A study of surface fracture in paste extrusion using signal processing

B. D. RUSSELL
Department of Chemical Engineering, New Museums Site, Pembroke Street, Cambridge CB2 3RA, UK

S. BLACKBURN
IRC in Materials Processing, University of Birmingham, Edgbaston, Birmingham B15 2TT, UK

D. I. WILSON*
Department of Chemical Engineering, New Museums Site, Pembroke Street, Cambridge CB2 3RA, UK
E-mail: ian_wilson@cheng.cam.ac.uk

Published online: 21 April 2006

Gross circumferential fracture (GCF) is a surface defect sometimes observed during the extrusion of particulate pastes. In this study particular attention was paid to the frequency of fracture in pastes undergoing ram extrusion through dies with circular, square and rectangular slot cross section. Three methods for estimating the fracture frequency were employed. The first was determined from the observed fracture periodicity, the second was determined from the application of short time Fourier transformation (STFT) techniques to the pressure signal measured at the die plate and the third was the application of the geometrical result first reported by Domanti and Bridgwater (2000). All were found to be in agreement within experimental error where failure was observed. In some materials the fracture frequency was still observed in the frequency domain after fracture ceased in the extrudate, indicating the existence of a flow instability in the paste: if the extrudate was smooth throughout the process STFT detected no peak. Fracture was more pronounced with short dies and/or where the paste experienced high wall shear stress. Square dies were more prone to fracture than circular dies (corner effects) and the fracture spacing could be related directly to the hydraulic diameter. Rectangular slot dies were more prone to irregular fracture. It was shown that lowering the die wall friction by using Teflon-lined dies reduced the propensity of the paste to fracture for all die geometries tested.

1. Introduction

Solid-liquid pastes featuring a high volume fraction of solids (typically $> 50\%$) frequently exhibit visco-plastic behaviour and are extruded to generate a wide range of products with controlled shape and relatively high density. Surface defects are a particularly unwelcome feature owing to the loss of surface finish or their impact on the mechanical strength of the product. The liquid fraction can provide some degree of interparticle cohesion (e.g. via capillary interaction) but the tensile stresses generated during extrusion can exceed this and lead to sharkskin [1] or gross circumferential fracture (GCF) as illustrated in Fig. 1. This work will refer to the surface defects as GCF, to differentiate from the sharkskin term used in polymer literature. This is because the physical systems, and thus fracture mechanisms, are believed to be significantly different.

Domanti and co-workers [2, 3] undertook a systematic study of GCF in ceramic pastes, and compared their observations with models for fracture or flow instabilities based on ductile metal strain analyses such as that by Pugh and Low [4]. These metal analyses were not able to explain all the features observed, which is not unexpected given that pastes exhibit non-isotropic strength (weaker in tension) and often feature a range of crack initiators, e.g. agglomerates, air bubbles, laminations (where flows join) *etc*. An alternative approach is to attribute GCF to phenomena which are known to cause surface fracture in polymer

*Author to whom all correspondence should be addressed.

0022-2461
DOI: 10.1007/s10853-005-5154-y

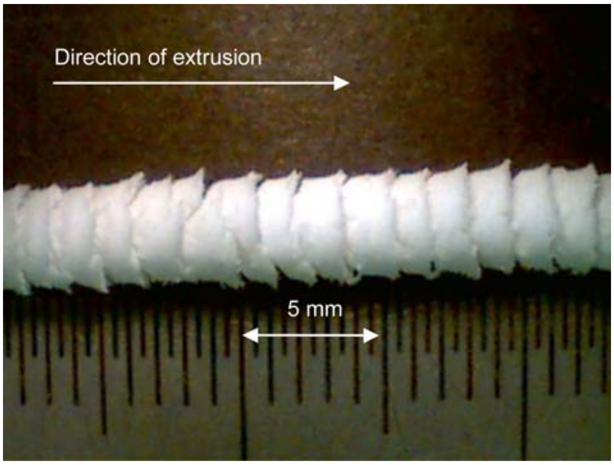

Figure 1 Illustration of gross circumferential fracture for a microcrystalline cellulose paste undergoing ram extrusion. Conditions: die land length/diameter $L/D = 48/3$ mm/mm, mean extrudate velocity $V = 10$ mm/s.

extrusion, including surface instability (e.g. stick-slip, [5]), flow instabilities (e.g. [6]) and compressibility effects (e.g. [7]). The difficulty arising in these approaches is how to quantify the parameters corresponding to melt properties for these heterogeneous multi-phase materials. This paper reports phenomenological observations which suggest that a complete understanding of GCF in pastes will feature contributions from both of these areas.

Previous phenomenological observations include those by Domanti *et al*. [3], who reported that increasing the die land length, L, and decreasing the die entry angle reduced surface fracture in all pastes tested. For their α-alumina based pastes containing glucose additives in particular, it was found that increasing the extrusion velocity and using a smaller reduction ratio increased the severity of surface fracture, in agreement with the findings of Harrison *et al*. [1], who also noted that GCF in pharmaceutical pastes was much less likely to occur for dies with a L/D ratio (where D is die land diameter) greater than 1. Kulikov and Hornung [8] reported that GCF in clay/oil mixes extruded through tubular dies could be avoided by using an annular die with a long central rod, but the mechanism for this result has yet to be established. Stick-slip mechanisms are known to cause surface defects in the ejection of compacts (e.g. [9]) featuring particulates with low liquid content, but the existence of a Coulombic friction or sticking wall interaction is seldom reported for pastes. On the contrary, their steady shear behaviour is often characterised by extensive slip at solid walls [10].

The mechanisms underlying GCF and related phenomena in pastes are not well understood and there is therefore a need to provide more information on the processes involved. This paper presents results from a study of surface fracture where signal processing techniques have been used to extract quantitative information on characteristic stresses and forces associated with fracture from normal stress measurements made by pressure transducers located near the extrusion die plate. Three pastes exhibiting different fracture behaviour were studied in a ram extrusion system and the extracted parameters are compared with qualitative observations.

An important feature explored in this work is the observation by Domanti and Bridgwater [2] that the period of fracture observed in their circular die geometries, i.e. the spacing between encircling circumferential cracks, was approximately equal to the die radius. For a process operating at constant throughput, this geometrical result links the frequency of fracture to the die geometry, and suggests that forces associated with the fracture mechanisms should feature this periodicity. For axisymmetric ram extrusion from a circular barrel of diameter D_{B} through a die land of diameter D at ram velocity V_{R}, the associated 'defect frequency', F_{D}, is given by

$$F_{\mathrm{D}} = \frac{V_{\mathrm{R}}}{\lambda_{\mathrm{p}}}\left(\frac{D_{\mathrm{B}}}{D}\right)^2 \tag{1}$$

where λ_{p} is the fracture period ($= D/2$ for circular dies). Amarasinghe and Wilson [11] used simple frequency analysis methods to interrogate pressure transducer data collected during experiments where various pastes exhibited GCF on extrusion through circular dies. Discrete peaks in the power spectrum were evident at the F_{D} value but the magnitude of the periodic component was not discussed in detail. This paper builds on that earlier work, employing more rigorous techniques to search for periodic components and quantifying these components for different materials, geometries and surfaces. The effect of geometry and particularly corners on GCF is also investigated, as is the scope for reducing GCF by using dies lined with Teflon (PTFE).

