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Effects of liquid phase migration on extrusion of microcrystalline cellulose pastes

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Abstract

The behaviour of water-based microcrystalline cellulose pastes undergoing ram extrusion has been investigated. Factors affecting the redistribution of water within the extruding paste and the upstream barrel compact, such as the initial water content, extrusion rate and die geometry, have been considered. The rates of dewatering for these given systems were characterised by the gradients of the extrusion pressure-ram displacement profiles. A linear relationship between the pressure-displacement gradient and the inverse square root of the paste velocity was obtained for a given paste and extrusion geometry. At velocities where water migration was significant, the extrudate was found to have a higher water content than that of the paste in the barrel at any given time; both the extrudate and the barrel paste decreased in water content with increasing ram displacement. Spheronisation of extrudate samples has shown that the redistribution of liquid during extrusion is an important factor affecting the quality of the spheres. A paste flow model, incorporating pseudo-plastic and shear deformation terms, was used to predict the change in extrusion pressures caused by liquid phase migration. The model parameters were obtained as functions of water content and gave good agreement with the experimental extrusion profiles. © 2000 Published by Elsevier Science B.V.

Keywords: Liquid phase migration; Microcrystalline cellulose; Ram extrusion; Spheronisation

1. Introduction

The manufacture of products by extrusion of solid–liquid pastes is widespread in the chemical, food and pharmaceutical industries. The bulk characterisation of the rheological properties of many pastes is now well established via a number of testing operations, such as ram extrusion (Benbow and Bridgwater, 1993). However, the effects of certain dynamic phenomena during extrusion, most notably liquid phase migration or 'dewatering', have yet to be fully described in a physically-based quantitative manner. The pharmaceutical industry frequently manufactures spherical pellets in the size range of 500–1500 μm using an extrusion/spheronisation process, which can be either filled into capsules or compacted into tablet form (Vervaet et al., 1995). The redistribution of liquid within a paste results in non-uniform material properties, such as the bulk yield stress, which can

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subsequently affect the extrusion process and product quality (Bains et al., 1991). Hence, an understanding of liquid phase migration is essential in designing the formulation and processing conditions for a given product.

2. Materials and methods

2.1. Material preparation

Mixtures of microcrystalline cellulose (MCC) powder (Avicel PH101, FMC Corp., Philadelphia, PA) and varying amounts of deionised water were prepared into pastes using the following standard procedure. The dry powder charge (range: 0.35–0.55 kg) was placed in a Hobart planetary mixer (A200 Model) and the water slowly poured onto the surface of the stirred powder bed. The paste was mixed for 10 min, after which pastes containing 45, 50 and 55 wt.% water (on a wet basis) were pugged once through a die plate featuring 4 mm diameter holes in order to break down agglomerates. Pastes containing 60 and 65 wt.% water were also prepared, but these were too wet to pug efficiently. The pastes were stored in sealed polythene bags for 24 h at room temperature to allow the water to equilibrate throughout the mixture. The freshly mixed pastes are often warm and inhomogeneity can be introduced by diffusion and evaporation during cooling in the open.

2.2. Experimental methods

2.2.1. Experimental apparatus

The extrusion apparatus consisted of a computer-controlled ram extruder (Dartec 100 kN screw strain frame), incorporating a cylindrical 25 mm internal diameter stainless steel barrel and various concentric cylindrical square entry dies (i.e. with an entry angle of 90°). A stainless steel ram was attached to the cross-head of the strain frame, with a close fitting phosphor-bronze seal that prevented leakage of the paste around the ram. The barrel was filled with paste and the ram operated at a given velocity. The applied force was monitored using a load transducer situated within the cross-head and the ram displacement was recorded. A detailed description of the apparatus is reported elsewhere (Burbidge et al., 1995). Extrudate samples were loaded into a 200-mm diameter cross-hatched plate surface spheroniser (Caleva Ltd., UK), running at a constant speed of 1000 rpm for 5 min per sample and the shape and size of the resulting particles were noted.

2.2.2. Characterisation

The pastes were characterised using the Benbow–Bridgwater procedure (Benbow et al., 1991), which allows the paste behaviour to be described in terms of six geometry-independent parameters. The subsequent flow equation predicts the extrusion pressure in terms of the paste velocity and the die and barrel geometry. The pastes were extruded through a number of cylindrical 3 mm diameter square entry dies of varying lengths (6–48 mm) at extrudate velocities ranging from 1.16 to 57.9 mm s^{-1}. The characterisation parameters were determined by a standard software package that incorporated a least squares fit of the extrusion pressure–extrudate velocity relationship.

