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Backlash: a feature of the cyclic deformation of ceramic pastes

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Abstract

This paper offers a mathematical model to explain the rather peculiar results obtained in the cyclic tension–compression tests on various stiff ceramic pastes. The experimental results indicate a material with low strength in the middle of the cycle, rapidly strengthening at the extremes. Additionally there is a pronounced Bauschinger effect on strain reversal. The mathematical model is based on a previously published model for dry granular materials. This was specifically designed to simulate cyclic deformation, but a modification to include capillary tension allows excellent agreement between the simulation and experimental results on pastes. This demonstrates that the mechanics of granular materials is the appropriate basis for modelling these and similar materials rather than modified polymer rheology. © 2000 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Mixes produced for plastic forming can be thought of as stiff pastes consisting of ceramic particles and a liquid. Such pastes commonly contain clay, oil or aqueous polymer solutions to give them plasticity. It is important for these mixes to bestow satisfactory properties to the fired ceramic whilst maintaining formability in the green state. In order to achieve this, compromises need to be made in the formulation and these should be based upon a sound understanding of the nature of paste plasticity.

The origin of paste plasticity in green ceramics has been considered previously. For example, Norton[1] explained plasticity in terms of capillary tension. This view, while supported in the area of soil physics, has found little support in the ceramics literature until recently.[2,3] Unfortunately the relationship between processability and mix design still remains somewhat obscure and deserves closer study. An initial goal for such a study would be to determine whether the properties of the binder liquid or granular behaviour control the mechanical properties. For instance, if an aqueous polymer solution is used, is the resistance to deformation at a low level of stress due to gelling of the polymer or, as Norton's work suggested, friction between the ceramic particles pulled together by surface tension?

Different types of mechanical test support various models for pastes. Many tests use existing capillary or torsional rheometers and so concentrate on the determination of the rate dependence of the resistance to deformation rather than the strain dependence. Looking at these results alone, it might be argued that the solids just 'thicken' the polymer solution as in pastes with a lower solids concentration.[4] Others, for example Adams et al.,[5] have investigated the strain as well as strain rate dependence using compression tests. They fitted their results to expressions previously used to model the plastic forming of metals and obtained a reasonable description of the paste during monotonic loading. A more complete survey of the testing of pastes is contained in Chandler et al.[6] The phenomenon of backlash, however, will be seen to uncover the particularly complex nature of stiff pastes and expose the principally granular nature of these materials.

Backlash is a peculiar phenomenon seen in the cyclic deformation of stiff ceramic pastes. It is best described in Macey's[7] own words:

> If a portion of clay in the plastic state be held in the thumbs and fingers of both hands and alternately

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> pulled and compressed, a looseness can be felt. There is a range of easy movement culminating in each direction in a sudden resistance. The sensation is reminiscent of play in a joint or of the behaviour of a nut on a worn thread, from which analogy suggests the name of 'backlash'.

Many readers will have noticed this when working with clay, but it seems to be largely ignored in the literature. This may well be because it would appear to be of little direct use and sometimes even a nuisance. The only other connected work was concerned with the cyclic twisting of extruded bars[8] which will be discussed in the next section. Backlash does have some use, however, in that it provides an extreme test for any model of paste behaviour.

2. Experimental background

Cyclic testing of stiff ceramic pastes has received little attention in the literature. However the work which is reported highlights some very interesting phenomena.

Quantitative experiments were performed by Macey.[7] Extruded bars of various clay-based pastes were deformed in cyclic tension-compression. The absolute value of the strain rate was kept constant but the sign changed when the force limit was reached. Typical results after a few cycles of this deformation are shown in normalised form in Fig. 1A. Although, as can be detected qualitatively, the material is more resistant to deformation at the extremes of the cycle than it is in the middle, other important observations come to light:

i. there is a marked Bauschinger effect.[9] That is, yield occurs sooner on reversal than one would expect from perfect plasticity.
ii. The curve is not symmetric between tension and compression.
iii. There is a drift towards extension over successive cycles.