2. Experimental

Ram extrusion experiments were performed at room temperature using an instrumented, computer-controlled strain frame (Dartec SA 100 Load Frame with stroke transducer, Tamworth, UK) configured to act as a ram extruder. The paste was loaded into a 190 mm high cylindrical barrel with inside diameter 25 mm, compacted, then extruded through a range of axisymmetric square-entry dies (i.e. 90° entry angle). Further details of the extruder configuration can be found in Chen *et al*. [12]. Stainless steel was used as the material of construction unless stated otherwise. Extrusion pressure measurements were made using 3 mm diameter solid stage pressure transducers (Kulite XTM 190, Basingstoke, UK) calibrated over the required stress range. The transducers were located flush with the die plate or along the die land. Comparison studies indicated that the fluctuations in pressure signal were not significantly affected by the location of the transducer. Transducer signals were logged at speeds up to 1 kHz, depending on extrusion velocity: the sampling rate was selected to ensure that F_{D} was smaller than the Nyquist frequency.

TABLE I Rheological characteristics of pastes using 6 parameter Benbow-Bridgwater correlation (Equation A1)

Parameter	Starch [14]	Detergent	Glucose ceramic
σ_0 (MPa)	0.11	0.46	0.20
α (MPa.(s.m^{-1})$^{-m}$	1.3	1.5	1.7
m	0.33	0.15	0.91
τ_0 (MPa)	0.125	0.30	0.049
β (MPa.(s.m^{-1})$^{-n}$	0.30	2.1	0.13
n	0.36	0.61	0.46

Three different pastes were used in this work: a potato starch-based food dough (subsequently termed 'starch paste'), a detergent paste, and an α-alumina based ceramic paste similar to that used by Amarasinghe and Wilson but with the addition of glucose syrup which had been shown to increase the likelihood of fracture [13].

The detergent paste was used primarily as a control, as this material very rarely exhibited fracture in previous experiments. This material was therefore used to establish whether periodic signals, particularly those close to F_D, were evident when no fracture was observed visually. The glucose paste was known to fracture easily, and afforded comparisons with the starch paste in order to determine if the amplitude of the periodic signals associated with fracture, where this occurred, depended on the material, the process conditions, or both.

Additional shear behaviour measurements were carried out using a parallel plate controlled stress rheometer (Bohlin Instruments, Gloucestershire, UK, Model CVO 120 High Resolution) on the detergent and glucose pastes. The starch paste, due to its powdery nature, could not be tested in this device. Using roughened 25 mm diameter plates, 3 mm apart, the rotational velocity was increased until flow of the paste material became apparent. Internal deformation rather than slip was observed with the roughened plates and this allowed an estimate of an internal yield stress under shearing conditions, albeit in conditions dissimilar to those encountered in the die land during extrusion. The detergent paste did not flow at all within the parameters of the test; however, the glucose paste appeared to have a yield shear stress of approximately 20 kPa.

The rheological behaviour of the pastes was characterised separately using the approach described by Benbow and Bridgwater [10] summarised in Appendix 1 and the results are presented in Table I. A detailed rheological investigation of the starch paste was reported by Cheyne *et al.* [14]. The values of σ_0 indicate that for the experiments performed in this study the contribution of the total extrusion work due to deformation is relatively large compared to the shearing work in the die land which is related to τ_0, although geometric factors also play a role. Thus these types of pastes are termed stiff pastes, and they usually feature slip flow in the die land. Additionally, the values of the stress velocity-dependency factors m and n are all less than 1, indicating *shear-thinning* behaviour (i.e. viscosity decreases as more work is applied). It is also interesting to note that the detergent paste featured a noticeably larger τ_0 value than the other materials, indicating that the die-land contribution to the work required for extrusion is greater than the other cases.

TABLE II Parameters studied with variation in die diameter, D. Note orifice dies used, $L \sim 0$

D (mm)	L (mm)	V_R (mm/s)	No. of trials
1.5	0	0.5, 1, 5, 10	12
3.5	0	0.5, 1, 5, 10	12
4.5	0	0.5, 1, 5, 10	12
6.0	0	0.5, 1, 5, 10	12
9.0	0	0.5, 1, 5, 10	12
12.0	0	0.5, 1, 5, 10	12

TABLE III Parameter range in study of variation in die land length, L

L (mm)	D (mm)	V_R (mm/s)	No. of trials
6	3	0.5, 1, 5, 10	12
12	3	0.5, 1, 5, 10	12
24	3	0.5, 1, 5, 10	12
36	3	0.5, 1, 5, 10	12
48	3	0.5, 1, 5, 10	12

TABLE IV Parameter range in study of variation in die land velocity, V

V_R (mm/s)	V (mm/s)	$\dot{\gamma}_a$ (s^{-1})	D (mm)	L (mm)
0.5	35	93	3	6
1	69	185	3	6
5	347	926	3	6
10	694	1852	3	6

Since previous studies (e.g. [2, 15]) have shown that D, L, V, die geometry and die lubrication have an effect on the occurrence of circumferential surface fracture, these were all varied in the tests summarised in Tables II–IV. Studies over a range of ram velocities allowed the frequency dependency of the periodic stress components to be established as (*i*) either geometry dependent (as in equation (1)) or (*ii*) an inherent feature of the material (e.g. a natural relaxation characteristic). All data sets indicated that the periodicity, where observed, was directly related to the ram velocity, i.e. (*i*). Where fracture was observed, the angle, θ, periodicty λ_p and depth of fracture were also measured (see Fig. 2). Three different frequencies will be discussed: the (predicted) defect frequency, F_D; the observed frequency of fracture, F_O, determined from the measured periodicity and extrusion speed after an experiment; and the fracture frequency extracted from signal processing analysis, F_F.

The internal microstructure of selected detergent and glucose-ceramic extrudates was studied post extrusion using an X-Tek HM160 Computed Tomography System, with a spatial resolution of approximately 30 μm. No distinct variations in material density or porosity were evident over cross sections of fractured extrudates, except that the material bordering a fracture showed a small reduction in density (expansion) and the presence of cracks.

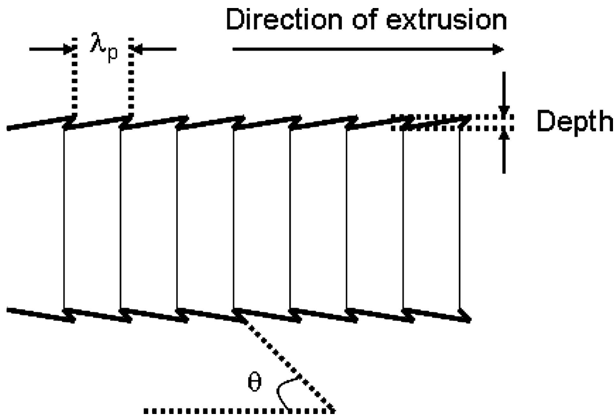

Figure 2 Illustration of physical measurements (period, angle and depth of fracture). Angle (θ) did not vary significantly in these tests and was typically 60° to the extrudate axis.

TABLE V Summary of die geometries

Die geometry	A_D (mm^2)	d_H (mm)	L (mm)
Circle	28.3	6	6
Slot	30	6.4	6, 48
Square	30.3	6.4	6

The effect of die geometry and surface condition was investigated using the series of dies detailed in Table V and Fig. 3. The non-circular dies all featured similar cross-sectional areas to the 3 mm circular dies. Separate extrusion experiments indicated that the force required to extrude the pastes through the different die geometries was reasonably well predicted by the Benbow-Bridgwater model given in Appendix I (equation A2) for brass dies. The use of Teflon die land surfaces, however, significantly reduced the force or extrusion pressure required to extrude the paste through the die at a given value of V, as well as reducing the extent of fracture. Separate extrusion tests with the glucose-ceramic paste indicated that similar extrusion pressures were observed for the brass and Teflon dies when the flow rate through the latter was raised by 100% for shorter round and square dies and 50% for longer round dies. This is an important observation as effects of die land (or capillary) materials are rarely reported in paste extrusion studies: where slip is a dominant phenomenon, it is understandable that the factors controlling adhesion to the wall will vary with the interaction (surface energy) of the wall material. Onoda and Hench [16] describe several methods by which wall adhesion can be modified by changing properties of the wall and the extrudate – including surface interaction (likely to be the dominant factor in the case of Teflon) and surface roughness. It was noteworthy that the Teflon surfaces were found to be more rough ($Ra \sim 0.6\ \mu$m) than the brass surfaces ($Ra \sim 0.3\ \mu$m), so that the use of smoother Teflon surfaces could be expected to give further reduction in defects.

