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Plastic behaviour of different ceramic pastes processed by extrusion

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Abstract

A preliminary characterization of the plastic behaviour of several industrially-prepared ceramic pastes is presented, which was performed in order to gain some insights for preparing ceramic formulations Al-rich sludge-containing adapted for extrusion forming. Stress–deformation curves obtained by compression had been performed and related with the different types of ceramic formulations and their moisture contents. Deformation amplitude and rupture stress revealed the most sensitive parameters, thus giving valuable information about the plastic behaviour of ceramic pastes, as well as important guide lines to design extrudable formulations containing. The stress–deformation curves obtained with common industrial pastes are reproducible and relative differences are determined by the composition and amount of added water. The extrusion of fully recycled formulations made of Al-rich anodising sludge was found possible, but further improvements in working conditions should be achieved.

Keywords: A. Extrusion; C. Plasticity; Al-sludge containing paste

1. Introduction

The extrusion of ceramics is a common processing technique that is specially adapted for the production of regular and constant cross-section bodies [1,2]. It is used both for traditional products, like tiles and bricks, and for the production of technical devices used in the electronic and automobile areas (e.g. catalyst supports) [3,4].

The plastic behaviour of an extrudable ceramic paste is controlled by different factors [5]: (i) volumetric fraction, shape and size distribution of the particles, (ii) type of inter-particle dominant forces, (iii) surface chemistry, (iv) packing density and (v) rheological behaviour of the liquid phase. The control of the liquid (continuous) phase properties is critical for the success of the process. As a general rule, it must be more viscous as low is the deformation (or shear) rate.

Most of the traditional ceramic mixtures show plastic-type behaviour, due to the presence of the plate-like morphology of the particles of the clay-based raw materials that are able to slide over each other due to the water retained in the interstitial spaces that acts as lubricant. When necessary, commercial plasticizers might be added, such as methylcellulose or hydroxipropyl methylcellulose [6]. The measurement and control of the plasticity is essential to achieve good fabrication conditions (correct shapes and low processing times). However, the common practice is somewhat empirical, due to the large number of influent parameters and the lack of sensitive quantification means for the evaluation of the complex relationship between the flow characteristics and as-extruded components' properties [7,8]. Moreover, processing parameters not related to the paste have also relevant effects on the extrusion definition. For example, the die configuration (length and cone angle) proved to be crucial in the extrusion of alumina-based components [9]. Benbow's equation [2] describes the relationship between the extrusion speed, the applied pressure and the length of the extruder.

The plastic behaviour of ceramic pastes is generally predicted by stress–deformation compression curves [10,11], as shown in Fig. 1. Up to the point A (yield point)

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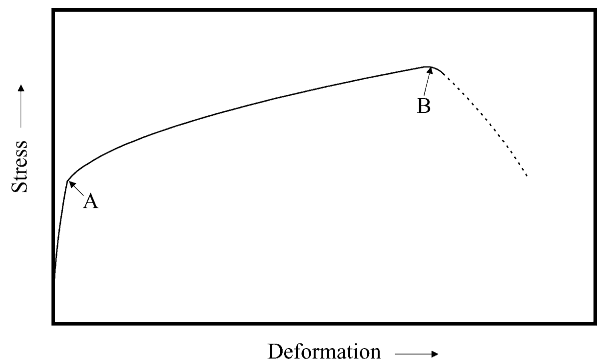

Fig. 1. Typical stress–deformation curve of a plastic ceramic paste.

the material shows elastic-type behaviour. Above this limit, a continuous increase of the stress induces plastic-type deformation. Finally, at the point B (maximum deformation) the structural failure will occur. Above this limit, clay-based material tends to shown an abnormal stress increasing instead of a decline, generally related with the enlargement of the area of contact with the support plates caused by compression forces.

This work presents a preliminary characterization of the plastic behaviour of several ceramic pastes, industrially-prepared, which was performed in order to gain some insights for preparing Al-rich sludge-containing ceramic formulations prepared in our lab and adapted for extrusion forming. Stress–deformation curves performed under compression tests could be related with the type of formulation and its moisture content.

2. Experimental

Industrial as-extruded cylindrical rods having diameters in the range of 8–17 cm, were collected from several traditional ceramic units: porcelain (Porcel, Oiã, PT), stoneware (Primagera 3, Bustos, PT), earthenware (Faianças as Primagera, Aveiro, PT), and terracotta (Argila Centro, Pombal, PT). Their average compositions are given in Table 1. A recycled paste exclusively made of Al-anodising sludge was also prepared, from a mixture (50:50 wt.%) of pre-calcined (at 1400 °C) and wet as-received material. In this case, the use of processing additives (1 wt.% of Zusoplast C28 plasticizer and 0.5 wt.% Zusoplast O59 lubricant, Zschimmer & Schwarz) was also tried [12,13] in order to cope the characteristic lack of plasticity of this material.

