ASSESSING THE PLASTICITY OF CERAMIC MASSES BY DETERMINING INDENTATION FORCE

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ABSTRACT

The methods currently being used to determine plasticity give rise to different problems (mainly lack of reproducibility and repeatability), which limit their application in controlling ceramic bodies and raw materials.

After critically analysing the procedures put forward in the literature, it was considered advisable to develop a new experimental procedure to assess plasticity in ceramic masses, based upon indentation trials performed on ceramic masses with differing consistencies, obtained by modifying moisture content.

A laboratory apparatus was designed and built, which, on connecting it to a computer, allows the test to be performed, yielding the information conveniently and precisely.

The Plasticity Index values obtained by this procedure were compared to those yielded by a traditional method, confirming their validity as well as their high degree of reproducibility and repeatability, which recommends its industrial use in controlling raw materials and ceramic bodies.

1. INTRODUCTION

1.1. PLASTICITY

Plasticity has traditionally been defined [1-3] as "the property of a material that enables it to be deformed without rupturing by the application of an external force that exceeds a critical value, and to retain that shape when the force is removed or reduced below this value". According to this definition, the material will only undergo plastic or permanent deformation when a certain critical value of force is exceeded, called *yield stress*.

As may be observed, the above definition is purely qualitative and only indicates the behaviour that a wide range of materials exhibit on applying a pressure on them that exceeds their yield stress. Furthermore, this behaviour depends on the conditions in which the force is applied (principally direction, duration and velocity), so that plasticity should not, strictly speaking be considered a specific material property.

Clay minerals develop this type of behaviour when they are mixed with water, a characteristic that has been widely used in forming traditional ceramic ware. The plasticity of these mixes mainly depends on the relation water/clay and the properties of the clay material (mineralogical composition, particle-size distribution, specific surface area, colloids, soluble salts, etc.). Thus, for a given clay composition or clay, masses having a high water content are really ductile and can undergo considerable deformation without rupturing on being subjected to relatively small forces; however, masses having a low water content break under small deformations.

One clay material is usually considered to be more plastic than another when it withstands greater deformation than the other without rupturing. This comparison must be performed at the moisture contents for which both have the same yield stress. On the other hand, if the comparison is effected at moisture contents for which these have the same maximum deformation value without rupture, the more plastic material will have an appreciably higher yield stress.

1.2. INDUSTRIAL IMPORTANCE OF PLASTICITY

It is common practice to relate the plasticity of clay pastes to their behaviour in different manufacturing process stages prior to firing (dispersion, moulding, drying, transport, etc.). This is possible because the properties of the clay mineral that determine plastic behaviour (particle-size distribution, specific surface area, mineralogy, etc.), also determine the properties controlling these stages (mechanical strength, permeability, compactness, etc.).

The existence of these empirical relationships makes plasticity a common parameter in formulating and adjusting clay compositions, in which materials of differing plasticity are mixed until a mixture is obtained that performs optimally.

As a rule, optimal plasticity can be considered as the minimum plasticity required for the moulding process to be performed satisfactorily, without any subsequent deformations or problems involving low mechanical strength in the green or dry pieces arising.

Bodies with low plasticity processed by extrusion, need to be formed at an excessively low consistency in order to obtain suitable flow in moulding, which causes deformations to arise easily in the green piece. Moreover, these compositions usually have a narrow range of working moisture contents, as their consistency varies considerably with moisture content. However, high plasticity entails greater drying difficulties and a greater tendency for cracks to form, owing to greater water retention and low permeability.

In the bodies moulded by pressing, the behaviour of the pieces during the mould release step is highly influenced by the ratio *green mechanical strength/ release force*, with fissures and laminations appearing when this ratio is low [4]. This release force, just as the properties mentioned above (mechanical strength, permeability, etc.), is related to the plasticity of the material at the moisture content at which pressing takes place. On the other hand, in compositions prepared by the wet process, the higher their plasticity, the greater the difficulties involved in deflocculating them.