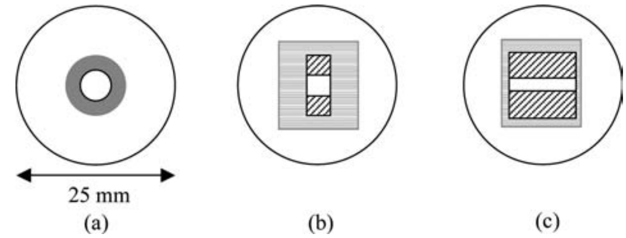

Figure 3 Schematic of die land geometries detailed in Table V. Grey area indicates region covered by Teflon in reduced wall friction dies. Hatched areas indicate recessed entry. (a) circular, 3 mm die land; (b) square; (c) rectangular slot. Not to scale.

3. Spectral analysis

Amaransinghe [17] used a conventional Fourier Transform (e.g. [18]) to calculate the average amplitude of a particular frequency component in a pressure signal over the sampling time. If the amplitude of the periodic signal varies with time, then conventional Fourier analysis gives erroneous results so the Short Time Fourier Transform (STFT) was employed here. The STFT divides the data set into a number of contiguous blocks, of length N, and transforms this shorter data series into the frequency domain using the conventional Fourier Transform. The average value of the amplitude of the sinusoidal component in this short block is simply the spectral density, Φ, of the relevant frequency component. Ideally, N will be very small but Fourier methods require many data (ideally, infinity) in order to give accurate information [19]. A compromise is therefore required between useful time domain resolution (small N) while preserving accuracy (large N). Numerical investigations [20] have indicated that for the series studied here, an N value of approximately 100 yields a reasonable compromise.

Other real-time spectral analysis methods have also been considered, such as wavelet analysis [19] and a Bayesian parametric estimator which has been investigated in detail by Bretthorst [21] and also Ó Ruanaidh and Fitzgerald [22]. These have proved more versatile for continuous extruders (see [20]) whereas the STFT is sufficiently robust for analysis of ram extrusion data.

4. Results and discussion

4.1. Occurrence of fracture

Circular dies will be considered first. GCF was not observed with the detergent pastes in any of the experiments outlined in Tables II–V. Some variation in surface roughness was occasionally evident, such as is shown in Fig. 4 for an orifice die (negligible length, where steady flow in the die land is not accomplished). This type of surface defect is not periodic in nature and is not considered further here. No periodic stress components were expected for this case: F_D for this experiment was 13 Hz, and the power spectrum in Fig. 5a confirmed the absence of any periodic stress components. This result contrasts sharply with the starch paste under the same process conditions,

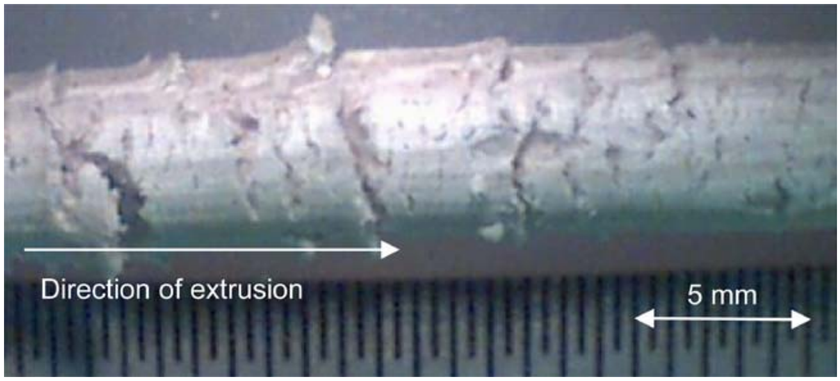

Figure 4 Illustration of non-periodic surface roughness observed on detergent extrudate under severe conditions (orifice die, $D = 9$ mm, $V_R = 1$ mm/s).

where fracture was observed at an F_O value of approximately 13 Hz. The corresponding power spectrum from this run in Figs. 5b and c shows a distinct peak at approximately 12 Hz. Most cases with circular dies featured closer agreement between F_O and F_F than shown here: some of the mismatch arises from error in the die and velocity measurements, and the assumption of paste incompressibility.

The starch and glucose pastes proved more successful in terms of promoting fracture. However, it was noted that in all cases where GCF was observed, this occurred in the initial stages of extrusion; thereafter, the extrudates appeared smooth. This is discussed in further detail below. Occurrence of fracture was found to depend strongly on the die land length. For $D = 3$ mm dies, fracture was observed consistently with starch dough for L/D values of 6/3 and 12/3 mm/mm, while the glucose paste only exhibited GCF with the $L/D = 6/3$ mm/mm die (or shorter). No fracture was observed with $D = 1.5$ mm dies for either paste, whilst for diameters greater than this fracture did occur.

The depth of fracture did not appear to depend on the diameter for a given value of V_R, and was almost constant at approximately 0.5 mm at $V_R = 1$ mm/s. For both glucose and starch materials, in cases where the die geometry promoted fracture, the depth of fracture depended on ram extrusion velocity. At low velocities, such as $V_R = 0.5$ mm/s, the average depth of fracture was approximately 0.5 mm for $D = 3$ mm. With the highest velocity, $V_R = 10$ mm/s and $D = 3$ mm, this increased to approximately 1.5 mm. These observations are consistent with the behaviour reported by Domanti and Bridgwater for ceramic paste formulations.

4.2. Detection of periodic stress components

Where fracture was observed visually in circular dies, a peak in the frequency domain was also detected, similar in frequency to both F_D and F_O. Where no fracture was observed visually, as with all detergent paste tests, no peaks were evident in the frequency domain close to F_D (or F_O). Fig. 6 shows the correlation between the calculated parameters, F_D and F_F, and the measured value, F_O. The plot features many data points overlapping, and shows that all three indicators were effectively equal, within the bounds of experimental error. The mechanism underlying GCF is thus demonstrated to be strongly related to geometry in these configurations, confirming the result of Domanti and Bridgwater [2].

The direct correlation between the occurrence of fracture and the existence of a distinct peak in the frequency spectrum, in the region of F_D, suggests that signal processing techniques could be employed to detect or monitor the occurrence of periodic surface fracture. Russell *et al.* [20] discuss the applicability of different algorithms for estimating F_F on-line. It should be noted that false negative results, i.e. where fracture was observed visually and no peak was evident in the frequency spectrum, were only recorded in tests featuring low fracture frequencies where the periodic component was not distinguishable from the low frequency noise.

