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# From dry pressing to plastic forming of ceramics: Assessing the workability window

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## HIGHLIGHTS

- Ceramic powders can be shaped only while in the funicular state.
- Dry pressing is effective above the pendular-to-funicular state transition.
- The workability window for plastic forming begins at maximum particle packing.
- The workability window extends to the funicular-to-capillary state transition.
- Dried body density can be used to quantitatively define the workability window.

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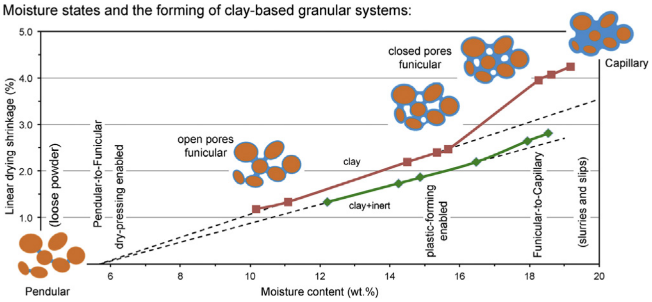
## GRAPHICAL ABSTRACT



## ABSTRACT

The shaping water content in clay-based ceramic building materials has conflicting implications in shaping and drying, decisively contributing to the cost-effectiveness of the industrial process. This work addresses the long-standing difficulties related to the quantification of the workability window for plastic forming. From uniaxial compression stress-strain curves, the yield stresses of mixtures of a red-firing clay and a ground basalt rock with different moisture contents were obtained. Combination of those results with the wet and dried bodies densities, showed that ceramic powders can be shaped only while in the funicular state: for this clay, dry pressing is effective above the pendular-to-funicular transition (~5.7 wt% moisture) and the workability window for plastic forming begins at the maximum dried body density, which signals the transition from open-to-closed gas pores in the funicular state (~15 wt% moisture), and extends to the funicular-to-capillary transition (~18 wt% moisture). Drying did not alter the moist particle structure, which enables the expeditious determination of the workability window from dried body density and initial moisture content.

## 1. Introduction

Processing of clay-based ceramic products, from raw materials conditioning, through shaping, to high temperature firing, includes a variety of steps, each of which having its own peculiarities and stringency. Because wet clays become plastic and can easily be shaped by plastic forming, water is a key ingredient throughout processing. Water helps most grinding operations and is thus generously used in industrial practice, leading to many shaping processes based on the direct use of ceramic suspensions (slips or slurries). But water must be removed from the ceramic pieces after shaping and before firing. The more water to eliminate, the costlier

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the process will become. Therefore, to enable processing with less water, a number of additives have been introduced to promote stability of concentrated suspensions and plasticity of ceramic pastes, while alternative processes and equipment enabled shaping from nearly dry powders [1]. In any case, the ceramic formulation always is a powder-air-water mixture, *i.e.* a wet granular system whose moisture content can typically vary, in industrial practice, from the ~5 wt% in dry-pressing of spray-dried powder granules to the ~40 wt% in slurries used in slip-casting.

Within such wide range of added water variation, specific characterization methods have evolved to best describe the behaviour of the particular granular system, both during shaping and during water removal (drying), and guide the corresponding industrial processing [2]. Dry-pressing is mostly concerned with powders that should flow easily to be shaped into ceramic pieces without density gradients [3], whereas slip-casting requires the ability to control the suspensions rheology and stability [4]. In between those two, industrial plastic forming of ceramic pastes relies on what is commonly described as the "workability window" of the wet ceramic paste, meaning the added water range within which the ceramic paste can be shaped into a green body without rupturing nor slumping [5].

When water is first added to a dry powdered clay, an increase in particle cohesion is observed. It is common industrial knowledge that pressing defects are to be expected when the spray-dried powders are "too" dry, the pressing efficiency being best when the powders moisture content surpasses 5 wt%. Cohesion then increases with the moisture content, as the water progressively displaces the air from the pores between the particles, to reach a maximum when the body becomes plastically deformable (*i.e.* the paste yields without rupturing). The corresponding added water is the so-called Atterberg plastic limit, *PL*. For water contents below the plastic limit, the paste is too hard to be shaped by plastic forming. As a rule, higher moisture contents lubricate the solid-solid friction and result in lower yield stress (or elastic limit, as in the stress-strain curves normally produced for plastic forming of metals) and easier plastic deformation (*i.e.* lower energy consumption), so cohesion decreases again [2]. However, if the moisture content is too high, above the so-called Atterberg liquid limit, *LL*, the paste turns into a slurry and starts behaving like a viscous fluid. From this arises the definition of the plasticity index, *PI*, of a ceramic paste ($PI = LL - PL$), whose measurement and control are needed to optimize the processing conditions. Because the yield stress needs to be sufficiently high so that the wet shaped products do not sag under their own weight, the ideal consistency for plastic forming and the workability window of plastic ceramic pastes lies between the Atterberg limits, *i.e.* occurs for water contents above the plastic limit and below the liquid limit.

