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Maldistribution of fluids in extrudates

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Abstract

Solid–liquid pastes featuring high volume fractions of particulates are frequently used in ceramic forming operations. When pastes are used it is important that the particulate distribution remains uniform throughout the body. The stresses imposed during extrusion processing can, however, promote differential flow between the solid and liquid phases giving rise to product and processing problems. Reliable models for predicting phase distribution changes in these multi-phase systems are in their infancy.

This paper reports progress towards developing simulation techniques and practical systems to verify the numerical approaches. Pastes containing glass spheres suspended in a highly viscous Newtonian fluid have been extruded at various speeds and solids loadings. Load and liquid content data are presented which form the basis for model verification. Soil mechanics approaches are used here to encapsulate the inherently multi-phase nature of these systems. The modified Cam–Clay model has been implemented in a finite element analysis simulation of ram extrusion using the ABAQUS platform. The simulation requires regular and extensive remeshing and monitoring of the conservation of mass. Predictions of extrusion pressures and deformation behaviour are compared with the experimental data for a series of square-ended and conical dies.

Keywords: Extrusion; Soil mechanics; Phase migration; Paste; Modelling

1. Introduction

Particulate pastes are widely used in many fields, not least ceramics for extrusion and injection moulding or as intermediates in other processes such as slip casting. They can be considered to be two-phase systems comprising solid particulates and a liquid matrix. For continuous and defect-free processing these two phases should not separate during flow. Phase segregation or migration leading to maldistribution will generate density variation in the finished product and in extreme cases the process of extrusion or moulding may stop. Phase migration is more normally recognised during processing by excess liquid being observed at the die face or on the extrudate surface.

There have been a number of reports in the literature describing the phenomena.[1,2] These studies and others[3] have attempted to predict the behaviour, considering the influence of die geometry, composition and other process variables on the onset and magnitude of phase migration. Generally it is accepted that phase migration is manifest when the relative velocity of the liquid phase to the solid phase is high, and that this is induced by local pressure gradients and the permeability of the solid skeleton. The existing models are either one-dimensional, geometry-specific or not capable of fully reflecting all aspects of the material's constitutive law. Two-phase models that fully incorporate the characteristics of pastes are well established in the field of soil mechanics and are commonly used at small strains. In this paper, these geotechnical approaches are implemented in the large strain environment of extrusion and the simulation results are compared with experimental data obtained from a model material of similar structure undergoing the same deformation.

2. Experimental pastes

Simplified pastes were prepared with regular solid and liquid phase characteristics that allowed their behaviour to be modelled reasonably reliably. These pastes comprised tightly sized glass spheres with a mean particle size of 190 μm (Omya UK Ltd., UK) mixed with a Newtonian fluid with a viscosity of 300 Pa s at 20 °C (glucose, Lloyds Chemist, UK). These two materials were blended together using a Z-blade mixer (Morton, UK) for 30 min. Pastes were prepared with nominal solid loadings of 64.2, 68.7 and 72.8 wt% solids (50, 55 and 60% by vol. assuming

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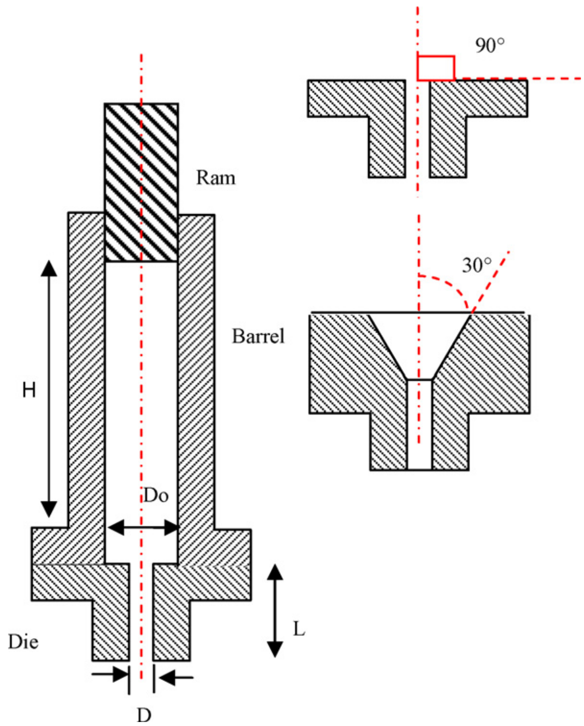