False positives, i.e. where a periodic component was observed in the frequency spectrum but fracture was not observed visually, were *not* recorded under circumstances where no fracture occurred. However, periodic components corresponding to F_D continued to be evident in the frequency spectrum in many tests after visible fracture had stopped. Examples of this behaviour for the glucose and starch pastes are shown in Fig. 7, which plots the amplitude of the periodic component corresponding to fracture, a_F, calculated by STFT, over time. For the starch paste in Fig. 7a, surface fracture was only observed during the initial stages of extrusion, i.e. [$t < 50$ s], after which the extrudate surface appeared smooth. The Figure shows that the end of visible fracture was accompanied by a change in the magnitude of a_F. In the case of the glucose paste in Fig. 7b, fracture was found to occur at 6.8 Hz and was also only observed prior to $t \sim 10$ s, but in this case the end of visible fracture is not accompanied by a clear change in a_F: this difference was observed consistently. This observation suggests that GCF is associated with a flow instability, which generates stresses in the extrudate which are large enough to overcome the cohesive interactions in the paste. For a ceramic or metal, the growth of a fracture would be described in terms of a fracture toughness or brittle failure strength (see, for example, the

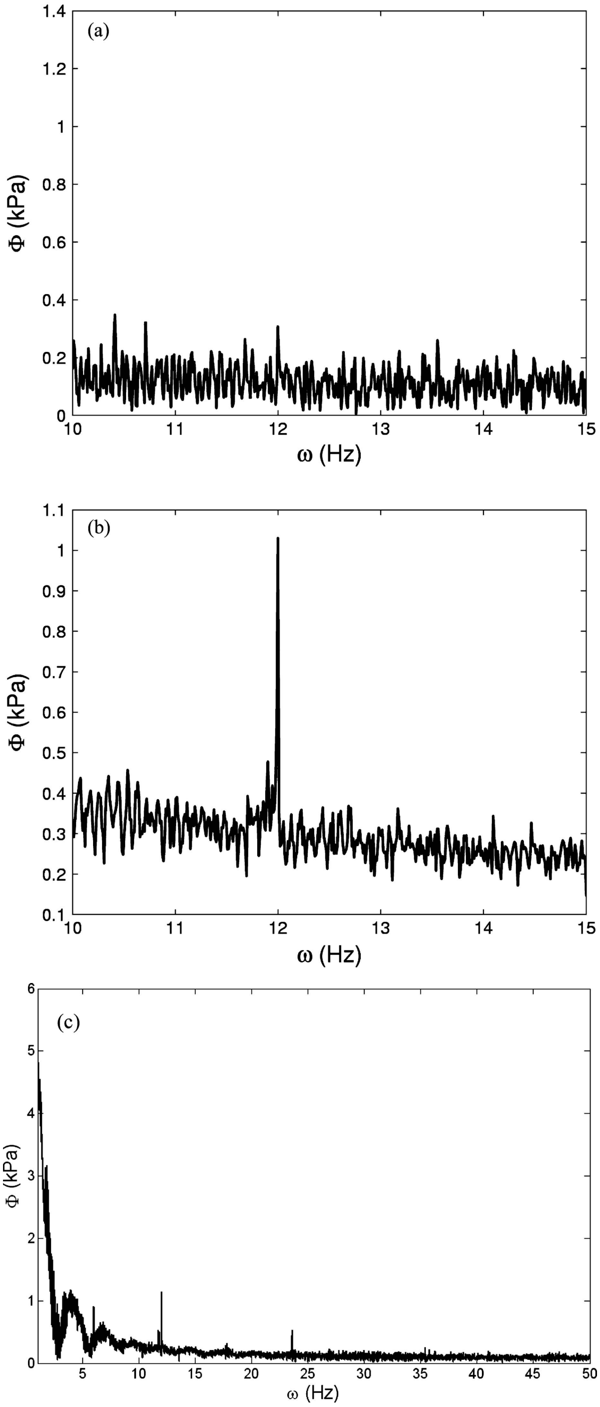

Figure 5 (a) Spectral density of data gathered from ram extrusion of detergent paste with no regular fracture observed, as in Fig. 4; (b) Spectral density of data gathered from ram extrusion of starch paste when $F_O \sim F_D = 12$ Hz; (c) Plot of entire spectrum up to Nyquist Frequency (50 Hz), showing harmonic at 24 Hz. Conditions: $L/D = 0/4.5$ mm/mm, $V_R = 1$ mm/s.

analysis by Bentham and Koiter [23] for growth of an edge crack in a cylindrical bar under tension), but analyses of this form rely on elastic energy considerations which are not immediately transferable to visco-plastic materials such as these pastes. However the disappearance of GCF on further extrusion, despite the presence of periodic stresses, indicated that the tendency of the material to fracture did not remain uniform. It was noteworthy

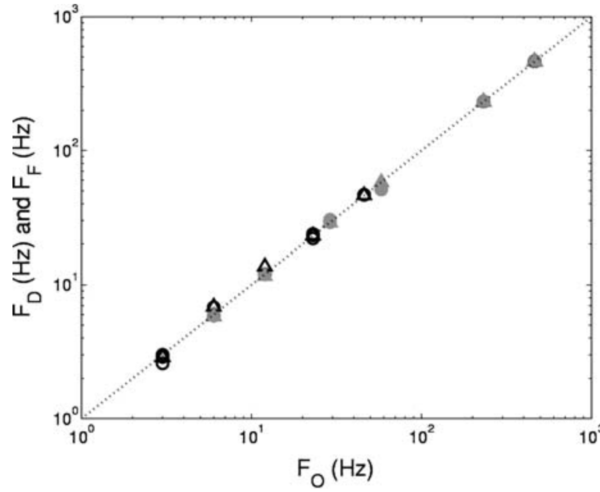

Figure 6 Comparison between observed frequency of fracture (F_O) and defect frequency (F_D—triangles) and peak in frequency domain (F_F—circles) for circular dies (various lengths). Grey—glucose paste, black—starch paste; dashed line shows line of equivalence.

that the mean extrusion pressure did not vary significantly over the course of these experiments apart from the initial transient as paste filled the die land etc.

The most likely explanation of this behaviour was that the ram extrusion configuration generated a distribution in material cohesion (or fracture toughness), either via compaction of material to eliminate voids (potential crack initiation sites) or density (and therefore strength). Such variations are likely to arise during the compaction of these materials in the barrel, where small frictional forces can result in density differences and a variation of stress transmitted through the material. The consolidation step is unlikely to feature wall lubrication, which is established during steady flow. This postulation was tested by two experiments for each paste. Firstly, extrusion was performed with a varying level of fill in the barrel and uniform consolidating stress (∼4 MPa). No fracture was observed when the barrel was filled to less than two barrel diameters, whereas GCF was observed during the initial stages of extrusion for greater levels of fill. Secondly, the starch paste billets generated by the compaction stage were removed from the barrel and the strength of the paste quantified using an indentation tester: the glucose paste was not stiff enough in the green state to give meaningful data. Fig. 8 shows the strength – presented as the maximum force recorded by the indenter – as well as the a_F parameter obtained from extrusion of a similar billet. The compact strength was determined by the amount of force required by an indenter to break the section of paste taken from the barrel. The paste was positioned on a flat plate, so that this was not a standard bending test but affords a comparative measure of the compact strength. It can be seen that fracture occurs when the initial, weak, paste is extruded, marked by a large a_F value: the periodic flow instability remains after fracture has finished, when more cohesive material is being extruded, and its amplitude increases – albeit without incurring fracture – as the final,

