \frac{4}{\tau_w^3}\int_0^{\tau_w} \tau^2 f(\tau)\,d\tau \quad (8)$$

Thus $Z(\tau_w)$ which has been described by Oldroyd (1949) as the effective slip coefficient can be evaluated by determining apparent fluidity $(4Q)/(\pi R^3 \tau_w)$ for each die radius at individual wall shear stress values. By plotting these values of apparent fluidity as a function of the reciprocals of the die radii, a straight line should result with a slope equivalent to $Z(\tau_w)$ and an intercept equal to $\phi(\tau_w)$. By repeating this procedure at different values of τ_w, $Z(\tau_w)$ can be evaluated as a function of τ_w.

Berghaus (1951) suggested that when a concentrated suspension flowed through a die a boundary layer of solvent would be found at the die wall. Since the boundary layer slides directly at the wall the slip produced will be dependent on the shear stress at the wall. Hence this boundary layer will be analogous to the general boundary layer hypothesis proposed by Oldroyd (1949). By modifying Eqn. 5, Berghaus (1951) developed a method of evaluating the anomalous flow behaviour at the wall of a capillary using the equation:

$$V_w(\tau_w) = R_2 \cdot \frac{R_1}{R_2} - R_1 \cdot \frac{1}{\tau_w^3}\int_0^{\tau_w} f_1(\tau_w) - f_2(\tau_w)\,dr \quad (9)$$

where V_w is the slip velocity and R_1 and R_2 are two different die radii.

The relationship between the apparent fluidity and $1/R$ has not always been found to be a simple proportionality for all rheological systems. Jastzrebreski (1967) reported that for concentrated pastes the effective slip coefficient was dependent on both the wall shear stress and die diameter, and accordingly modified Eqn. (5) to:

$$\frac{2Q}{\pi R^3 \tau_w} = \frac{\beta_c(\tau_w)}{R^2} + \phi(\tau_w) \quad (10)$$

where β_c is an alternative slip coefficient defined as $Z(\tau_w)/R$.

Eqn. 4 predicts that the shear rate (α) is a function of the shear stress (τ) and the material properties $f(\tau)$ and hence a plot of shear rate as a function of wall shear stress should be unique for each material tested provided that time dependence is absent, flow is laminar, and that there is no slip or the slip has been accounted for. This unique material property, 'its flow curve', can be fitted to various mathematical models. Farooqi and Richardson (1980) reported that pastes of kaolin could be described by the following model:

the Bingham Body Model: $\tau_w = \tau_y + k^{\alpha}$ (11)

where τ_y is the yield value and k is a constant, while Beazley (1972) showed that it was possible to detect 4 types of flow behaviour for a deflocculated clay suspension. The 4 types of flow behaviour were differentiated by 4 values for the degree of non-Newtonian flow when fitted to the power law model as follows:

$$\tau_w = k \cdot \alpha^{n'} \quad (12)$$

where k′ is the power law constant and n′ the degree of non-Newtonian flow. It would be advantageous to describe the two types of flow behaviour for the pastes reported by one mathematical model. The Herschel-Bulkley (1926) model combines Eqn. 11 and 12 and hence can generally be applied to pastes and wet powder mass systems. The Herschel-Bulkley model has been modified by Benbow (1971) and applied to the extrusion of a concentrated alumina suspension as follows:

$$\tau_w = \tau_y + k^* \cdot u^{(1-n')} \quad (13)$$

where k* is a modified power law constant.

It is the purpose of this paper to discuss the applicability of the theory described above to a particular wet powder mass used in extrusion/spheronisation.

Materials and Methods

Microcrystalline cellulose (Avicel PH101-FMC Corporation, Marcus Hook, Pennsylvania, USA) is commonly incorporated into formulations for the manufacture of spherical granules produced by extrusion/spheronisation (Reynolds, 1970). This material was combined with an equal quantity of the widely used water-soluble diluent lactose (Unigate regular grade) so that the formulation studied closely resembled a practical pharmaceutical formulation.

The blend of microcrystalline cellulose and lactose was mixed with a fixed quantity of water (1.2 times the weight of Avicel PH101 used) in a Hobart planetary mixer and passed through a 2.84 mm aperture screen to remove lumps. Fifty gram samples were taken from the bulk wet mix and each was packed to a constant volume into the 2.54 cm diameter barrel of a ram extruder by applying hand pressure. Each filled barrel was then mounted on the C-piece and aligned underneath the piston of the extruder which was attached to the crosshead of a servo-hydraulic press (Dartec, Stourbridge) via a calibrated load cell. The piston was then lowered into the filled barrel at various selected crosshead speeds and the displacement of this piston, once in contact with the packed material in the barrel of the extruder, was recorded against load on an X–Y recorder (Bryans Instruments, Mitcham, Surrey). Dies of varying diameter (1–2 mm) and lengths (2–20 mm) were located beneath the barrel of the extruder. The moisture contents of both the extrudate and the plug of material remaining in the barrel after extrusion were determined by drying overnight in an oven at 60°C.