Fig. 1. Geometries of the experimental system. Symbols and numbers in text. Only 90° and 30° angles shown.

saturation): levels in each batch were measured before extrusion as the viscosity of the fluid prevented precise make up. The paste was extruded from a ram extruder with a barrel diameter, D_0, of 25 mm and a die diameter, D, of 3 mm and length, L, to die diameter ratio of 8. The barrel was filled to a height, H, of 120 mm and 100 mm extruded (where possible). The entry angle to the die was 90°, 60°, 45°, or 30° as shown in Fig. 1. The assembly was conditioned at 20 °C and 40% RH for 1 h, prior to extrusion. Extrudates were formed at ram velocities of 1, 2, 5, 10, and 20 mm min^{-1}. For the 90°, 60°, and 45° entry dies only the intermediate solids loading was investigated. During each run, samples of paste were taken from the feed, of the extrudate at 5 mm ram displacements, of any extrudate emerging once ram motion ceased, and of the paste plug remaining in the barrel. Liquid phase content was measured by thermal removal of the glucose at 500 °C. From the data collected, plots of load (N) and solids content (%) as a function of barrel displacement (mm) could be constructed.

3. Experimental observations

Typical plots of pressure–displacement are shown in Fig. 2 for the high solids loading paste. It can be seen from these profiles that as the paste is first compacted, there is a steep rise in the load. Once extrusion starts the load rises less rapidly. In non-phase separating pastes no pressure rise with displacement was observed during this stage of the process but in phase migrating pastes the pressure generally rises with displacement. If the load required for extrusion remains below the upper limit for the extruder (30 kN) then a further rapid rise in extrusion load is observed as the final material is extruded. This is attributed to the

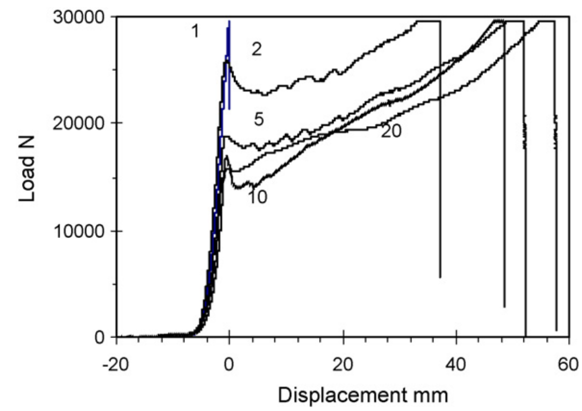

Fig. 2. Experimental load–displacement curves for high solids loading paste passing through 90° die. Numbers are ram speed in mm min^{-1}.

change in flow pattern when the ram is close to the die and the removal of static material which may accumulate in the corners of steep angled die entries. In these pastes, when there was no apparent phase migration in terms of solids content variation, the load required for extrusion increased with increasing ram speed. This was observed when the ram speed was fast and/or the solid loading low. In Fig. 2 the load is observed to rise with reducing ram speed and to exceed the extruder capacity at progressively shorter ram displacements. This was most extreme at 1 mm min^{-1}, when there was virtually no extrusion before the load limit was reached. The load curves can be directly related to the solids loading curves shown in Fig. 3. It can be seen that as the velocity of extrusion is reduced the initial concentration of solids in the extrudate is reduced. The solids loading generally rises as extrusion continues. The gradient of the rise is generally reduced with increasing ram velocity. This is indicative of the paste's propensity to phase migration.

The load–displacement and solids loading–displacement plots for the conical entry dies for the paste with intermediate solids loading are shown in Figs. 4 and 5 respectively. Capillary flow models would suggest that there would be a slight rise in the load required for extrusion with decreasing entry angle. The results show that this may be the case in the early stages of flow in the system being examined here. There is no significant variation in the slope of the load curves from the commencement of extrusion to the static zone removal region. Following the

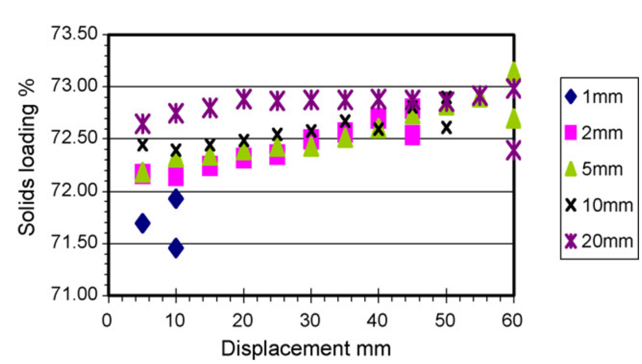

Fig. 3. Experimental solids loading–displacement curves for high solids loading paste passing through 90° die. Numbers are ram speed in mm min^{-1}.