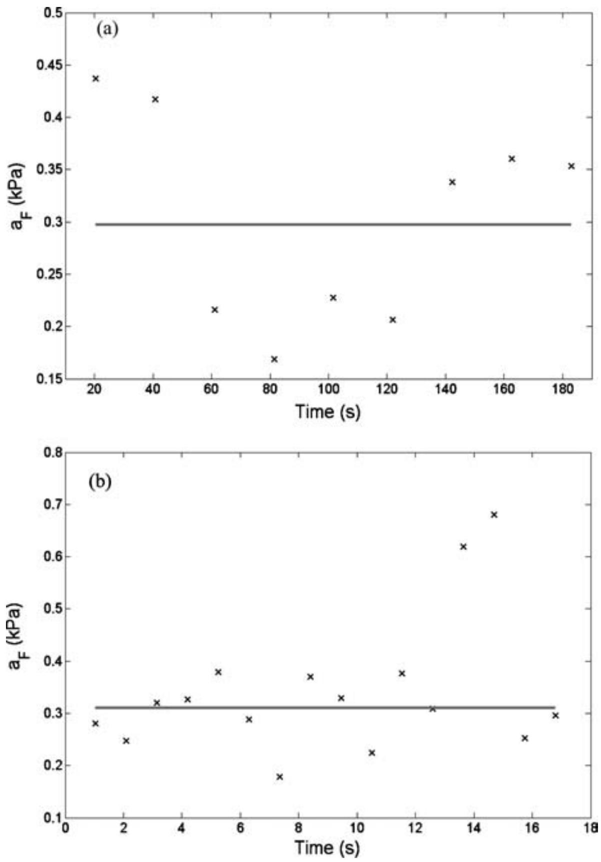

Figure 7 Amplitude of periodic stress component, a_F, during ram extrusion of (a) starch paste (conditions: $L/D = 0/4.5$ mm/mm, $V_R = 0.5$ mm/s); (b) glucose paste (conditions: $L/D = 6/3$ mm/mm, $V_R = 5$ mm/s). Crosses: amplitude from STFT, grey line: amplitude from overall FT.

stronger material passes through the die. This behaviour is consistent with the observations of Pugh and Low [4] and Kulikov and Hornung [8], who observed a reduction in fracture by increasing the stress or pressure on the material being extruded by raising the hydrostatic pressure and increasing frictional resistance post extrusion, respectively.

These pastes therefore exhibit features associated both with granular solids, particularly sharp fracture, and continuum materials such as polymers and metals. The underlying mechanism, linking periodicity and geometry, has not been elucidated further here, but the pool of available information has been extended.

4.3. Amplitude of periodic stress components

It was not possible to obtain a_F values for the detergent paste as this did not exhibit fracture under these tests. Fig. 9 compares the values obtained for the glucose and starch materials during fracturing periods against the effective wall shear stress, τ_{eff}, for all sets of ram velocity, die land length or die diameter studied. The τ_{eff} values were estimated from the measured extrusion pressure values using $= \Delta P\ D/4L$, where the pressure difference was corrected for any die entry contribution in the transducer

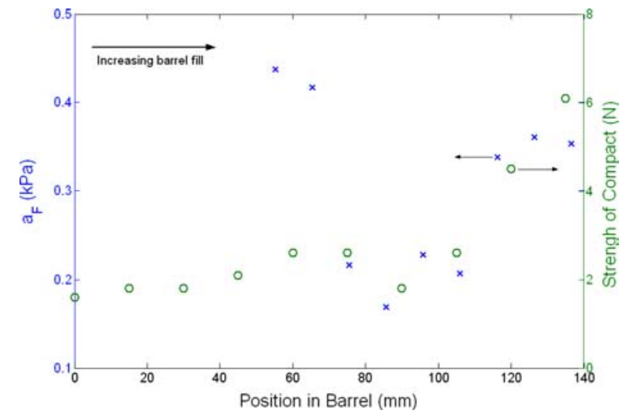

Figure 8 Relationship between starch paste strength measured by indentation testing of a compacted billet (circles) and amplitude of periodic stress component (crosses). Data plotted against location in barrel billet after compaction or a_F corresponding to when a zone would be extruded. Periodic stress component taken from data in Fig. 7a. Conditions: $L/D = 0/4.5$ mm/mm, $V_R = 0.5$ mm/s.

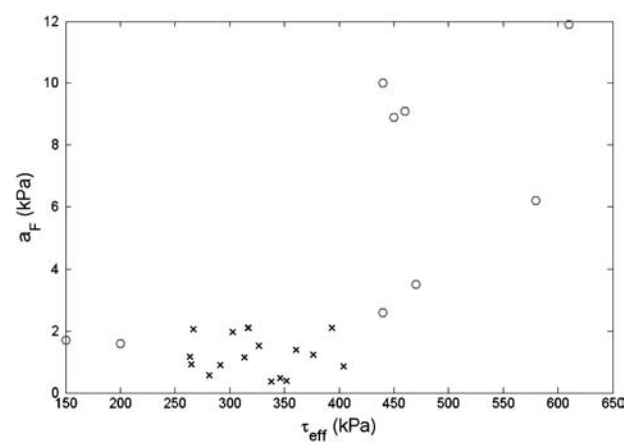

Figure 9 Effect of wall shear stress on the amplitude of periodic stress component from ram extrusion of starch (crosses) and glucose (circles) pastes. Circular dies, various *L*/*D* and velocities.

reading using equation (A1). The individual data sets did not show any systematic difference for each of these variables. It is immediately evident that a_F appears to be relatively insensitive to the nature of the material, with a weak dependence on τ_{eff}, suggesting a weak frictional interaction, or possibly a compressibility contribution. The magnitude of the a_F values is also noteworthy, at two orders of magnitude lower than the estimated wall shear stress, and around three orders of magnitude smaller than the corresponding mean extrusion pressures. This result is consistent with the observations that ceramic and other green pastes are very weak in tension (Benbow and Bridgwater [10]) compared to compression. An interesting area for further work would be to compare these a_F values with estimates of cohesive interactions owing to liquid bridge effects.

The observed depth of fracture did not map simply with a_F, being more sensitive to ram velocity and die land shear stress rather than die land length and absolute extrusion pressure. This suggests that the depth of fracture is related

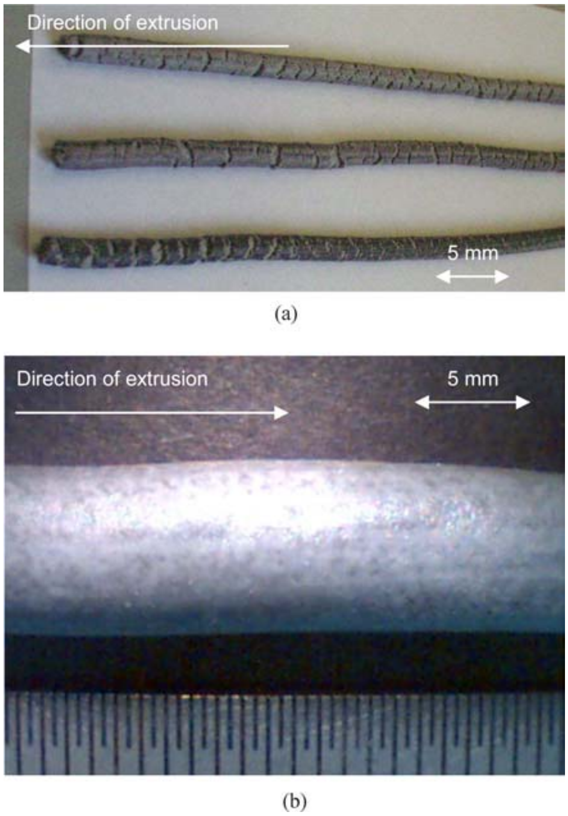

Figure 10 Elimination of GCF using a Teflon die land wall in circular geometry. (a) steel die; (b) Teflon lined die. Conditions: glucose ceramic paste, $L/D = 6/6$ mm/mm, $V_R = 1$ mm/s.

to the elastic strain energy generated by the shear stress in the die land which is released on fracture.