The equation proposed by Benbow and Bridgwater, which is used to predict the extrusion pressure, P, in terms of the mean extrudate velocity, V, consists of the summation of two terms:

$$P_1 = 2(\sigma_{y0} + \alpha V^m) \ln\left(\frac{D_0}{D}\right) \tag{1}$$

$$P_2 = 4(\tau_0 + \beta V^n) \frac{L}{D} \tag{2}$$

where σ_{y0} is the initial bulk yield stress of the paste, τ_0 is the initial paste-die wall shear stress, α and β are velocity factors and m and n are velocity indices — these six terms are the so called characterisation parameters and hence, the terms within the brackets can be described as velocity-dependent stress parameters with zero factors; D_0 is the barrel diameter, D is the die diameter, and L is the die length. The first term, P_1, corresponds to the pressure drop due to the change in cross-sectional area during plastic flow deformation (Fig. 1); the second term, P_2, corresponds to the pressure drop caused by flow of the paste through the die. The latter term is mainly

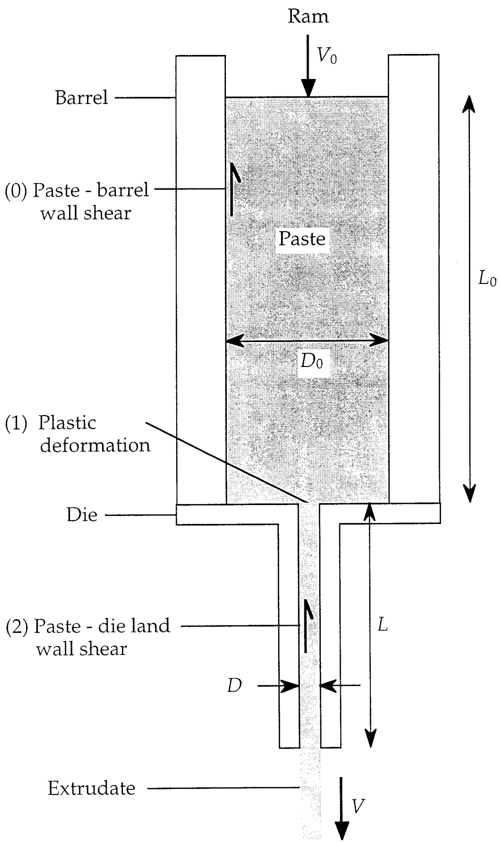

Fig. 1. Detail of the paste extrusion process.

due to frictional shear, particularly that associated with slip at the die walls. An additional pressure drop term can be included to account for shear at the barrel wall:

$$P_0 = 4(\tau_0 + \beta V_0^n)\frac{L_0}{D_0} \quad (3)$$

where V_0 is the ram velocity and L_0 is the height of the paste in the barrel. Since L_0 decreases during extrusion, this pressure drop term can be relatively significant for long ram displacements. However, for the characterisation process, P_0 was found to be negligible at the operating velocity range and ram displacements, in comparison with the other two pressure drop terms.

The resulting characterisation parameters are listed in Table 1. All the pastes show a yield dominance, with σ_{y0} and αV^m being $> \tau_0$ and βV^n for a given water content and extrudate velocity; all four parameters decrease with increasing water content. The yield and shear stresses confirm a logarithmic relationship with respect to water content (Bridgwater, 1996). There is no observable trend for m and n, although it should be noted that a value of n less than unity indicates a shear-thinning paste. The characterisation parameters thus generated were used in subsequent calculations to predict the extrusion pressure with respect to water content for a certain die geometry and extrudate velocity (see Section 3.1).

2.2.3. Extrusion study

2.2.3.1. The effect of extrusion velocity. It has been shown that the flow velocity is a strong factor governing the migration of water in extruding pastes (Harrison et al., 1985). In order to investigate the nature of the liquid movement, a series of extrusions were performed with a 55 wt.% water–MCC paste. A charge of ≈ 68 g of paste was put into the barrel and tapped down with the ram by hand, resulting in a paste height of ≈ 160 mm and was extruded through a 48 × 3 mm (length × diameter) die for a ram displacement of 100 mm.