Plasticity is also a useful quality control parameter, owing to its sensitivity in detecting changes in the physico-chemical properties of the clay materials. These changes are quite frequent as a result of the intrinsic heterogeneity of the deposits they come from.

1.3. ASSESSING PLASTICITY

1.3.1. Existing methods

Generally, two kinds of methods can be distinguished for assessing plasticity [1]:

a) **Indirect methods.** These do not directly evaluate plastic behaviour but other properties related to it. Some of the most widely employed indirect methods currently in use are:

- *Atterberg method* [5-7]. The interval of moisture contents is determined at which the clay is workable. To do so, a plasticity index is defined as the difference between the moisture contents the material possesses at the liquid limit, or moisture content beyond which the mass is insufficiently consistent to be mouldable, and the plastic limit, or minimum water percentage required to make the clay mouldable without cracks arising.
- *Pfefferkorn method* [8-10]. This method determines the moisture content at which a cylindrical test piece, of standard dimensions, undergoes a 30% axial compression caused by the impact of a metal disc of constant weight, released from a standard height.
- *Barna method* [11]. This investigator considers the plasticity mechanism to be a function of the colloidal water of the material, and defines an index as the ratio between colloidal water and tempering water.
- *Mechanical strength* [1][12]. This method is based on measuring the modulus of rupture of dry test pieces, considering that the higher plasticity is, the greater mechanical strength will be.

Although the indirect methods have been highly criticized by various authors, as they do not really assess plastic behaviour, they are currently used in routine controls owing to the low cost of the equipment used and the short time required for the tests.

b) **Direct methods.** These methods determine the effect moisture content of the clay body has on the relationship between an applied force and the resulting deformation.

One of the simplest, widely used direct methods is the one developed by Moore [1][13-14]. This investigator defines a plasticity index as the ratio of the pressures applied to produce two arbitrarily chosen deformation values (10% and 50%), determining these values in the compression pressure/deformation curve obtained on compressing a cylindrical test piece between two plates at a constant rate. Other tests that are also used involve bending, torsion, shear, and extrusion testing [1-3][15-18].

The direct methods are considered to define the plastic behaviour of the material most closely. Characterization of the behaviour of a plastic mass can only be performed by determining its different rheological parameters [19] (yield stress, viscosity, limit deformation without rupture, viscoelastic parameters, etc.); however, their usefulness in routine controls is restricted, mainly owing to the cost of the equipment required, so that they are only used in research and in materials with a high added value.

The main criteria to be taken into account in choosing a method for assessing plasticity, will depend on the manufacturing process used (manual moulding, extrusion, pressing, casting), as it may be convenient to subject the material to conditions resembling those it will undergo in the industrial process, and on the purpose of the measurements (characterizing raw materials, compositional control, etc.). The speed at which results are obtained will be primordial in quality control, while it is not such a determining factor in characterization. Finally, assessing actual plastic behaviour will be especially relevant in forming processes in the plastic state.

The above methods have been qualitatively compared in Table 1. This comparison has been performed from references found in the literature and from our own experience.

Table 1. *Qualitative comparison of varying methods for assessing plasticity. The rating scheme used is : 0=deficient, 1=average, 2=suitable and 3=optimal.*

	CRITERIA				
METHODS	Speed	Cost	Precision	Sensitivity	Flow*
MOORE	3	1	1	1	2
ATTERBERG	1	3	1	3	1
PFEFFERKORN	1	3	2	2	0
BARNA	0	3	2	0	1
RESISTANCE	1	1	2	1	0

(*) Plastic behaviour assessment

The method proposed by Atterberg is observed to have some advantages over the other methods examined, which is why it is widely employed. These reasons involve its low cost and sensitivity in distinguishing between materials that have slightly different behaviour. In spite of this, the method's lack of precision is a significant drawback, which limits its use in controlling materials.