Results and Discussion

The force/displacement profile recorded (Fig. 1) can be conveniently divided into 3 separate stages.

(i) Compression stage. The material packed into the barrel was found to consolidate into a dense plug prior to commencement of flow within the die. The apparent density of the plug (calculated by simple measurement of its weight and volume) at the commencement of flow within the die was found to be approximately equivalent to the apparent particle density of the wet powder mass, implying that the material had consolidated until all voidage had been eliminated. This equivalence of density is in agreement with the findings of Sheppard and Clare (1972) who showed that a metal powder densified to the value of the metal itself prior to flow in a ram extruder. The force/displacement profile at this stage was found to be

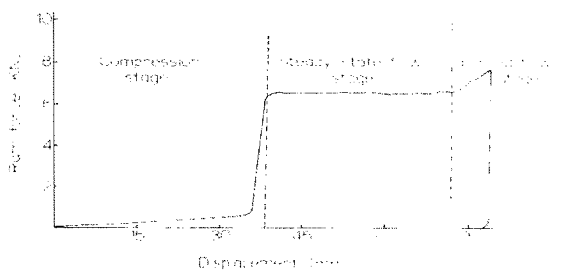

Fig. 1. A ram force/displacement profile for a microcrystalline cellulose/lactose/water mix (ram speed 4 mm·s^{-1}, die diameter 1.5 mm, L/R ratio 12).

independent of die dimension and extrusion rate, but dependent on barrel dimension and material mix.

(ii) Steady state flow stage. Once the material had begun to flow in the extruder, flow patterns in both the die and the barrel of the extruder were created. These patterns were found to remain unchanged provided that the extrusion force remained constant, i.e. a steady-state flow condition had been created (Harrison et al., 1984). The extrusion force monitored under these steady-state conditions was found to be independent of the packing of the material in the barrel and the position of the piston in the barrel, but dependent on the barrel diameter, die diameter, die length, die design and extrusion rate. The extrusion force (ram force) was found to increase with increasing die length and extrusion rate, and decrease with die diameter at a fixed barrel diameter. The dependency of the extrusion force on die length was found to be at variance with the results contained for metal extrusion where the extrusion force was independent of die length because the die wall was lubricated with a liquid film which minimized the shear stress at the die wall (Sheppard and Clare, 1972). Thus the dependency of extrusion force on die length in this wet powder mass extrusion implies that the wall shear stress will be dependent on the rheological properties of the wet powder mass and the experimental conditions selected.

(iii) Forced flow stage. As the piston approached the die, flow patterns within the barrel were no longer maintained resulting in an increased in extrusion force which was dependent on the displacement of the piston. The displacement of the crosshead of the servo-hydraulic press was stopped as soon as force flow was detected (Harrison, 1982).

The piston pressure values calculated from the force applied during steady-state divided by the surface area of the piston were plotted as a function of the length-to-radius ratios of the die (Fig. 2). The results shown in these plots confirmed the direct proportionality between piston pressure and length-to-radius ratio predicted from Eqn. 1. Hence it was possible to calculate a wall shear stress value and an 'upstream pressure loss' as the slope and intercept of these plots, respectively, at each extrusion rate. However, deviation from the simple proportionality was exhibited when the die length to radius ratio exceeded 16, independent of extrusion rate. This deviation was found to be more pronounced at large die diameters, where the die length was longer for an equivalent ratio, implying that the deviation had occurred because a moisture gradient had been created within the die during extrusion.

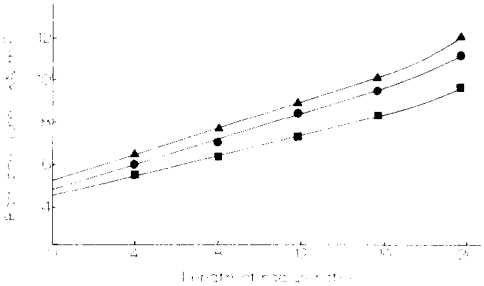

Fig. 2. Ram pressure as a function of length-to-radius ratio at various ram speeds (die diameter 1 mm). ▲, 6 $mm \cdot s^{-1}$; ●, 4 $mm \cdot s^{-1}$; ■, 2 $mm \cdot s^{-1}$.