Table 1
Characterisation parameters obtained using the Benbow–Bridgewater approach for MCC pastes of varying water content

Water content (wt.%)	σ_{y0} (MPa)	α (MPa m s^{-1})$^{-m}$	m	τ_0 (MPa)	β (MPa m s^{-1})$^{-n}$	n
45	1.4	5.8	0.21	0.39	2.0	0.59
50	0.61	4.7	0.28	0.080	0.61	0.29
55	0.19	2.7	0.34	0.051	0.42	0.37
60	0.027	0.40	0.37	0.011	0.076	0.42
65	0.0080	0.057	0.26	0.0012	0.016	0.37

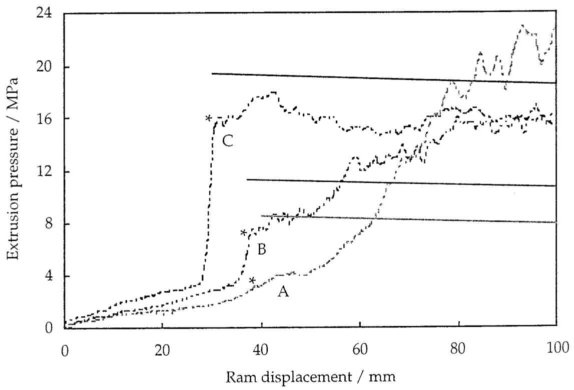

Fig. 2. Extrusion pressure-ram displacement profiles for MCC paste (initial water content 55 wt.%) through a 48 × 3 mm die at ram velocities (A) 0.0167 mm s^{-1}, (B) 0.0833 mm s^{-1} and (C) 0.833 mm s^{-1}; dotted lines = experimental results, solid lines = Benbow–Bridgwater predictions calculated using characterisation parameters for a constant water content of 55 wt.%; * indicates onset of extrusion.

The experiment was repeated for various ram velocities ranging from 0.0167 to 0.833 mm s^{-1} (corresponding to extrudate velocities of 1.16–57.9 mm s^{-1}) and the resulting extrusion pressure recorded. The apparatus was dismantled and the paste remaining in the barrel was carefully ejected by manually pushing the ram through the barrel. The resulting compact was cut into six discs of thickness 10 mm which were weighed and dried in an oven at 60°C for 24 h and the water content of each slice was determined to within ±0.3–0.4 wt.%.

Some of the obtained extrusion pressure-ram displacement profiles are shown in Fig. 2. The initial gradually increasing curve represents compaction of the paste within the barrel, leading to the onset of extrusion, when the pressure increases rapidly (especially for the higher velocities) due to the deformation of the paste at the die entry and the frictional shearing as it passes through the die land. As the ram velocity decreases, the onset of extrusion is delayed. This effect may be associated with a more effective particle packing at slower velocities (Youshaw and Halloran, 1982).

The initial value of the extrusion pressure decreases with decreasing velocity, as expected from the Benbow–Bridgwater equation. This equation, now including the paste-barrel wall frictional effect (P_0 from Eq. (3)), predicts a steadily decreasing extrusion pressure for the given characterisation parameters obtained for a paste of constant 55 wt.% water content; this is evident for the highest velocity. However, the pressure steadily increases during extrusion for the slower velocities, indicating an increase in magnitude of the yield and/or shear stresses brought about by a decreasing water content. The permeability of water through a soil is a function of porosity and degree of saturation (Huang et al., 1998). Similarly, the porosity of a compacted paste sample decreases with an increasing applied stress and therefore, extrusion at slower velocities produces a paste with a higher permeability, which allows the water to flow more freely.

At the lower velocities, a water-paste suspension was seen to drip out of the end of the die land before extrusion commenced and the first flow of extrudate was noticeably wetter than the main flow. This indicates an initial flow of water-saturated extrudate (Delalonde et al., 1996). Also, the onset of extrusion occurred at lower stresses than those predicted by the Benbow–Bridgwater equation, consistent with initial extrusion of a paste with a higher water content than that of the bulk. Flow instabilities resulting from severe liquid migration have also been observed for capillary extrusions of highly filled suspensions (Yaras et al., 1994).