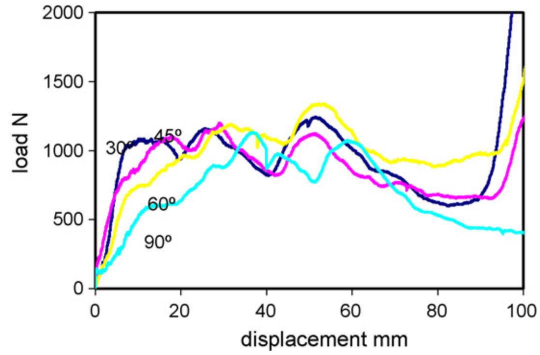

Fig. 4. Experimental load–displacement curves for medium solids loading paste passing through 90°, 60°, 45° and 30° dies. 10 mm min^{-1}.

arguments presented for the square entry die this suggests that there was only slight evidence of phase migration under these conditions. The solids loading data appear to confirm this where there is no significant change in slope in the curves recorded for the different dies.

4. Model development

The modified Cam–Clay soil mechanics constitutive law has been used in the ABAQUS Lagrangian solver to simulate the paste (axisymmetrically) undergoing ram extrusion at a fixed ram velocity. The liquid is Newtonian and Darcy's law is used to represent the interaction between the two phases. The Carman–Kozeny relationship is used to estimate the permeability of the solids skeleton. There is some concern in the literature[4–6] that with non-uniform particles its predictions may not be valid. As the model assumes spherical particles and the pastes feature ballotini then the unmodified relationship is applied, at least as a first estimate.

Key assumptions in the model are: gravitational and inertial forces are neglected; isothermal conditions; the particles and liquid are assumed to be incompressible; the paste is assumed to be saturated (no entrained air). The total stress on the paste at any location, e.g. at the ram, is supported by both the pore liquid (as pore pressure) and the solids matrix (termed effective stress). This is known as Terzaghi's principle.[7]

A detailed derivation of the Cam–Clay yield surface is given elsewhere.[8] Under an increasing isotropic (effective) stress, the particulate matrix compacts elastically and then plastically. During elastic or plastic shear, the material may dilate, compact, or shear at constant volume (critical state); this is an important feature of highly filled granular materials and its extent depends on the internal angle of friction of the particles, ϕ, and the Poisson's ratio of the solid skeleton, υ. These parameters were taken from the literature, although a triaxial testing program is currently underway to obtain them. The values used here were 0.01 and 0.05 for the elastic (κ) and plastic (λ) bulk moduli, respectively, 45° for ϕ and 0.49 for υ. The initial, pre-extrusion isotropic (effective) stress in the paste, was due to surface tension effects at the surface of the paste that was exposed to atmosphere. The initial crushing stress of particulate matrix was assumed to be twice this value, giving the initial value of the overconsolidation ratio as 2. This is likely to be too low, although gives an initially equal balance of compaction and dilation during shear (critical state). The von Mises yield criterion and associated flow are assumed for this material. The friction condition at the extruder wall was unknown. As pastes have a strong tendency for wall slip, a smooth wall was assumed in all simulations as a first approximation.

The geometry is modelled using 800 quadrilateral elements. Adaptive remeshing (using existing code[9] and methodologies[10] as a basis) was employed to overcome the high mesh distortion as the extrusion process continues, using the L2 error in effective pressure stress as a guide for mesh construction. The use of adaptive remeshing in soil mechanics simulations has been extremely limited thus far due to difficulties in implementation[11] and was a major issue, as it had to be performed ∼2000 times for the simulations described here, mostly due to distortion in the elements flowing around the die entry corner. As a result, the model absorbs much processor time and the provisional simulation results were attained for a shorter barrel length ($H/D_0 = 1.5$), die-land length ($L/D = 4.2$) and contraction ($D/D_0 = 0.2$) than used in the experimental programme. The less-severe contraction resulted in decreased element distortion at the die entry corner, and therefore the frequency of remeshing was reduced.