The last two effects were not present in cyclic torsion tests performed by Astbury and Moore.[8] These experiments were also on extruded bars of clay-based pastes, which were strained sinusoidally. The other important features were, however, reproduced (see Fig. 1B). It transpires that the tension–compression asymmetry is an important feature so recently the authors performed slightly modified tension-compression tests. These were again on extruded bars but on this occasion:

- The bars were not made from clay-based pastes but from well mixed pastes consisting of A17 alumina (manufactured by Alcoa) and aqueous-based binder (see Table 1).The test was arranged so that the displacement rate of the cross-head followed a square wave in time with equal amounts of extension and compressive deformation within each cycle.
- The test started with small amplitude displacements which slowly rose to a set level. This procedure eliminated the bias effects of initiating the full deformation with compression rather than tension (or vice versa).
- During the test the length of the gauge-length was measured using a laser extensometer. This prevented the compliance of the gripping system from introducing error into the results.
- A novel feature of this test was the gripping technique. Springs were carefully screwed into the ends of the cylindrical sample. These areas were allowed to dry while the surface of the central portion was kept moist.

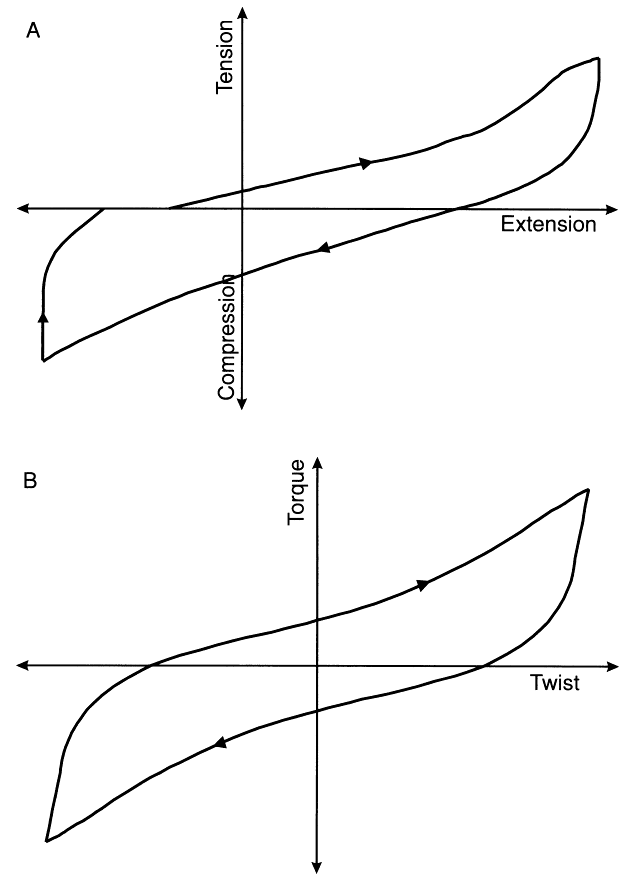

Fig. 1. (A) One loop of the results of Macey's tension–compression tests on clay; (B) the steady-state results of Astbury and Moore's torsion tests on earthenware redrawn on conventional axes.

A stress–strain loop after a few cycles at a steady amplitude is shown in Fig. 2A. The important features seen in the clay–water pastes are reproduced. On this occasion the limits of displacement were controlled and although tension-compression asymmetry can be seen there was no continuing drift.