4.4. Effect of die wall lubrication

Table VI summarises the experiments with circular dies which were repeated with a Teflon sleeve inserted in the die land. Significant reductions in the extent and occurrence of GCF were observed for these Teflon-lined dies, which could be attributed to the reduced level of die land wall shear stress compared to the brass or steel versions at a given velocity. The recorded extrusion pressures were consistently lower for the Teflon dies than for their brass/steel counterparts, which was attributed to die land wall effects as the die entry component is dominated by internal extensional shear rather than wall conditions.

The role of die land lubrication was investigated by a crude attempt at matching the die land shear stresses. A reference was generated using a steel or brass die of a particular geometry at a ram velocity of 0.5 mm/s. Extrusions were then performed for the same material using the Teflon die land of the same geometry, with the ram velocity varied until the extrusion pressure matched that in the reference. Table VII reports the ratio of ram velocities used for Teflon over the steel/brass equivalent in this 'shear stress matching' exercise. The die entry component on the extrusion pressure will also increase with ram velocity, but the *m* values in Table I indicate that this contribution generally varies less strongly than the die land shear stress component. The circular and square dies required the same velocity increase for shear stress matching, which could be related to the fact that the square geometry is reasonably axisymmetric.

In many cases, the Teflon inserts eliminated GCF completely, as illustrated in Fig. 10. Signal processing confirmed the absence of periodic stress components. These results indicated that the onset of fracture is not simply a function of wall shear stress, rather that the response of the material is dependent on geometry, wall interactions and paste state.

4.5. Non-circular geometries

The use of die lands with corners in the duct cross-section introduced a different form of fracture, described by Onda and Hench [16] as 'edge tearing', 'dog's teeth' or 'feather edging'. The corners act as stress concentrators and therefore represent fracture initiation sites. Fig. 11a shows edge tearing in a square cross-section die, while Figs 11b and c show acute cracking along the short side of a slot die which we here term 'fronding'. The latter image shows that the periodicity of fracture on each side of the slot die could vary independently. It is noteworthy that these materials exhibit little die swell: this is chiefly a crack onset phenomenon.

As with circular dies, the depth of fracture was most sensitive to the ram velocity. The average depth of fracture was taken as the maximum extrudate width (distance from tip of one 'frond' to the other) minus the minimum extrudate width. The average depth of fracture at low ram velocities, 0.5 mm/s, was approximately 0.5 mm, which increased linearly to *c.* 2 mm when extruded at velocities of 10 mm/s. No dependence on the die land length was noted in terms of fracture occurrence for both the square and slot dies.

As with circular dies, periodic stress components were not evident in the STFT spectra for non-circular dies when

TABLE VI Parameter range in study of die land wall lubrication

Die geometry	L (mm)	V_R (mm/s)
Circle ($D = 6$ mm)	6	0.5, 1, 5, 10
Slot (15×2 mm)	6	0.5, 1, 5, 10
Slot (15×2 mm)	48	0.5, 1, 5, 10
Square (5.5×5.5 mm)	6	0.5, 1, 5, 10

TABLE VII Shear stress matching conditions (Sanders-Hewett and Davies [13])

Die geometry	Teflon:Steel/Brass V Ratio
Circle, $L/D = 6/6$ mm/mm	2.0
Slot, $L = 48$ mm	1.4
Slot, $L = 6$ mm	3.4
Square, $L = 6$ mm	2.0

(c)

Figure 11 Illustrations of fracture observed during ram extrusion of glucose paste through (a) square die (5.5 × 5.5 mm); and (b) slot die (2 × 15 mm), regular fracture, (c) slot die, irregular fracture and 'fronding'.

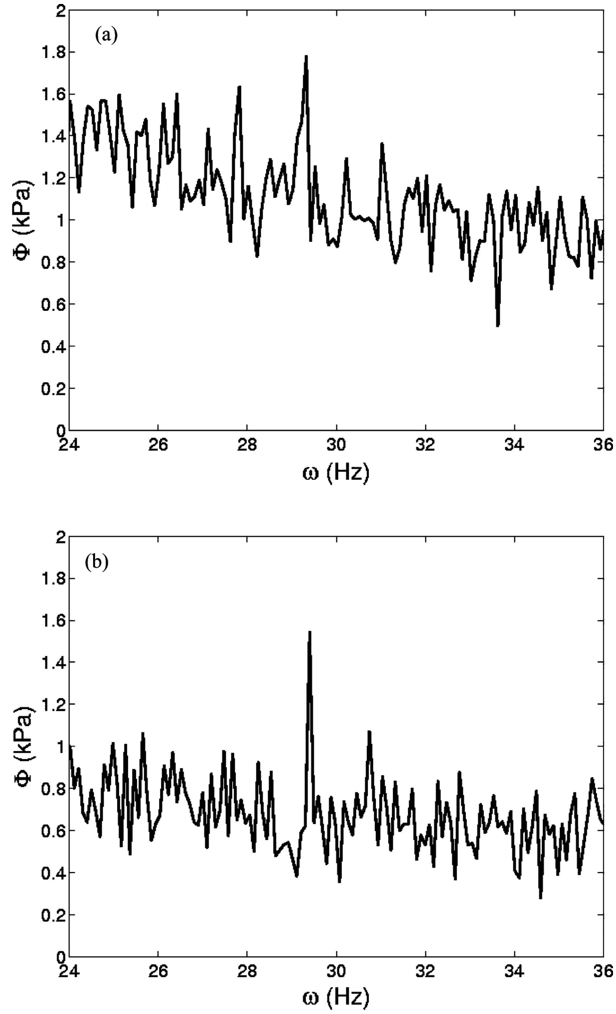

Figure 12 (a) & (b) Spectral density of starch paste extrusion data from notionally identical tests when $F_{\mathrm{O}} \approx F_{\mathrm{D}} \sim 30$ Hz for square die (5.5 × 5.5 mm) showing different baseline behaviour. Conditions: $V_{\mathrm{R}} = 10$ mm/s.

no visible fracture was observed. Where fracture was observed with square dies, peaks close to the F_{O} value were observed but were occasionally masked by higher levels of noise in the spectra. This is illustrated by Fig. 12 for notionally identical extrusions with the starch paste, where F_{O} was *c*. 30 Hz. The base line noise in Fig. 12a shows a slight decrease with increasing frequency as one would expect for white noise, but with Φ values significantly greater than those in 12b and which almost swamp the fracture peak. In both cases, the fracture was periodic and was observed throughout the entire extrusion. For non-circular dies, the corners acted as permanent crack initiation sites and promoted GCF throughout the extrusion.

As with circular dies, F_{F} correlated very closely with F_{O} for the square dies. When it was possible to identify a peak corresponding to the frequency of fracture, the temporal behaviour in a_{F} mirrored the trend reported above for circular dies. Fig. 13 shows such a profile for a case with a square die where fracture did cease during extrusion, after *c*. 4 s. Continuous fracture was the norm, so removing corners from die designs, or reducing their interaction with the material, therefore appear to be logical methods to avoid GCF in extruding complex shapes.

Data gathered from slot die experiments exhibited few peaks in the frequency domain, as the 'fronding' observed with these dies was not as periodic as in circular or even square dies. It is noteworthy that the fronding was restricted almost entirely to the shorter sides of the dies,

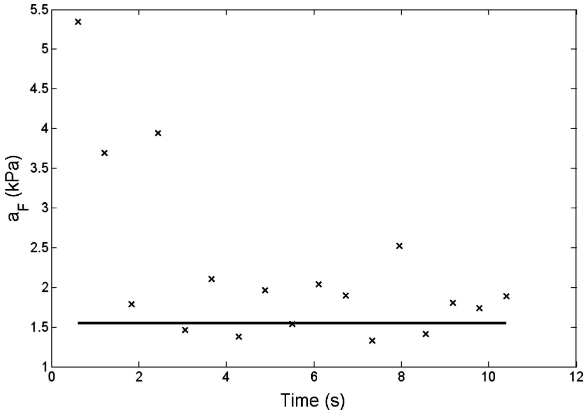

Figure 13 Amplitude of periodic stress component for starch paste extrusion through a 5.5 × 5.5 mm square die, when $F_O \approx F_F \sim 30$ Hz. Conditions: $V_R = 10$ mm/s. Fracture ceased in this case after ∼ 4 s. Solid line indicates average periodic stress component, taken from complete FFT.