Fielden et al. (1989) identified that the compaction stage prior to extrusion was the major contributor to water migration within MCC–lactose pastes. In view of this, all extrusion experiments are reported here in terms of ram, as opposed to extrudate, velocity. During single-ended pressing of dry powders in a similar geometry, a complex density distribution is developed within the compact due to the effects of die wall friction (Briscoe and Rough, 1998), with the average slice density usually decreasing down the compact in the direction of the ram movement. With a paste, however, the density distribution is affected by both the solids matrix porosity and the degree of liquid saturation. Accurate density measurements are required in order to confirm the development of a saturated paste zone at the bottom of the compact. However, the experimental errors involved in the simple sectioning and weighing analysis were too high to justify the calculations.

In order to investigate the water movement through the paste during compaction, a series of experiments was performed where the open die was replaced by a solid metal platen. A 55 wt.% water–MCC paste was packed by hand into the closed barrel and compacted at various speeds up to an applied pressure of 20 MPa (corresponding to ≈ 50 mm ram displacement). The paste compact was then ejected and sectioned into twelve 10-mm thick slices, which were dried in an oven. The subsequent calculated water profiles for two

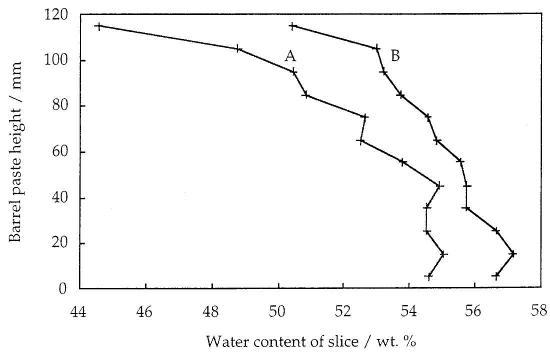

Fig. 3. Average water contents of 10 mm slices of MCC paste (initial water content 55 wt.%) compacted in a closed barrel up to 20 MPa applied pressure at speeds of (A) 0.0167 mm s^{-1} and (B) 0.0833 mm s^{-1}.

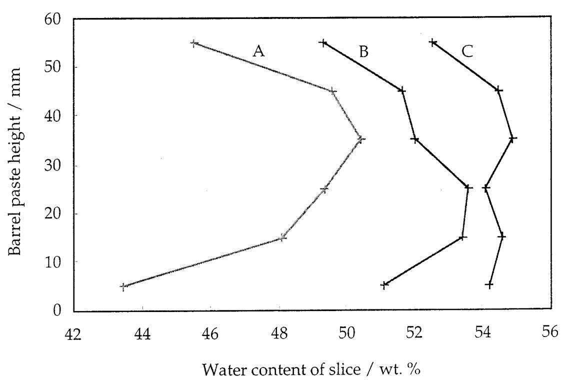

Fig. 4. Average water contents of 10 mm slices of MCC paste (initial water content 55 wt.%) remaining in barrel after extrusion through a 48 × 3 mm die at ram velocities (A) 0.0167 mm s^{-1}, (B) 0.0833 mm s^{-1} and (C) 0.833 mm s^{-1}; total ram displacement = 100 mm.

compaction speeds are shown in Fig. 3 and indicate a gradient of water content down each compact. This gradient is particularly steep at the very top and is probably due to leakage of water around the ram seal. The total water contents of the compacts were determined as 54.7 and 52.2 wt.% for ram speeds of 0.0833 and 0.0167 mm s^{-1}, respectively, indicating a considerable loss of water through the ram seal and/or die tooling at the slower speed. The loss of water over the paste in the barrel is 3.66 and 5.81 wt.%

Once extrusion commences in an open die system, the water gradient in the barrel is disrupted due to the formation of a static zone of drier paste near the die entry (Bayfield et al., 1998). The water content profiles of the paste remaining in the barrel after a ram displacement of 100 mm at various velocities through a 48 × 3 mm die system are shown in Fig. 4; these confirm a decrease in total barrel water content with decreasing ram velocity, along with a more severe water loss at the top of the compact and at the die entry. Upon dismantling the equipment, water (but no paste) was seen on the die-barrel seal for all velocities and around the ram seal for the lower velocities; hence, water is being lost into the extrudate and/or through the tooling, more so as the ram velocity decreases.