Table 1
The composition of our alumina paste used to give the results shown in Fig. 2A

Material	Volume fraction
A17 alumina	0.67
Aqueous PVA solution	0.33

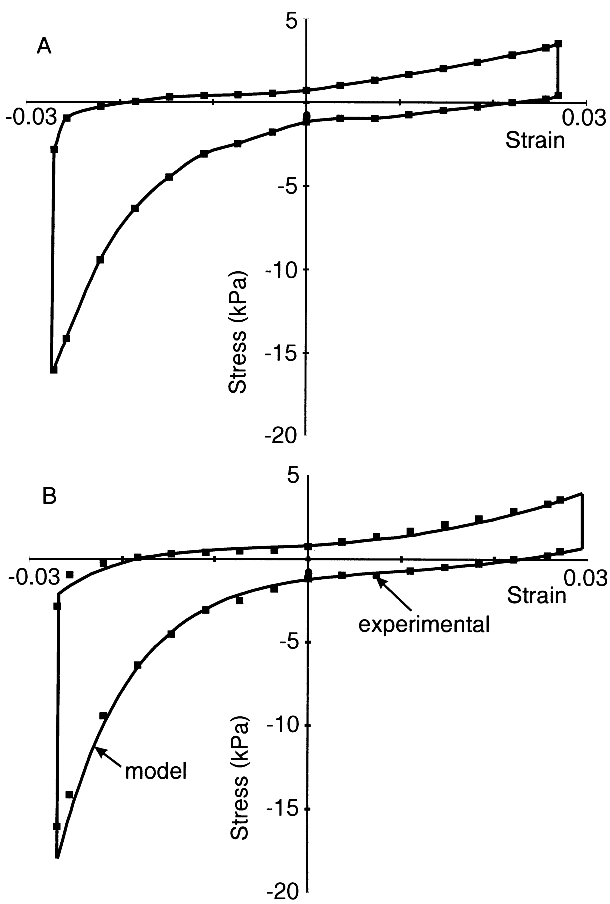

Fig. 2. (A) A stress–strain loop from cyclic tension–compression tests on alumina paste; (B) stress–strain loop from the mathematical model laid on top of the experimental data points for alumina.

3. Granular model

3.1. Basic ideas

What can be causing this peculiar behaviour? Macey[7] had some ideas but they do not form a complete theory. Bolton,[10] however, suggested that backlash might be explained by bringing together three previously observed phenomena.

Firstly, the deformation of an assembly of hard dry granules can involve significant volume changes as the granules either roll over or slide past each other. If a sample of granular material is subject to repeated cycles of small deformation about a mean position the granules progressively seem to find a more compact arrangement.[11] What is particularly important to the phenomena of backlash is that after a moderate number of cycles at small strain levels the volume appears to be related to the shear strain in a parabolic fashion.[12] The volume is a minimum at the centre of the cycle and increases approximately in proportion to the square of the deviation from the central position. This is very nearly reversible as long as the strain is not too large. If the strain is large and monotonic the dense packing can be disrupted producing a significant overall volume increase. Reynolds coined the term 'dilatancy' to describe this.[13] Only at very large monotonic strains (>20%) is the dilatancy exhausted after which deformation continues at the so-called critical state.[14]

Secondly, when a sample of paste is surrounded by air, the curvature of the menisci at the air–liquid interface induces liquid tension that pulls the granules together.[15] This curvature is increased if a small amount of fluid is removed and consequently the granules are pulled together more tightly, increasing the compressive forces at the contacts. However the dilatancy induced by particle rearrangement has the same effect, as it creates more internal voids to be filled with a constant volume of fluid. The presence of isolated air voids[16] in many commercially produced pastes will alleviate the very rapid rise in contact forces as these bubbles can expand accommodating some of the increase in pore volume.

Thirdly, the higher the compressive contact forces between the granules the higher the frictional resistance of the granular assembly to shear deformation. This assumes the granules are effectively in contact with their nearest neighbours.

Backlash can now be explained as follows. We argue that after a few cycles the particles have improved their packing in the central portion of the cycle and the parabolic relationship between volume and shear has been established. There will then be an effective surplus of fluid at the mean position and an effective deficiency of fluid at the extremes of the cycle. The fluid surplus produces little resistance to deformation in the central regions of the cycle and the fluid deficiency gives the strengthening at the extremes.

The remainder of this section develops these simple ideas in the form of a mathematical model but no new physical concepts are introduced.