The wall shear stress and the upstream pressure loss were calculated at each die diameter and extrusion rate; the data obtained are shown in Table 1. The upstream loss can be seen to be extrusion rate dependent presumably due to an increase in the frictional losses created by the shear of the material upstream of the die entrance as the extrusion rate was increased. Sheppard and

TABLE 1

Mean upstream pressure loss and wall shear stress values at different extrusion rates (ram speeds) and die diameters

Die diameter (mm)	Extrusion rate (ram speed) ($mm \cdot s^{-1}$)	Wall shear stress ($kN \cdot m^{-2}$)	Mean upstream pressure loss ($kN \cdot m^{-2}$)
1.0	1.5	215	4930
	2.0	238	5020
	4.0	291	5850
	6.0	301	6430
2.0	1.5	166	3260
	2.0	190	3340
	4.0	249	3620
	6.0	302	3870

Clare (1972) reported that the pressure exerted during metal extrusion, when lubricated dies were used, was extrusion rate dependent. This extrusion rate dependency was deemed to be insignificant when compared with the errors associated with the assessment of extrusion pressure. Consequently it was decided in the present work to determine a mean upstream pressure loss accompanied with a standard deviation to describe the extrusion rate dependence of the calculation mean upstream pressure loss.

In metal systems, the extrusion pressure has been divided into two parts, a redundant pressure loss and a convergent flow pressure loss which is dependent on the reduction ratio (Eqn. 3). Using this analogy, the change in the value of mean upstream pressure loss with die diameter will be associated with the convergent pressure loss, and the pressure loss independent of die diameter will be associated with the redundant pressure loss, which can be considered to be predominantly due to the yielding of the material prior to flow. The results in Table 1 clearly indicate that the convergent pressure loss is not simply a function of the logarithmic reduction ratio, as stated in Eqn. 3, and therefore it is not possible to separate the convergent pressure loss and the redundant pressure loss without studying the change at larger die diameters. Thus, the mean upstream pressure loss can only be used as a value to describe the pressure required to initiate flow in the die of a ram extruder at particular die diameters. When the wall shear stress/shear rate (as $4u/R$) was plotted, (Fig. 3) it was found that the flow curve at a die diameter of 1 mm had significantly shifted to the right. This suggests that one of the following could have occurred: (i) flow was not laminar in the 1 mm diameter die; (ii) the consistency of the material was altered during the passage through the die; and (iii) the velocity of throughput at the die wall was not zero.

The flow within the die was found to be laminar by studying the flow of 'coloured material' during extrusion (Harrison et al., 1984). When the apparent fluidity was calculated (Oldroyd, 1949), it was found that the calculated values passed through a minimum value as the velocity of throughput decreased. This minimum value occurred when a moisture gradient was detected between the plug of material in the barrel and the extudate, implying that at a velocity of throughput below the minimum value of apparent fluidity, the rheological nature of the material altered to maintain the flow of material at a particular wall shear stress. In consequence it was decided to ignore all the data collected beneath this minimum value and assume that the consistency of the material mix was constant above the minimum apparent fluidity.

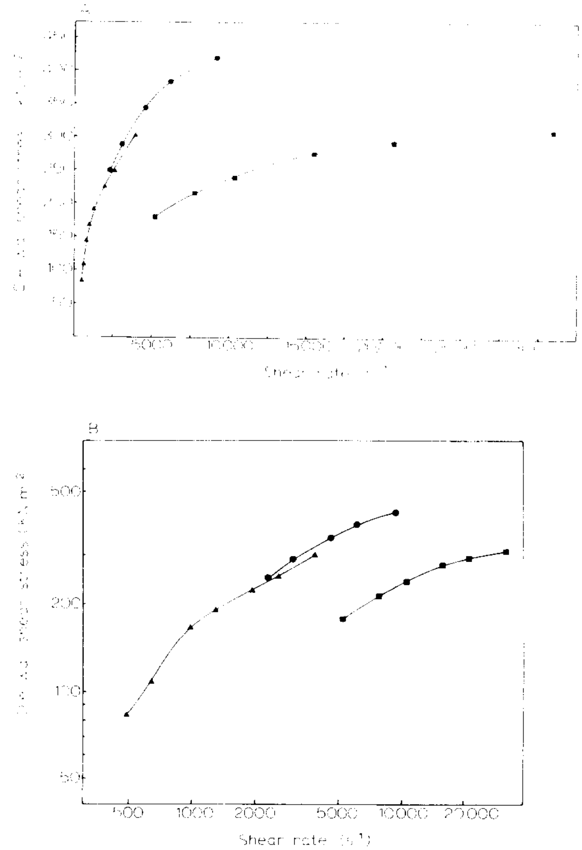

Fig. 3. Plots of die wall stress vs shear rate for dies of diameter ▲, 2 mm; ●, 1.5 mm; ■, 1 mm. A: linear plot. B: logarithmic plot.