The gradient of the extrusion pressure–ram displacement profile for a given die system increases with decreasing ram velocity, which has also been observed for icing sugar pastes (Bayfield et al., 1998). By assuming that the extrusion pressure increases linearly with the ram displacement, the measured gradient appears to be directly proportional to the inverse square root of the ram velocity (Fig. 5); this is analogous to the Terzaghi one-dimensional consolidation model for soils, which predicts a linear relationship between the settlement of a sample and the square root of time (Craig, 1997).

2.2.3.2. The effect of initial bulk water content. The effect of the initial bulk water content of an MCC paste upon the extrusion pressure is shown in Fig. 6 for ram velocities of 0.0833 and 0.833 mm s^{-1} through a 12 × 1 mm die. The length of the initial compaction stage decreases with increasing water

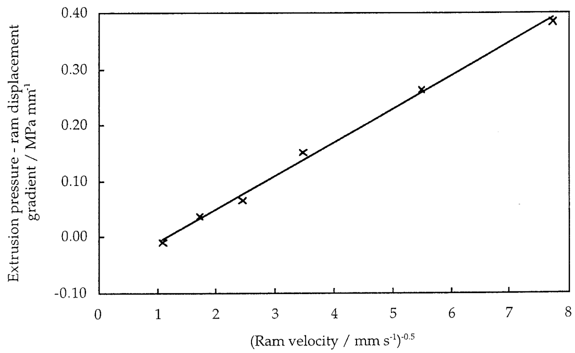

Fig. 5. Plot of extrusion pressure-ram displacement gradient against inverse square root of ram velocity for MCC paste (initial water content 55 wt.%); L/D = 48/3 mm, total ram displacement = 100 mm.

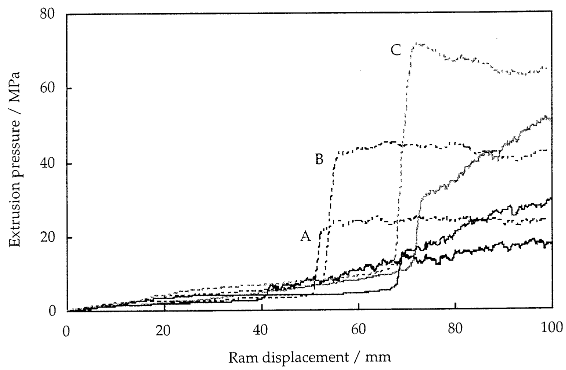

Fig. 6. Extrusion profiles for MCC pastes of initial water contents (A) 55 wt.%, (B) 50 wt.% and (C) 45 wt.% at ram velocities of 0.0833 mm s^{-1} (solid lines) and 0.833 mm s^{-1} (dotted lines); L/D = 12/1 mm.

content, implying more efficient particle packing (Harrison et al., 1985). At the higher velocity, the extrusion pressures are fairly constant, or steadily decrease (as shown by the 45 wt.% paste) due to the decreasing contribution of the paste-barrel wall shear stress as the compact height decreases. At the slower speed, an increase in extrusion pressure is seen and the pressure-displacement gradient increases as the water content decreases. This was also observed by Bayfield et al. (1998), and can be explained by the reduction in bulk yield stress for softer pastes, which results in less severe stress gradients within the deforming paste, thus decreasing the potential for liquid migration. The gradients were plotted against the inverse square root of the ram velocity for each paste (Fig. 7) and once again show a linear relationship.

2.2.3.3. The effect of die geometry. A number of extrusion runs were performed with 55 wt.% paste at a constant ram velocity of 0.0833 mm s^{-1} through dies of diameter 1 and 3 mm of various lengths. The paste remaining in the barrel after a ram displacement of 100 mm was sectioned and dried as before. For each diameter it was found that the die length, L, hardly affected the final water content profile in the compact (an example of which is shown in Fig. 8), indicating that the paste-die wall shear pressure term, P_2, weakly

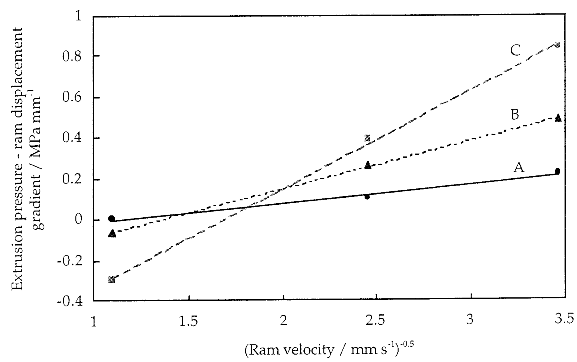

Fig. 7. Plot of extrusion pressure-ram displacement gradient against inverse square root of ram velocity for MCC pastes of initial water contents (A) 55 wt.%, (B) 50 wt.% and (C) 45 wt.%; L/D = 12/1 mm, total ram displacement = 100 mm.