3.2. Mathematical preliminaries

Consider a volume of paste lying in a Cartesian coordinate system x_i with i taking values 1 to 3. It is deformed such that the instantaneous velocity vectors of a small region of the material have components υ_i and result in the components of the small displacement vector u_i. The components of the rate of strain tensor e_{ij} are given by

$$\dot{e}_{ij} = \frac{1}{2}\left(\frac{\partial \upsilon_i}{\partial x_j} + \frac{\partial \upsilon_j}{\partial x_i}\right) \tag{1}$$

and the components of the strain tensor e_{ij} are

$$e_{ij} = \frac{1}{2}\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right). \tag{2}$$

The components of the stress tensor needed to produce this deformation are σ_{ij}.

It is useful to split each of these tensors into two parts:[17] the strain rate and strain tensors can be split into volumetric and deviatoric parts. The volumetric parts are defined, using the summation convention, by $\dot{e} = \dot{e}_{kk}$ and $e = e_{kk}$, respectively. The components of the deviatoric part of the tensors can then be defined as

$$\dot{d}_{ij} = \dot{e}_{ij} - \frac{1}{3}\dot{e}\delta_{ij};\ d_{ij} = e_{ij} - \frac{1}{3}e\delta_{ij}, \tag{3}$$

where δ_{ij} is the Kronecker delta and takes the value of unity when $i = j$ and zero otherwise. Similarly for the stress tensor, the mean stress is defined as $\sigma = \sigma_{kk}/3$ and the components of the deviatoric stress tensor as

$$s_{ij} = \sigma_{ij} - \sigma\delta_{ij}. \tag{4}$$

3.3. Dry granular model

Dry granular materials can be modelled within the context of plasticity. They have a yield surface, which is pressure dependent, and deformation is essentially rate independent. At present the state of modelling is such that one needs separate constitutive models for large strain monotonic loading and small strain cyclic loading. The model presented here was developed recently to simulate the later and is appropriate to granular materials that have previously suffered small levels of cyclic strain so that a dense packing is produced. The approach is to start with:

- a rule linking volume strain to shear deformation;
- an expression for the rate at which energy is dissipated by rate independent plastic deformation;
- and the assumption that all the work done on the material is dissipated.

Once these are established then flow rules and yield surfaces can be derived mathematically[18] using straightforward procedures involving the mathematical theory of envelopes. The details of the model used in this paper are published elsewhere,[19] but the important aspects rely on the following detailed assumptions:

i. the initial dense state is isotropic which allows scalar variables to be written in terms of tensor invariants;
ii. the quadratic dilatancy rule is

$$e = \nu d_{nm} d_{nm}, \tag{5}$$

which extends the parabolic relationship alluded to earlier to the three dimensional case and introduces what we take to be a material constant ν;
iii. the incremental energy balance for a unit volume of material

$$\sigma_{ij}\dot{e}_{ij} = \dot{\mathcal{D}}, \tag{6}$$

where $\mathcal{D}$ is the rate of dissipation and is chosen for this model to be

$$\dot{\mathcal{D}} = -\sigma\mu\sqrt{\dot{d}_{nm}\dot{d}_{nm}} \tag{7}$$

and μ is a material constant.

If the mathematical procedures developed by Chandler[18] are applied the yield function[19]

$$\left(s_{ij} + 2\nu\sigma d_{ij}\right)\left(s_{ij} + 2\nu\sigma d_{ij}\right) - (\sigma\mu)^2 = 0 \tag{8}$$

is produced. This incorporates a type of kinematic hardening and has been shown to simulate the cyclic behaviour of granular materials quite well as long as the strains are small and the material is in a dense state of packing.[19]