All the formulations were extruded through a cylindrical die (diameter of 33.0 mm) in order to obtain rods, which were then cut into test probes of the same diameter and about 43.0 mm length. Stress–deformation tests were carried out by plastic compression (Lloyd Instruments LR 30 K) in appropriate metallic moulds by following reproducible conditions. A minimum of three specimens per composition were tested. The compression tests were conducted at a constant rate of 2.0 mm/min until a maximum deformation of about 70% or the ultimate limit of the load cell (500 N) was reached. Fig. 2 shows a schematic representation of a cylindrical probe at the beginning and at the end of the compression test.

The moisture contents of the samples were varied and adjusted by their simple exposure to more dry or more wet environments. To reach equilibrium conditions, the samples were kept under the desired conditions for long periods of 1 week or more.

All the calculations necessary to plot the stress–deformation curves and evaluate the plastic behaviour of the samples were performed considering that the volume of the samples was kept constant along the test, assuming that the area of contact sample/support was inversely proportionally to the height. This approach, acceptable along the first stage of the test, tends to loose validity when high compression strengths are applied (near the end of the test), especially after cracks formation in the sidewall (as show in Fig. 2B).

3. Results and discussion

3.1. Industrially-prepared pastes

Fig. 3 shows stress–deformation curves of all industrially-prepared formulations. Initial testing conditions correspond to those use in the fabrication units that supplied the as-collected pastes, which therefore, possess the required degrees of homogenization and moisture for the respective processes. It can be seen that despite their different compositions and moisture contents, all the curves exhibit very similar trends. After a small yield stress, less perceptible in the more plastic compositions, the curves show a first shear-thinning-type behaviour followed by near

Table 1
Typical tested batch formulations (wt.%) of industrially-prepared pastes

	Composition (%)						
	Ball clay	Kaolin	Quartz	Feldspar	Calcite	Talc	Red clays
Porcelain	0–5	45–50	25	25	–	–	–
Stoneware	25	25	24–28	18–22	–	6–10	–
Earthenware	20	32–38	26–30	–	8-14	–	–
Terracotta	–	–	0–5	–	–	–	95–100[a]

[a] Generally it was a mixture of two red clays of different plasticity levels.

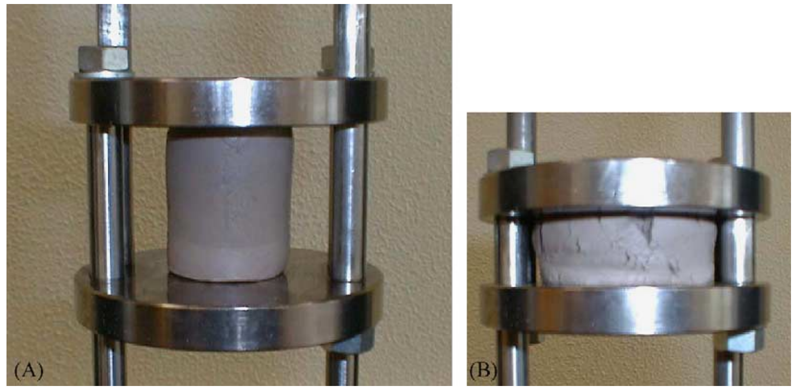

Fig. 2. Views of cylindrical probes used on stress–deformation tests: (A) at the beginning and (B) at the end of the test.

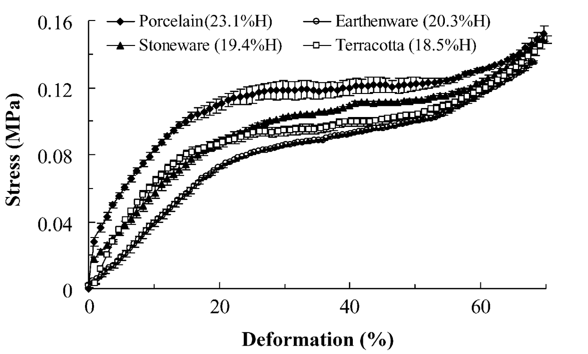

Fig. 3. Stress–deformation curves of several industrial formulations previously optimised for extrusion.