Plasticity can be influenced by factors related not only to the ceramic paste itself but also to the shaping process and, despite the apparent simplicity of the definition [5], the workability of a ceramic paste is rather difficult to quantify (very dependent on the technician's skill) and transpose to the specific shaping process. The simple control of paste plasticity is not enough to ensure the absence of drying defects (cracks, warp) and efforts have been made to extend the simpler plasticity measurement methods (*e.g.* Atterberg, Pfefferkorn, indentation) [5,6] to other more elaborate, costlier methods that evaluate the relationship between an applied force and the resulting deformation (*e.g.* stress/strain curves, rheometry) [7,8]. Albeit more popular in industry, the former are still limited and industrial practice tries to avoid such defects by keeping the water content low and by the incorporation of degreasing (inert) materials. Thus, the need persists for a less qualitative assessment of the workability window of ceramic pastes.

As mentioned above, dry granular materials are non-cohesive. If the ceramic formulation is regarded as a wet granular system, *i.e.* a powder-air-water mixture, the body cohesion results from the surface tension at the water-air interface [9]. Liquid induced cohesion depends on the moisture content and so do the observed changes in the mechanical properties of the body. The various levels of moisture result in different liquid distributions around the solid particles and can be divided in as many behavioural states, namely, adsorbed, pendular, funicular, capillary and slurry [9–11]. There is no significant cohesion in the presence of only adsorbed water and neither in slurries, in which particles are fully immersed in liquid and there is no capillary action at the surface.

The intermediate states are those that matter when studying the mechanical behaviour of moist ceramic pastes. For a given array of solid particles, in the pendular state particles are held together by liquid bridges that form between contacting particle pairs. Cohesive forces act through those liquid bridges. A cohesion stress $\sigma_c$ can be defined (Eq. (1), relating the average force $F$ per liquid bridge with the average number $k$ of bridges per particle, the average particle size $d$, and the particle packing density $\nu$ [9]:

$$\sigma_c \approx \nu \frac{k}{\pi d^2} F \tag{1}$$

Eq. (1) shows that cohesion will be stronger when the particles are better packed and in the presence of more liquid bridges. In the pendular state the gas phase (air) is continuous and the liquid phase (water) is discontinuous. As the number of liquid bridges increases, cohesion progressively increases until the liquid occupies ~25% of the available empty (gas) space [12]. From then on the system enters the funicular state, in which the liquid phase is continuous around contact points and also fills some pores, but there still remain voids filled with air (the funicular state can exist both with open gas pores or with closed gas pores). When only closed gas pores remain, the number of bridges stops changing significantly and the cohesion stress is expected to depend only on the particle packing density, which is mostly determined by the shaping process. In the capillary state all voids between particles are filled with liquid, but the surface liquid is drawn back into the pores due to capillary suction (liquid menisci form at the surface). If the transition from open-to-closed gas pores within the funicular state and the associated change in cohesion mechanism brings to mind the description of the Atterberg plastic limit [5], the entrance to the capillary state reminds the description of the Atterberg liquid limit.

The application to moist clays of compressive workability tests commonly used for metals has been tried [7], seeking a relationship between the plastic work carried out from yield stress to rupture and the plasticity index (and the workability window), but the results were rather qualitative and not very conclusive. However, if cohesion in a body is a measure of the particles ability to remain attached, there must be a cohesion related parameter that might enable a better quantification of the workability of a ceramic paste. This work seeks an interpretation of the measured plastic behaviour of clay-based green bodies in the light of the wet granular systems theory (*i.e.* as powder-air-water mixtures), looking for a quantitative assessment of the workability window of ceramic pastes, one more reliable and easier to determine than current plasticity index evaluation.

## 2. Experimental

A typical clay from the ceramic industry at the Caí River valley in the Rio Grande do Sul State, Brazil, was used as plastic raw material. For the role of inert material a basalt gravel from the Serra