During the simulation the average volume fractions of liquid and solid were monitored in order to ensure conservation of mass. This was particularly important for the liquid phase as after several time steps a systematic error in the liquid volume fraction was observed and a correction algorithm was required. The extrudate was periodically cut within the model and the average solids loading calculated. These results, and the load sustained at the ram, can be directly related to the experimental data.

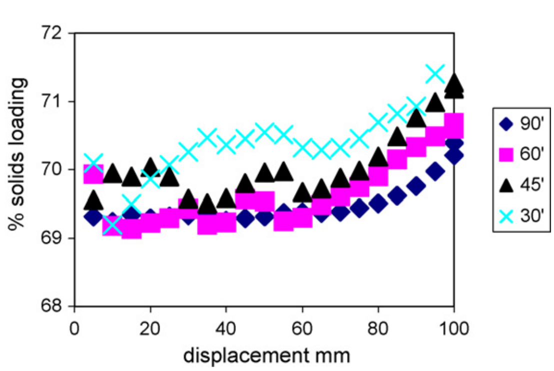

Fig. 5. Experimental solids loading–displacement curves for medium solids loading paste passing through 90°, 60°, 45° and 30° dies.

5. Model predictions

The extrusion load predicted by the model rises more rapidly than the experimental system for the 90° entry angle (Fig. 6A), but this is to be expected as the experimental paste is degassed (and compacted) during the initial ram displacements, whereas the model assumes an initially air-free (saturated) state. The model predicts a slower rise in pressure for the other entry angles, and this is because simulations in conical entry geometries start with an unfilled cone, giving a slow increase in predicted extru-

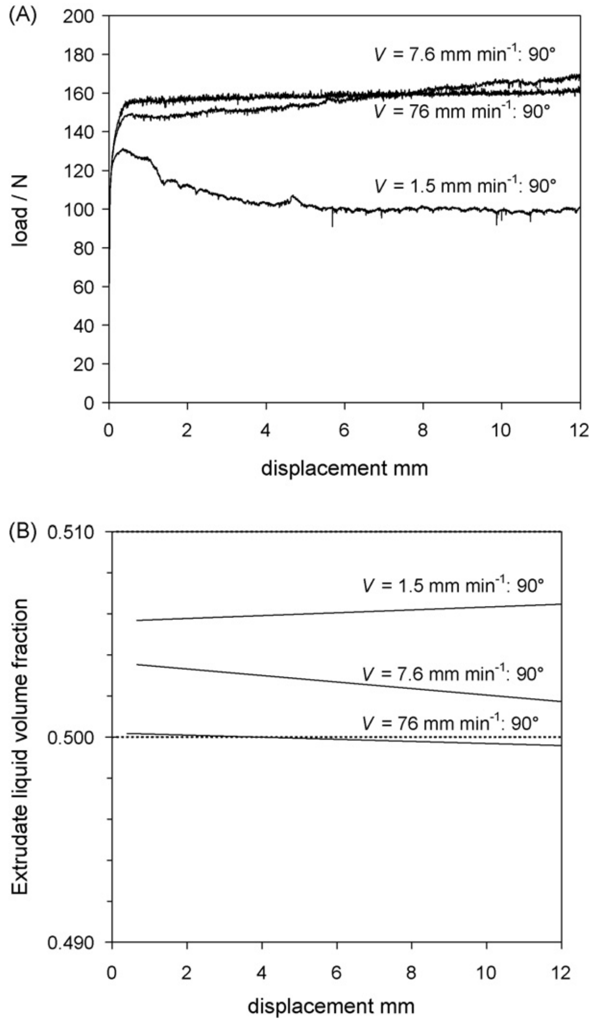

Fig. 6. (A) Simulated extrusion load profile for medium solids loading. (B) Simulated phase volume profile for medium solids loading (dashed line indicates starting liquid phase volume fraction in paste). All through 90° entry die and different flow rates.