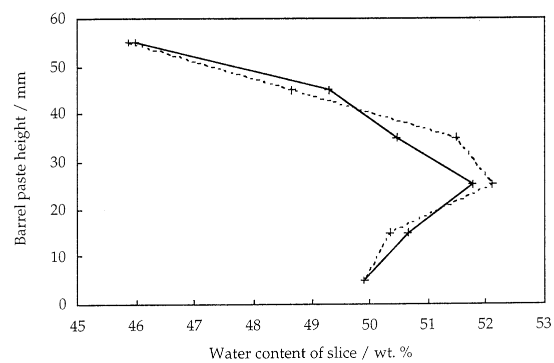

Fig. 8. Average water contents of 10 mm slices of MCC paste (initial water content 55 wt.%) remaining in barrel after extrusion through dies of L/D = 12/1 mm (solid line) and L/D = 4/1 mm (dotted line); ram velocity = 0.0833 mm s^{-1}, total ram displacement = 100 mm.

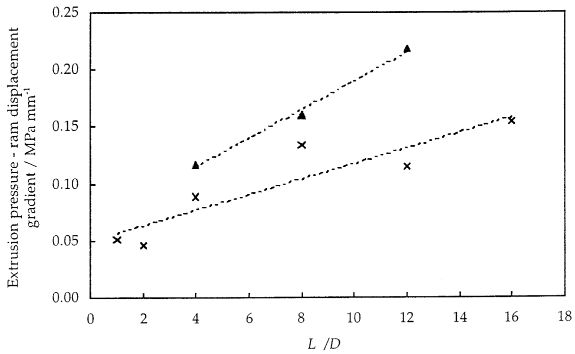

Fig. 9. Plot of extrusion pressure-ram displacement gradient against L/D for extrusion of MCC paste (initial water content 55 wt.%) through dies of diameter 1 mm (triangles) and 3 mm (crosses) of various lengths; ram velocity = 0.0833 mm s^{-1}, total ram displacement = 100 mm.

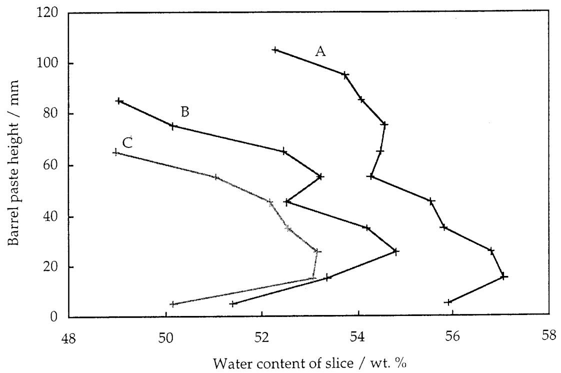

Fig. 10. Average water contents of 10 mm slices of MCC paste (initial water content 55 wt.%) remaining in barrel after extrusion through a 48 × 3 mm die for total ram displacements of (A) 50 mm, (B) 70 mm and (C) 90 mm; ram velocity = 0.0833 mm s^{-1}.

influences the mechanism of water migration in the upstream paste for a given die diameter (Eq. (2)). Newton et al. (1995) also found that the die length had no influence upon the quality of spheres produced by spheronisation of MCC–lactose–water paste extrudates. Hence, water migration is solely influenced by the stress distribution within the barrel arising from compaction and deformation of the paste. Hence, the pressure drop across the paste in the barrel only (P_0+P_1) and not the total extrusion pressure should be used to define the gradient for an indication of the rate of dewatering when comparing dies of different geometry. In these experiments, however, the pressure drop due to the die wall shear, P_2, cannot be uncoupled from the measured extrusion pressure. In order to achieve this, a system with a pressure transducer at the die entry would be appropriate.