It is therefore evident that the dependence of the shear stress/shear rate curves on die diameter was due to the existence of slip at the die wall. Thus before a mathematical model can be fitted to describe the rheological properties of this wet powder mass, the actual shear rate at the die wall has to be estimated by eliminating slip. This can be done by estimating a slip velocity at the die wall, substituting this value into Eqn. 13 and calculating the actual shear rate values. Table 2 gives values for the slip velocity estimated by the method described by Berghaus (1957). It can be seen that these values are dependent on the die diameter ratios used to calculate them implying that the slip velocity in this type of system is not a simple function of both the wall shear stress and die diameter as suggested by Oldroyd (1949) and Jastzrebreski (1967). The results at the two largest die diameters suggest a negative or negligible slip implying that the wall shear stress/shear rate profile for the 2 mm diameter die could be used to fit a mathematical model to this system. However, the non-linearity of the wall shear stress/shear rate profile for the 2 mm diameter die (Fig. 3) indicates that the material does not conform to a recognizable non-Newtonian system governed by the power law mathematical model. The deviation at low wall shear stress values can be considered to result from the significance of the yield stress in the estimated wall shear stress value. The deviation of high shear rate can be assumed to result from slip at the die wall, but in order to fit a mathematical model this deviation has to be ignored.

A mathematical model that takes account of both non-Newtonian flow and a yield stress is the Herschel-Bulkley model but unfortunately this model can only be solved tentatively as it requires the solution of three variables from two unknowns

TABLE 2

Calculated slip velocities (Berghaus 12) at selected wall shear stress values and die diameter ratios

Wall shear stress ($kN \cdot m^{-2}$)	Slip velocity at selected die diameter ratios ($m \cdot s^{-1}$)		
	1.0 mm/ 2.0 mm	1.0 mm/ 1.5 mm	1.5 mm/ 2.0 mm
157	1.09	0.82	0.28
205	1.69	1.17	0.15
253	3.16	2.03	−0.08
300	7.84	5.13	−0.12

(Harrison, 1982). A solution to this model gives values of n of 0.68 ± 0.01, k of $3.77 \pm 0.74 \times 10^{-4}$ and τ_w of 4788.8 m^{-2} — unrealistic results. A possible explanation of this is the fact that the model requires the determination of a power law function alongside two linear functions and hence small errors in the power law term result in the very large errors in the linear function. It must be concluded, therefore, that realistic values for the solution of this model will only be obtained if some estimate of the yield value of this material could be obtained.

A re-evaluation of the values obtained for the mean upstream pressure loss (Table 1) shows that the values are larger than those for the shear stresses. The upstream pressure loss is a term that describes the yield stress required to initiate flow in the barrel along with the frictional pressure losses associated with flow in the die. It is doubtful whether these frictional losses would be the predominant component of this term and hence the yield stress upstream of the die will probably be larger than the value of the wall shear stress in the die. Because of the large upstream pressure loss values and the poor fit of the experimental data to a mathematical model, it must be concluded that the wall shear stress values evaluated at different shear rates do not describe the rheological properties of the bulk material but only the properties of the lubricating layer which is itself dependent on the die diameter, extrusion rate and bulk properties. The thickness and consistency of this lubricating film can only be determined if the bulk of the material is assumed to flow through the die as a plug and the lubricating layer as an annulus. It is therefore essential to find a method to accurately determine the yield value of the material by either using wider barrel diameters or using an alternative rheometer to evaluate the yield value of a plug of material formed in the barrel of the ram extruder during extrusion.

Conclusion

While it has not been possible in this work to evaluate the exact rheological properties of a wet powder mass system consisting of microcrystalline cellulose, lactose and water by capillary rheometry due to die wall slip and complex changes associated with the convergence of flow into the die, rheological information has been gained to describe the system under experimental conditions similar to those employed in the extrusion and spheronisation of this material.

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Symbols

A_0	Cross-sectional area of the barrel
A	Cross-sectional area of the die
A_0/A	Reduction ratio
A_m	Pressure loss in the 'redundant' zone
B_m	Pressure loss in the 'convergent' zone
F_A	Force required to move material through the die
F_V	Force that tends to retard flow
L	Length of the die
P_0	Finite pressure loss due to viscous dissipation
P_T	Total pressure loss
ΔP	Pressure difference between two ends of a die
Q	Volumetric throughput
R	Radius of the die
$S(\tau_w)$	Die wall slip
V_w	Slip velocity
$Z(\tau_w)$	Effective slip coefficient
k	Power law constant
k*	Modified power law constant
n	Degree of non-Newtonian flow
r	Distance from centre of die
u	Velocity of throughput at a distance r from centre of die
α	Shear rate
α_w	Wall shear rate
β_c	Alternative slip coefficient
$\delta(\tau_w)$	Intercept in Oldroyd Eqn. 13
τ	Shear stress
τ_w	Wall shear stress
$\tau\mu$	Constant associated with the resistance of a metal to deformation
τ_y	Yield value.

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