3.4. A model for pastes

Stiff pastes contain just enough liquid to flood the porosity but still have curved menisci on the free surface. The effect of this pore liquid and its surface tension is to produce a pore fluid tension (t) which is reacted against by increased granule-granule contact forces. This is neatly incorporated into our existing model by subtracting the pore tension from the mean applied stress to give an effective stress $\sigma' = \sigma - t$ which is then used in the yield function.[20] This is known as the Principle of Effective Stress and is well proven by a great many experiments on granular materials.[14] The yield function is therefore modified to become

$$(s_{ij} + 2\nu\sigma' d_{ij})(s_{ij} + 2\nu\sigma' d_{ij}) - (\sigma')^2 = 0. \tag{9}$$

To complete the model all that is needed is to specify a relationship between the pore tension and the volume strain. For the sake of simplicity, we take

$$t = t_0 + ke. \tag{10}$$

where t_0 and k are assumed to be material constants. The entire model has therefore four adjustable parameters ν, μ, t_0 and k.

4. Simulation of the tension–compression tests

Imagine a cylindrical column of paste, with its axis lying along the x_1 axis. It is acted upon by an applied axial stress σ_A but by no imposed stress on the curved surface. The mean applied stress is therefore $\frac{1}{3}\sigma_A$ and the non-zero components of deviatoric stress are

$$s_{11} = +\frac{2}{3}\sigma_A \quad (11)$$

$$s_{22} = -\frac{1}{3}\sigma_A \quad (12)$$

$$s_{33} = -\frac{1}{3}\sigma_A \quad (13)$$

We assume that the deformation produced in the column is homogeneous. (This assumption seems broadly true for the small deformations considered here but at larger strains the formation of bands of localised shear is common.) Let the axial strain be denoted by e_A and the lateral strain by e_L. The volume strain is therefore given by

$$e = e_A + 2e_L \quad (14)$$

and the non-zero components of deviatoric strain become

$$d_{11} = +\frac{2}{3}(e_A - e_L) \quad (15)$$

$$d_{22} = -\frac{1}{3}(e_A - e_L) \quad (16)$$

$$d_{33} = -\frac{1}{3}(e_A - e_L). \quad (17)$$

The dilatancy rule [Eq. (5)] can then be reduced to

$$e_A + 2e_L = \frac{2}{3}\nu(e_A - e_L)^2 \quad (18)$$

and the yield function [Eq. (9)] becomes

$$\frac{2}{3}\sigma_A^2 + \frac{8}{3}\sigma_A\nu\sigma'(e_A - e_L) + \frac{8}{3}(\nu\sigma'(e_A - e_L))^2 = \mu^2(\sigma')^2 \quad (19)$$

where

$$\sigma' = \frac{1}{3}\sigma_A - \frac{2}{3}k\nu(e_A - e_L)^2 - t_0. \quad (20)$$

The final mathematical model for the tension–compression test is as follows: given e_A the values of σ_A that solve Eqs. (18)–(20) give the axial stresses needed to extend and compress the column of paste. Specifically, if e_A is specified then e_L can be calculated from Eq. (18). Eqs (19) and (20) then reduce to a quadratic equation in σ_A. The results for one set of parameters (see Table 2), chosen to match the shape of the stress-strain curves seen experimentally, are shown in Fig. 2B.

5. Discussion

5.1. Features reproduced

The model, without being particularly sensitive to the values of the parameters used, can mimic almost all the features of the curves seen experimentally. These include: the weakening in the central region; the strengthening at the extremes of the cycle; the noticeable Bauschinger effect and the difference between tension and compression.

How are these features reproduced? The weak centre and strong extremes are simply a result of a combination of the quadratic dilatancy, effective stress and particle friction as indicated previously. However the tension–compression asymmetry is a result of the compressive stresses increasing the effective pressure between the particle contacts while the applied tensile stress reduces it. The Bauschinger effect occurs in these materials because the pore tension resists the volume expansion near the extremes of deformation while upon strain reversal the pore tension assists the volume reduction at the onset of plastic deformation.

5.2. Parameter values

Even though the model can be made to work quite well, are the values of the parameters reasonable? Let us take these in turn.