Newtonian plateaux, and finally ending with shear-thickening-type branches. The onsets of the shear-thickening-type branches would correspond to the starting of cracks formation in the sidewall (as show in Fig. 2B) due to tensile stresses generated in that region. In fact, the liquid film surrounding particles act as lubricant and enable particle rearrangements during deformation, while keeping particles together due to capillary forces. However, when the applied tensile stress exceeds the magnitude of capillary forces, water tends to recede while air is allowed to enter into the pores in the sidewall of the deforming body, as show in Fig. 2B. Rupture stresses change between 0.06 and 0.11 MPa and the extension of the plastic-deformation region is almost constant. Leanest formulations (made of less plastic raw materials) such as porcelain, require higher water amounts to reach suitable plasticity levels and optimal workability. On the other hand, terracotta paste is made of plastic clays that naturally contain high amounts of fine plate-like particles requires lower water content (optimal amount of about 18.5%). For all the compositions the rupture occurs for a deformation level of 50–55%. We might then conclude that the necessary previous adjustments of paste characteristics have been made in order to optimise the extrusion process, and since typical industrial working conditions are not very different, their plastic behaviours results similar.

An attempt to approach the moisture levels of all pastes was made in order to evidence the influence of different formulations on the plastic behaviour. The stress–deformation curves presented in Fig. 4 show that the leanest porcelain paste is also the shortest one, requiring rupture stress higher than 0.2 MPa, while the terracotta paste shows highest ability to deform without rupture and require the lowest stress levels for deformation along almost all the deformation range. Stoneware and earthenware pastes show intermediate behaviour, although closer to the terracotta paste, as expected since relative to the overall compositions

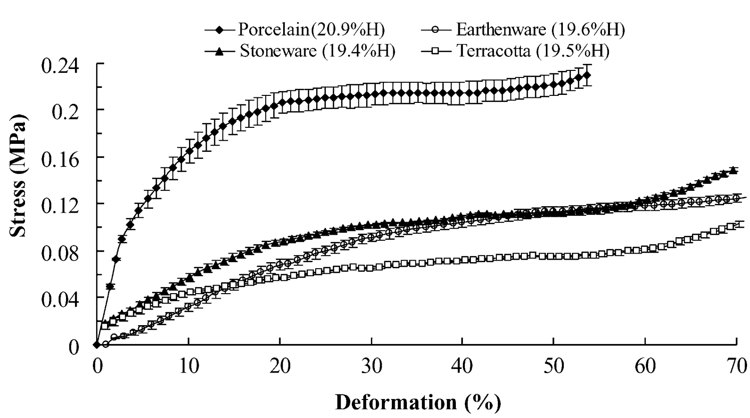

Fig. 4. Stress–deformation curves of several industrial common formulations showing similar humidity levels.

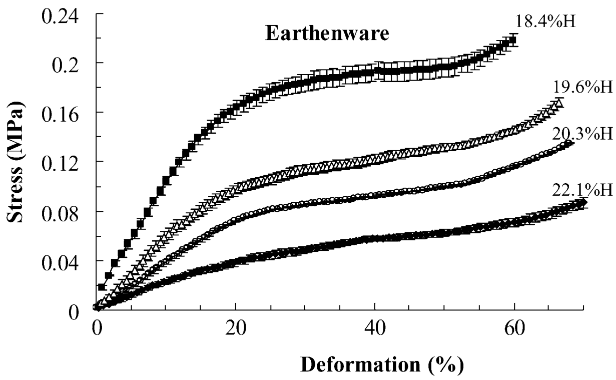

Fig. 5. Effect of the water content on stress–deformation behaviour of an earthenware industrial paste.

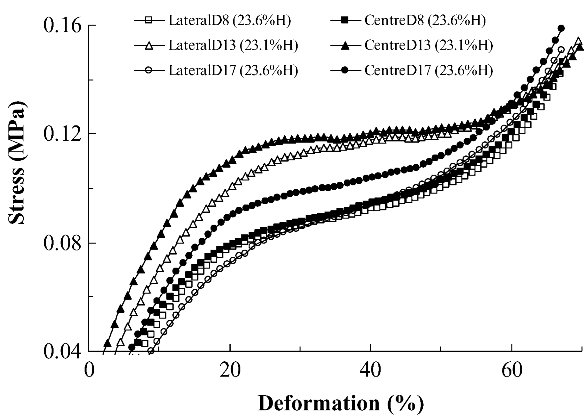

Fig. 6. Effect of the water content on stress–deformation curves of porcelain industrially-extruded rods having different diameters.

these formulations contain about 40 wt.% of ball clays and 10 wt.% kaolin, i.e. a total amount of clay-based materials of about 50 wt.%. Fig. 5 shows stress–deformation curves of earthenware formulations having different moisture levels. It can be seen that small variations in the moisture content (of the order of 1%) induce strong changes in the plastic behaviour. In general, the less humid samples are able to support higher stress levels before rupture but become shorter, showing less extended deformation in the plastic region.