sion load as it gradually fills. In the experimental system this filling process is masked by the pre-extrusion process of filling the barrel and the slower general pressure build up as the paste is de-aired. At a 90° die entry angle there is a change in pressure profile with ram velocity. At the highest ram speed, there is no change in extrusion load with displacement, suggesting no phase migration. This is consistent with experimental observations. At the intermediate velocity there is a slight rise in pressure with displacement, again as observed experimentally. The load falls at the lowest ram speed and this is attributed to initial dilatancy in, and significant softening of, the entry region drawing fluid away from the ram area and into the earlier samples of extrudate. Thus the material no longer acts as a perfect plastic on the macro scale and shows behaviour similar to that seen in the experiment with reduced loads being required at low ram speeds. Similar behaviour is predicted for the conical entry dies, in Fig. 7A. It should be noted that if there were no phase migration the load profile from the model would be the same for all speeds as the paste is modelled as a perfect plastic. We have not demonstrated that the experimental paste used here is a perfect plastic and would more generally classify its behaviour as pseudo-plastic but this may be an artefact of phase migration as predicted by the model.

The predicted solid loadings suggest that there would be a general decrease in solids content of the first formed extrudates as ram speed is reduced for the 90° entry die, Fig. 6B. This is reflected in the experimental data. Further the model also predicts that there would be virtually no phase migration detected in the conical entry dies over and above that found in the 90° data, Fig. 7B. This again is consistent with the experimental data.

There are however some issues which remain to be resolved between the model and the experimental data. The remeshing process was initially a complex issue and restricted the reduction ratio and billet volume, i.e. H. We believe these to be close to resolution. At the time of writing even though the general trends are similar the model parameters were taken from the literature and this clearly leads to significant magnitude differences between the model and experiment. The greatest discrepancy currently comes with respect to the load prediction. This is strongly affected by the (overly low) value used for the initial

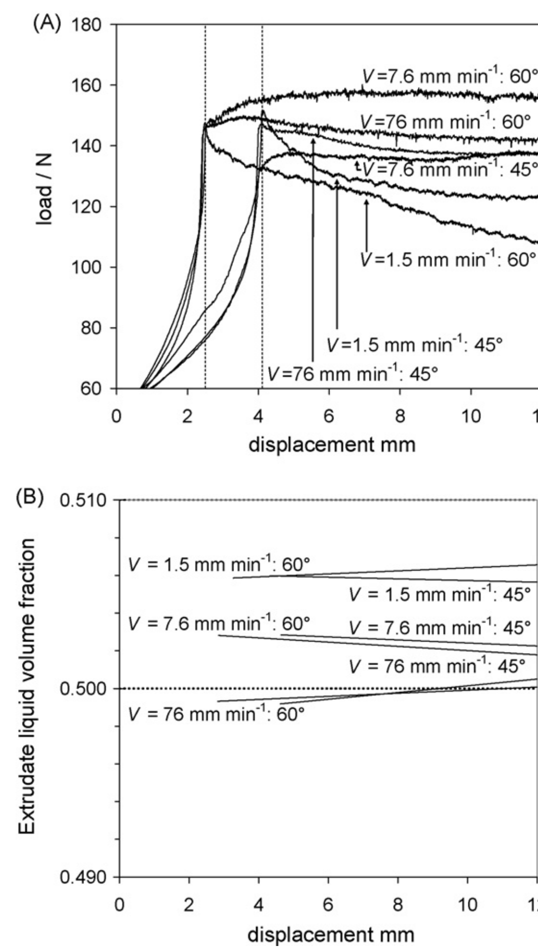

Fig. 7. (A) Simulated extrusion load profile for medium solids loading (dashed lines indicate when conical die entry zone is just full). (B) Simulated phase volume profile for medium solids loading (dashed line indicates starting liquid phase volume fraction in paste). All through 60° and 45° entry die and different flow rates.

crushing strength of the particle matrix; this positively correlates with the initial extrusion pressure from the model. Triaxial cell work continues to measure the required material parameters to rectify this, and experiments to measure the likely amount of wall friction are being undertaken.

Also, some of the predicted changes in solids loading are extremely fine and it is difficult to attain such accuracy in the experimental system, which mask these subtleties.

Given that the model contains estimates of the required parameters, it clearly predicts behavioural trends similar to those seem for the experimental material.

6. Conclusion

The existence of phase migration in highly loaded suspensions has been verified and quantified experimentally. Models based on a soil mechanics approach to two-phase systems have been developed which predict phase migration during flow from a ram extruder and shows similar trends to the experimental data. Refinement of the model and the required input data should improve the predictive capacity of the model. Phase migration is shown by both model and experiment to be dependant on the speed of extrusion and the formulation and to a lesser extent on the geometry.

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