The value of t_0 is chosen to give good agreement at the central region of the cycle. Is this consistent with the particle size of the alumina? In order to check this we made two assumptions: the surface tension of the polymer solution is of the same order as water (0.07 Nm^{-1}); the liquid tension induced is proportional to the product of the surface tension and curvature. If these are correct the radius of curvature would need to be of

Table 2
The parameter values for the simulation shown in Fig. 2B which mimics the experimental results shown in Fig. 2A

Parameter	Fitted value
t_0	1.6 kPa
μ	0.5
ν	10.0
k	250 kPa

order 1 μm during the cycle to provide enough pore tension. This is within a physically realistic value for a powder with a mean particle size of 3 μm.

The value of μ is chosen to give the appropriate ratio of tensile to compressive strength. The value would not be inappropriate for a dry soil[14] and is therefore not unreasonable for this material.

The value of $k/t_0\nu$ controls the overall shape of the extending and compressing curves. Values from 5 to 50 give curves in line with the general shapes seen experimentally, so the model seems quite robust. If $k/t_0\nu$ and the stress range are kept constant then changing ν stretches or compresses the strain axis. The value of ν used does not lead to particularly high volume differences. For example the difference in volume between the centre and extremes of the cycle in the model is less than 2%. This is not excessive and is much less than the range of stable packing of spheres, which is about 8% of the bulk volume.

As mentioned earlier, most practical pastes incorporate some internal air filled voids. These are likely to be dispersed through the paste providing expandable cavities so that on dilation the increase in porosity can be accommodated locally and not just from fluid drawn from the outer surface. By ignoring the effects of elevated air pressure in the bubbles one can estimate that t_0/k will be of the same order as the volume fraction of air present (a few percent[16]). The value used in the simulation, therefore, does not seem unreasonable.

5.3. Further work

The basic assumption of this work is that in the central position the material has the minimum volume. While adequate for displacement controlled deformation where extension is balanced by compression, this is clearly inappropriate for a more general pattern of deformation. For instance, if the deformation was cyclic, yet had a bias toward either extension or compression, then the position of minimum volume would probably drift as in Macey's original experiments. The incorporation of this feature is beyond this paper as it requires the use of finite strain measures, it is however currently under investigation at Aberdeen.

In results, not reported here, in which the specimens were deformed at different rates, there appears to be a relatively weak rate dependency. That there is some is hardly surprising because of the presence of the viscous liquid. Further developments may therefore add viscous dissipation to the model if required.

As an additional investigation of this mechanism some of the values of the parameters used could be determined independently. This would provide a stringent test of the model and it is our intention to do this.

A useful universal model of granular mechanics is currently well beyond reach. However, there are a number of models that simulate various types of granular behaviour. The model used here, to simulate these cyclic tests, is not suitable for extended monotonic deformation or for situations where particle deformation is an issue. In these circumstances modern critical state models (for example, Ref. 21) are more appropriate and attempts to simulate processes such as extrusion would be worthwhile using these types of model.

There are a number of mechanical properties that are not covered by these types of model and these include the limits to the ductility of the paste and its adhesion to surfaces. The detail of the surface chemistry of the binder may well have a more pronounced influence on these properties than it does on the strength.

6. Conclusions

i. Cyclic compression testing of synthetically plasticised alumina powder reproduces the peculiar features seen in clay-based pastes by Macey.[7]
ii. Cyclic tension–compression tests for small strains can be successfully simulated using a model, originally developed to simulate the behaviour of dry powders, supplemented by capillary tension and the principle of effective stress.
iii. The parameters used in the simulation all have reasonable values so it is safe to assert that the granular character of these materials dominates their behaviour. This supports the idea that other aspects of paste behaviour such as compaction and extrusion could also be modelled using primarily granular mechanics.
iv. This shows that, as long as the fluid does not drain away, its main role, in the tests discussed here, is to provide surface tension that pulls the particles together. For processes where high pressures are used, however, the situation will be very different. The presence of the liquid will then tend to reduce the interparticle forces by sustaining much of the applied pressure itself.

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