As previously mentioned, porcelain paste (Fig. 6) requires higher water contents for flowing. In order to evaluate radial changes on this leanest paste prepared with 23.1–23.6 wt.% water, three different samples were collected from industrially-extruded rods having diameters of 8, 13 and 17 cm. Then, different slices from the central and external regions were collected and tested. Although the evolution of the respective curves is very similar, samples retired from the central zone tend to show slightly higher rupture stress values and more extensive plastic deformation. This observation is in accordance with expected better homogenization and compacting conditions in the middle of the extruded cylinders. By contrast, attrition forces are stronger in the region near the extruder wall and preferential particle orientation and lamination of clay-based pastes increases causing some mechanical degradation [10,11]. This effect is stronger for wider rods, and only very small differences are noticed in 8 cm diameter samples.

3.2. Sludge-containing pastes

The experiments using the common industrial pastes were basically conducted to check if the actual (and new) measuring conditions could give representative and accurate, results since their plastic behaviours are somewhat known. Once proved the adaptability of the technique for the prediction of relevant plastic characteristics, the new recycled pastes were then tested. Fig. 7 shows stress–deformation curves, denoting that pure (without additives) sludge formulations possess low plasticity. For the formulation containing 39.2 wt.% water, a maximum stress of about 0.6 MPa is reached for a deformation level of only 10%, decreases afterwards for higher deformation levels, while rupture is rapidly achieved. Formulations prepared with higher amounts of water (43%) and containing

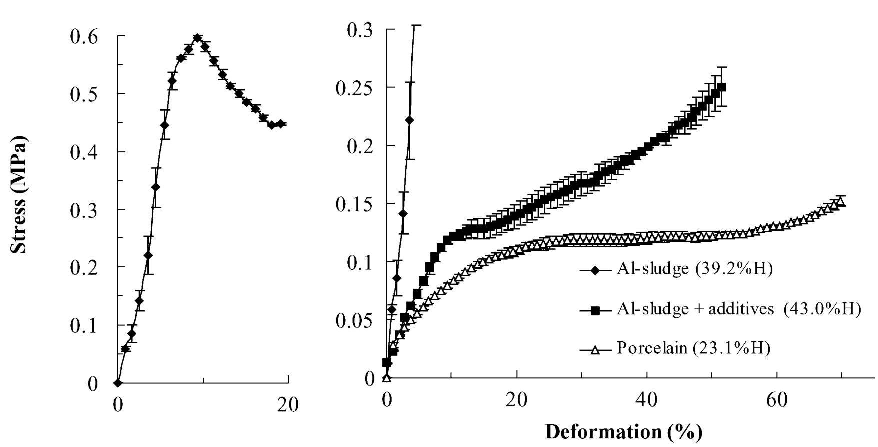

Fig. 7. Typical stress–deformation curves of Al-sludge pastes. The complete run of the pure paste is detailed in the left side. Comparison with a porcelain formulation is also shown.

plasticizer and lubricant agents (1 wt.% of Zusoplast C28 plasticizer and 0.5 wt.% Zusoplast O59 lubricant) show higher plasticity and better workability. Its behaviour is more close to that of an optimal porcelain paste, but the plasticity region is still incipient and its workability is far from the desirable conditions. Further improvements should be obtained, which deserves deeper optimisation studies, now in course.

4. Conclusions

Stress–deformation curves of compression tests seem to be a suitable method for the characterisation of the plastic behaviour of ceramic pastes generally used for extrusion forming. In particular, deformation amplitude and rupture stress are sensitive parameters for the prediction of the plasticity and its dependence on the composition and moisture content. Results obtained with common industrial pastes are reproducible and relative differences are determined by the composition and amount of added water. The extrusion of fully recycled formulations made of Al-rich anodising sludge was found possible, but further improvements on working conditions should be achieved in order to get well shaped pieces (e.g. tubes). Selection and adjustment of suitable plasticizers and/or lubricants or the combination of natural plastic materials, such as clays, are possible routes.

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