As the diameter is decreased, the total water content of the compact was found to decrease, the average water contents being 51.5 and 49.5 wt.% for the 3 and 1 mm dies, respectively for all die lengths. Since the extrudate velocity, V, varies with the inverse square of the die diameter, the pressure terms P_1 and P_2 are expected to be higher for smaller diameters at a given L/D and ram velocity (Eqs. (1) and (2)); hence a greater rate of dewatering is seen as the diameter decreases. The gradients of the resulting extrusion pressure-displacement profiles are shown in Fig. 9 as a function of L/D and suggest a possible linear dependence, with the smaller diameter exhibiting higher gradients. The increase in total gradient with the die length for a given diameter and paste velocity is expected since P_2 is a function of L, although the contribution due to the increasing P_0 and P_1 terms via dewatering is also present. However, as explained previously, this increase in gradient cannot be used as an absolute measure of the rate of dewatering between dies of varying length.

2.2.3.4. Paste water content as a function of time. In order to investigate the water content of the extrudate and barrel paste with time, extrusion experiments were carried out at a ram velocity of 0.0833 mm s^{-1} through a 48 × 3 mm die, where the final ram displacement varied from 40 to 100 mm in 10 mm increments. Both the paste in the barrel and the extrudate produced during these intervals were sectioned and dried. The water contents of the paste in the barrel are shown in Fig. 10 for various ram displacements; the paste exhibits dewatering as the extrusion proceeds and the development of a static zone at the die entry. The highly compacted paste at the top also shows a loss of water, due to liquid migration into the paste below and/or through the ram tooling. Fig. 11 shows the average water contents of the extrudate collected during 10 mm ram displacement

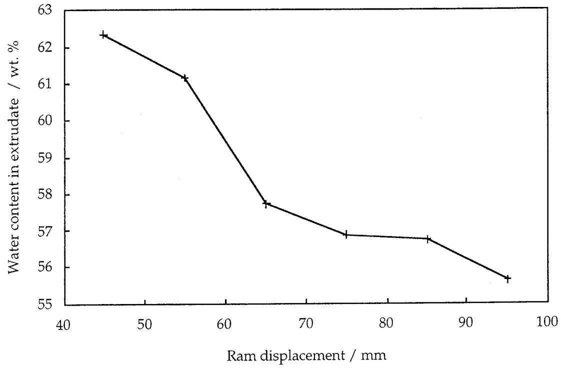

Fig. 11. Average water contents of extrudate collected in 10 mm ram displacement intervals for MCC paste (initial water content 55 wt.%); $L/D = 48/3$ mm, ram velocity = 0.0833 mm s^{-1}.

intervals; the extrudate has a higher water content than the paste in the barrel at any given time and the extrudate water content decreases throughout the experiment. These features were also reported by Yu et al. (1999) for alumina pastes.

2.2.4. Spheronisation study

MCC pastes of initial bulk water contents ranging from 45 to 60 wt.% were extruded through 4×1 and 12×1 mm dies at ram velocities of 0.0833, 0.167 and 0.833 mm s^{-1}. For each experiment, all of the extrudate collected over a ram displacement of 100 mm was spheronised. An indication of the size (measured to within ± 0.5 mm) and type of particle produced is reported in Table 2; the results are the same for both dies, i.e. the die length does not affect the sphere quality (Section 2.2.3.3). The 60 wt.% paste was too wet to spheronise properly and formed large agglomerates for all ram velocities, whereas the 45 wt.% paste produced cylindrical particles and a significant number of fines (small particles resembling a powder), due to an insufficient amount of water.

At relatively slow extrusion velocities, the extrudate has a higher water content than the bulk and decreases with time (Fig. 11); so if the extrudate water content lies within the range of 45–60 wt.%, one would expect spheres with a wider particle size distribution than those produced by extrusion at higher velocities (which have a narrower water content range). This is the case for the 55 wt.% paste, the maximum size of the particles increasing with decreasing extrusion velocity. However, an opposite trend is seen for the 45 wt.% paste. This can be explained by a more compacted extrudate being produced at the higher velocity, since the extrusion pressure is relatively high (Fig. 6) and is therefore more difficult to break up in the spheroniser. The quality of spheres produced by the 50 wt.% paste was independent of the velocities used; hence the extrudates lie within the domain unaffected by the dual liquid redistribution and compaction-related problems of spheronisation.