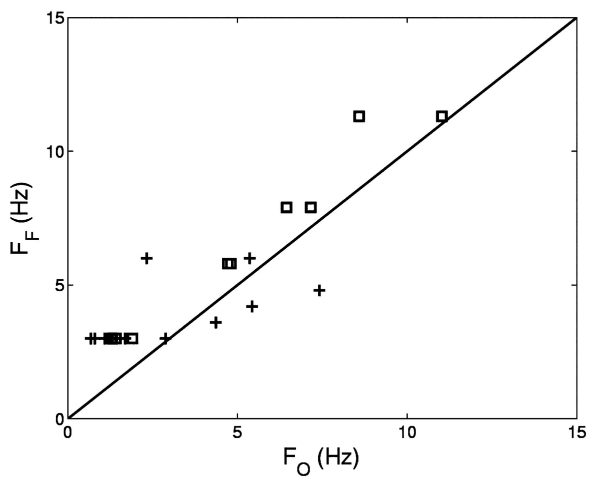

Figure 14 Comparison between observed fracture frequency (F_O) and peak in frequency domain (F_F) for square (□) and slot (+) dies. Grey line: line of equivalence. Glucose paste, various *L*/*D* and velocity values. The square dies show a stronger correlation.

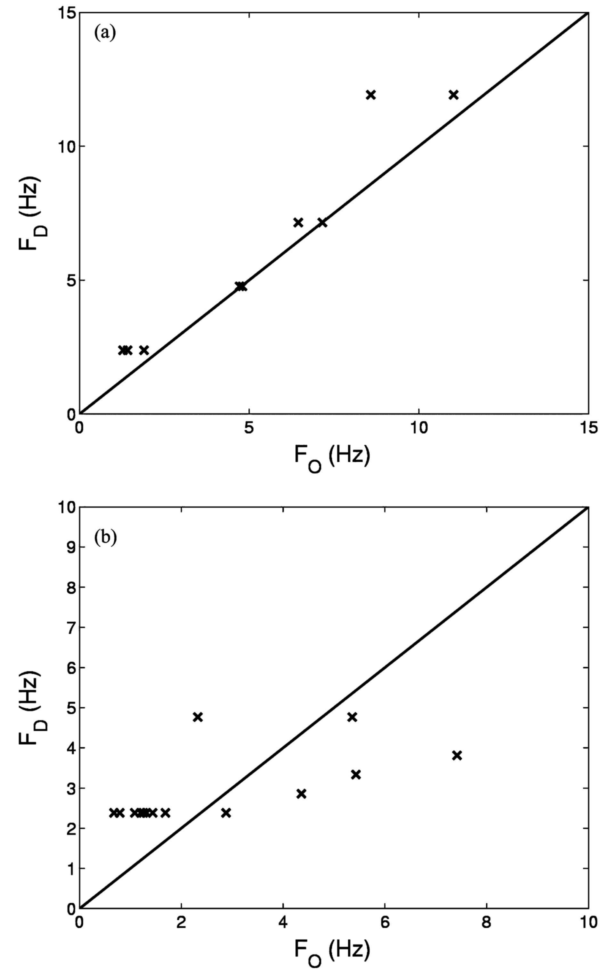

Figure 15 Comparison between observed frequency of fracture (F_O) and defect frequency (F_D) calculated using hydraulic diameter as characteristic length scale for ram extrusion with (*a*) square dies (5.5 × 5.5 mm) and (b) slot dies (15 × 2 mm).

where the corners were closest, and would frequently vary in nature between the different sides. The variability in fracture periodicity resulted in considerable differences between F_F and F_O values for slot dies, particularly at low frequencies (and extrusion rates). This variation is reflected in Fig. 14: there were distinctly different trends for square and slot dies.

The use of Teflon inserts in the square and slot dies again reduced the extent of fracture. With square dies, circumferential cracking was usually eliminated, and fracture restricted to a small region next to the corner (if any). With the slot dies, the extent of fronding was less severe and often more regular.

Calculation of the 'defect frequency' requires knowledge of the appropriate length scale for the periodicity in these systems. One candidate is the hydraulic diameter, d_H, given by $4 \times (A_D)/C$, possibly justified by GCF being the result of a flow instability. Fig. 15 shows that d_H gave a good correlation for the square (and round) dies, whereas the slot dies did not show any systematic trend.

5. Conclusions

The phenomenon of gross circumferential fracture in the ram extrusion of soft-solid pastes has been studied using a number of different paste materials across a range of operating parameters known to give rise to fracture in previous studies. When fracture did occur, the effects of these operating parameters were broadly in line with previously reported results, in particular that short die lands and high wall shear stresses promote fracture. For square entry dies with circular and square die land cross sections, the periodicity of the fracture planes exhibited a simple relationship to the geometry, as reported by Domanti and Bridgwater.

Insight into the mechanisms underlying GCF was provided by interrogation of the pressure signal measured at the die in the frequency domain. Short Time Fourier Transform techniques confirmed that the observed fracture periodicity was accompanied by a distinct peak in the power spectrum at the frequency corresponding to the observed fracture frequency. False negatives were not observed unless the signal to noise ratio was too small, such that the noise masked the periodic component. A key feature was that periodic components were not observed if fracture did not occur anywhere during an extrusion test, but would continue to be detectable after fracture had occurred, often in the initial states, but whenever GCF was no longer visible. These effects, which were observed for several pastes, indicate that this form of ram extrusion is susceptible to a flow instability which will generate GCF if the combination of shear stresses and material cohesion (or other factor analogous to a fracture toughness) are sufficient to induce cracking. The magnitude of the periodic component was small compared to the stresses imposed on the paste and exhibited a small frictional dependence.

Extrusion through non-circular dies was subject to fracture initiated at the stress-concentration corners, such that GCF was usually observed throughout an experiment. The observed periodicity was again observed in the frequency domain, and the length scale appropriate for the geometry criterion was found to be the hydraulic diameter for square dies. A wide variation in fracture frequency was observed with slot dies, including gross peeling and different periodicities for different sides, presumably due to the separation of the corner zones. For cases of regular fracture, the appropriate length scale for the geometric criterion was found to be the hydraulic diameter.

The use of a lubricating die land surface, namely Teflon, both decreased the wall shear stress in the die land and reduced, and sometimes eliminated, the extent of fracture. Teflon surfaces were not, however, able to suppress fracture initiation at corners.