Table 2
Results from spheroniation study of MCC pastes of varying initial water content extruded at different velocities; $L/D = 12/1$ mm

Water content (wt.%)	Ram velocity (mm s^{-1})	Type of particle	Maximum particle size (mm)	Amount of fines
45	0.833	Cylinders	3.0	Many
	0.167		2.0	
	0.0833		1.5	
50	0.833	Spheres	1.0	Few
	0.167		1.0	
	0.0833		1.0	
55	0.833	Spheres	1.0	Very few
	0.167		1.5	
	0.0833		2.0	
60	0.833	Agglomerates	4.0	None
	0.167		4.0	
	0.0833		4.0	

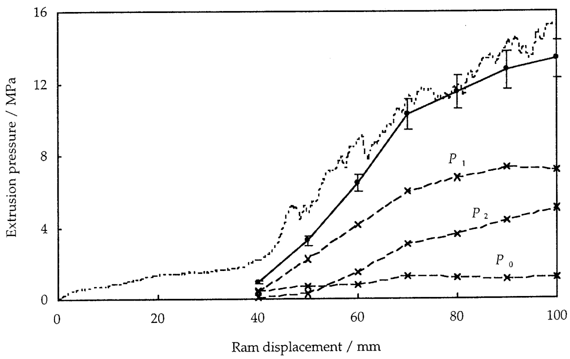

Fig. 12. Predicted extrusion pressure terms, P_0, P_1 and P_2 (dashed lines) and their sum (solid line) calculated using the Benbow–Bridgwater model for a 55 wt.% water–MCC paste undergoing dewatering during extrusion; $L/D = 48/3$ mm, ram velocity = 0.0833 mm s^{-1}; dotted line = experimental result.

3. Paste flow analysis

3.1. Prediction of extrusion pressures

As explained in Section 2.2.2, the Benbow–Bridgwater prediction of total extrusion pressure comprises the summation of three terms that represent the plastic deformation and frictional shearing of the paste. By using the characterisation parameters given in Table 1, each pressure term can be evaluated as a function of water content for a given die geometry and ram velocity; in this case a 48 × 3 mm die and a ram velocity of 0.0833 mm s^{-1} were chosen. The resulting generated data were fitted with parabolic expressions to produce pressure-water content calibration curves. For the P_0 term, a paste length of 10 mm was taken, since the sum of the shear contributions from these individual slices over the barrel compact length was used in the analysis.

The water contents of the 10 mm barrel slices determined at given ram displacements were used to calculate the corresponding pressure terms from the calibration curves; the bottom slice of the compact was assumed to undergo plastic deformation, thus producing the P_1 term. Burbidge et al. (1995) had proposed that the wetter paste upstream and not the static zone, undergoes this plastic deformation. After drying the bottom slice, the static zone could be broken away fairly easily exposing a truncated conical sample and so the volume occupied by the stagnant paste in this slice was estimated. This was ≈ 22% after a ram displacement of 100 mm, therefore the assumption of using the overall water content of the bottom slice for the die entry pressure drop was considered valid as a first approximation. The remaining slices of the compact contributed to the paste-barrel wall shear term P_0. The water contents of the extrudate collected during 10-mm ram displacements were used to determine the pressure drop due to the die wall shear (P_2). The predicted pressure terms and their sum are shown in Fig. 12 and show good agreement with the experimental results.

4. Conclusions

The redistribution of water in extruding microcrystalline cellulose pastes is an important feature that ultimately affects both the process performance and the final quality of spheronised products. Water migrates into the extrudate, resulting in non-uniform water contents within the barrel and the extruding paste. The flow of water through a material is controlled by the permeability of the solids matrix and hence, a relationship exists between the compaction history of the paste and its resulting water content distribution. The rate of dewatering can be characterised in certain cases by the extrusion pressure-ram displacement gradient and was found to be strongly dependent upon the paste flow velocity. For a given system, the length of the die does not seem to affect the mechanism of water migration.

A modified Benbow–Bridgwater analysis, which describes paste flow in terms of pseudoplastic deformation and wall shear terms, was found to describe the extrusion process adequately. This analysis incorporates the bulk yield stress and shear stress paste parameters, which are found to be logarithmic functions of water content. However, the analysis does not include the complications associated with a static zone at the die entry. With the development of a model predicting the flow of water through the solids matrix, obtained by relating the paste porosity to the permeability of saturated and unsaturated systems, the extrusion process can be investigated fully. A model of this nature shall be proposed by the authors in a future article.

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