Nomenclature

Roman

A_{B} Cross sectional area of barrel ($\mathrm{m}^{2)}$)
A_{D} Cross sectional area of die land (m^2)
A_{F} amplitude of periodic stress component (Pa)
C Perimeter (m)
D Die land diameter (m)
D_{B} Barrel diameter (m)
d_{H} Hydraulic diameter (m)
F_{D} Defect frequency (Hz)
F_{F} Frequency of periodic stress component in frequency domain (Hz)
F_{O} Observed fracture frequency (Hz)
L Die land length (m)
m Die entry yield stress velocity index
n Die land wall stress velocity index
N Length of data sub set
P Mean extrusion pressure (Pa)
Ra Surface roughness (μm)
t time (s)
V Extrudate velocity (m s^{-1})
V_{R} Ram velocity (m s^{-1})

Greek

α Die entry yield stress velocity factor (Pa (s m^{-1})$^{\mathrm{m}}$)
β Die land wall stress velocity factor (Pa (s m^{-1})$^{\mathrm{n}}$)
$\dot{\gamma}_a$ Apparent wall shear rate ($= 8\times V_{\mathrm{L}}/D$) (s^{-1})
Φ Spectral density (Pa)
λ_{p} Fracture period (m)
σ_0 Die entry yield stress (Pa)
τ_0 Die land wall yield stress (Pa)
τ_{eff} Effective die land wall shear stress (Pa)
θ Angle of fracture ($^\circ$)

Appendix: Benbow-bridgwater characterisation

The rheological behaviour of the pastes undergoing extrusion was characterised by capillary rheometry following the methodology described by Benbow and Bridgwater [10]. In this approach, a series of ram extrusion tests is performed through axisymmetric circular dies over a range of different die land L/D ratios and mean die land velocity V, and the force on the ram required to extrude the paste is presented as the average 'extrusion pressure', P. The data are then fitted to a 6-parameter equation, *viz.*:

$$P = 2\left(\sigma_0 + \alpha V^m\right)\ln\left(\frac{D_0}{D}\right) + 4\left(\tau_0 + \beta V^n\right)\left(\frac{L}{D}\right) \tag{A.1}$$

In this model the dominant contributions are attributed to plastic extension in the die entry (characterised by σ_0, α and m) and wall slip in the die land (characterised by the parameters τ_0, β and n). A detailed critique of this approach can be found in Blackburn *et al*. [24]. For non-circular axi-symmetric dies as employed here, Benbow and Bridgwater proposed a model for the extrusion pressure, P, in terms of the cross-sectional area of the barrel, A_{B}, die land, A_{D} and die land perimeter C of the form:

$$P = \left(\sigma_0 + \alpha V^m\right)\ln\left(\frac{A_B}{A_D}\right) + 4\left(\tau_0 + \beta V^n\right)\left(\frac{L\cdot C}{A_D}\right) \tag{A2}$$

Fig. A1 shows that this model gave reasonable agreement for extrusion of the glucose paste through the non-circular brass dies described in Table V. The paste parameters are reported in Table I and were obtained by characterisation tests in axisymmetric circular die lands with $2 < L/D < 16$ [13].

Acknowledgments

This work was funded as part of EPSRC project GR/N 24780. The starch materials and detergent paste were generously provided by United Biscuits R&D, High Wycombe, and Procter & Gamble Product Technology Centre, Newcastle-upon-Tyne, respectively. Support for BDR from the Cambridge Commonwealth Trust is much

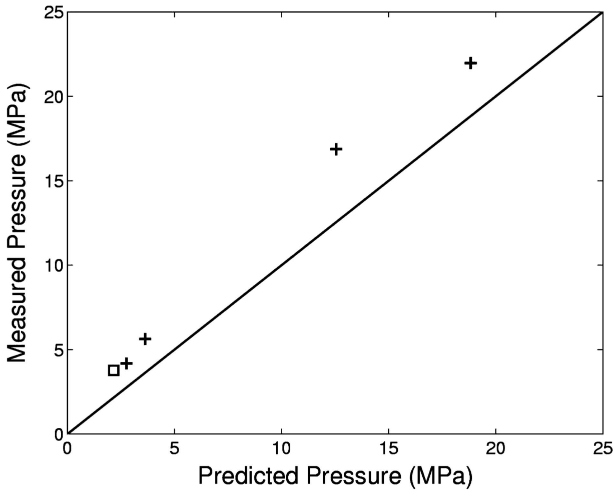

Figure A1 Comparison of measured and predicted extrusion pressure (Equation A.2) for glucose paste undergoing extrusion through brass slot (+) and square (□) dies. Grey line shows line of equivalence.

appreciated. Helpful discussions with Dr. Joan Lasenby and Prof. John Bridgwater are gratefully acknowledged, as is the work on the glucose paste performed by Sam Sanders-Hewett and Dyfrig Davies.

References

1. P. J. HARRISON, J. M. NEWTON and R. C. ROWE, *J. Pharm. Pharmacol*. **37** (1985) 81.
2. A. T. J. DOMANTI and J. BRIDGWATER, *Chem. Enging. Res. Des*. **78(A)** (2000) 68.
3. A. T. J. DOMANTI, D. J. HORROBIN and J. BRIDGWATER, *Int. J. Mech. Sci*. **44** (2002) 1381.
4. H. LL. D. PUGH and A. H. LOW, *J. Inst. Met*. **93** (1964) 201.
5. G. SORNBERGER and J. C. QUANTIN, *J. Non-Newtonian Fluid Mech*. **23** (1987) 123.
6. Y. W. INN, R. J. FISHER and M. T. SHAW, *Rheologica. Acta* **37** (1998) 6.
7. R. P. G. RUTGERS and M. R. MACKLEY, *J. Non-N. Fluid. Mech*. **98** (2-3), (2001) 185.
8. O. L. KULIKOV and K. HORNUNG, *J. Non-Newtonian Fluid Mech*. **98** (2001) 107.
9. R. A. DIMILIA and J. S. REED, *J. Amer. Ceram. Soc*. **66**(9) (1983) 667.
10. J. J. BENBOW and J. BRIDGWATER, in "Paste Flow and Extrusion" (Clarendon Press, Oxford, 1993).
11. A. D. U. S. AMARASINGHE and D. I. WILSON, *Chem. Enging. Res. Des*. **76(A)** (1998) 3.
12. Y. CHEN, A. S. BURBIDGE and J. BRIDGWATER, *J. Amer. Ceram. Soc*. **80** (7) (1998) 1841.
13. S. P. T. SANDERS-HEWETT and D. J. DAVIES, Part IIB Research Project Reports, Department of Chemical Engineering, University of Cambridge, 2003.
14. A. CHEYNE, J. BARNES and D. I. WILSON, *J. Food Enging*. **66**, 1.
15. O. L. KULIKOV and K. HORNUNG, *J. Non-N. Fluid Mech*. **107** (2002) 133.
16. G. Y. ONODA and L. L. HENCH, in "Ceramic Processing Before Firing" (John Wiley & Sons, NY, 1978).
17. A. D. U. S.Amarasinghe, Interpretation of paste extrusion data, PhD Dissertation, University of Cambridge, 1998.
18. Mathworks, Matlab Version 5.3, 1999.
19. S. MALLAT, in "A Wavelet Tour of Signal Processing" (Academic Press, 1998).
20. B. D. RUSSELL, S. BLACKBURN, D. I. WILSON and J. LASENBY, Application of On-Line Signal Processing to Paste Extrusion Data, submitted to IEEE Trans. *Ind. App*., 2004.
21. G. L. BRETTHORST, in "Bayesian Spectrum Analysis and Parameter Estimation" (Springer-Verlag, 1989).
22. J. J. K., Ó RUANAIDH and W.F. FITZGERALD, in "Numerical Bayesian Methods Applied to Signal Processing" (Springer, 1996).
23. J. P., BENTHAM and W. T KOITER, Asymptotic Approximations to Crack Problems, in "Methods of analysis and solutions of crack problems", edited by G.C. Sih, (Noordhoff International Publishing, Leyden, 1973).
24. S. BLACKBURN, A. S. BURBIDGE and H. MILLS, A Critical Assessment of the Benbow Approach to Describing the Extrusion of Highly Concentrated Particulate Suspensions and Pastes, Proc. XIIIth International Congress on Rheology, Cambridge, Paper 4-139-141, 2000.

Received 8 June 2004
and accepted 7 July 2005