1.3.2. Proposed new technique

The experimental technique determining the Atterberg limits, is based on work done by Atterberg, which was subsequently completed by Casagrande. This method is currently in widespread use. However, the results obtained exhibit low precision owing to:

- The experimenter. It is difficult to exactly determine the moisture contents at which the limits, especially the plastic limit, lie.
- The testing equipment. Fall height and spoon striking rate, as well as the dimensions and shape of the groover, also noticeably affect the value of the liquid limit, which is why they are laid down in a standard test [5], and must be checked at regular intervals.

These drawbacks can be overcome, determining the Atterberg limits by another procedure. According to some workers [20-21], the tests determining the Atterberg limits have been shown to measure, in an approximate manner, the water content at which the material has two specific shear strength values. Therefore, the Atterberg index could be obtained by measuring the shear strength of the material on mixing it with different amounts of water.

Figure 1 depicts the variation in shear strength or consistency of a clay mass with its moisture content [22-23]. According to the water content of a material, it can be found in four varying states: a solid, semi-solid, plastic or liquid state. The range of plastic behaviour lies between the Atterberg limits. The figure shows a consistency peak, whose moisture content corresponds to the true plastic limit, H_{PL} [24]. However, the liquid limit H_{LL}, lies at an arbitrarily defined consistency, as it is not really a physical limit [7][24].

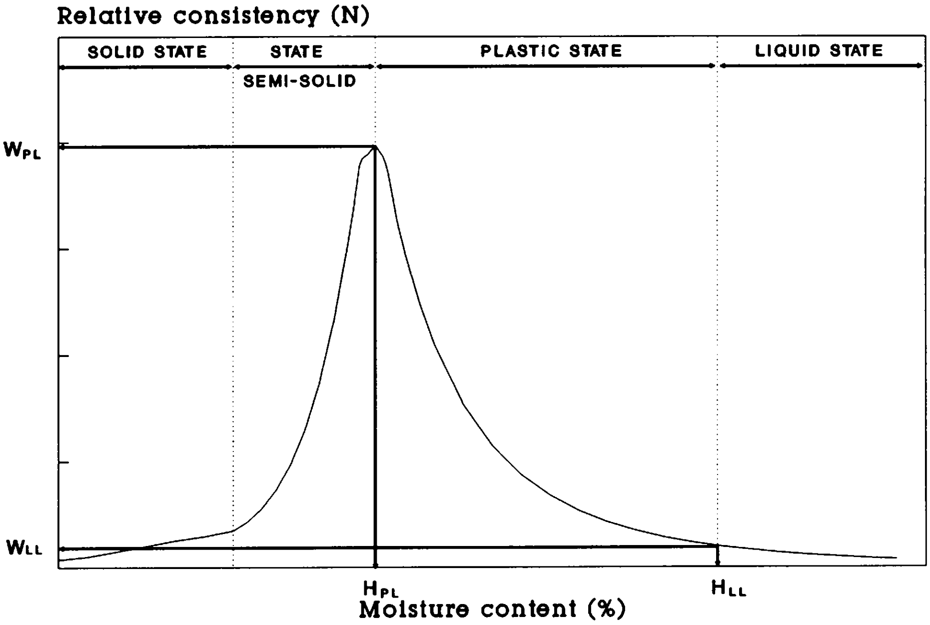

Figure 1. *Variation of consistency with moisture content.*

At the moisture content interval at which the material is in a plastic state, the variation in consistency with moisture content is approximately parabolic [3]. Therefore, this domain of the curve can be fitted to an easily linearizable equation, of the form:

$$W = a\,H^{b}$$

where:
W = consistency
H = moisture content (%)
a and b = characteristic parameters of the material

If the Atterberg limits are assumed to lie at preset consistencies, and a set of pairs of consistency-moisture content values are available in the plastic behaviour zone, linear fitting easily allows the moisture content of these limits to be obtained. There is in this method a certain error in calculating the plastic limit, as the curve in its proximity is not parabolic; nevertheless, owing to the steep slope of the curve in this zone, the error in calculating moisture content is very small.

2. OBJECTIVES

In spite of the existing interest in assessing plasticity, for quality control of clay raw materials and bodies, as well as for body formulation, it is not currently routinely performed, as a result of the different problems involved in the existing tests (lack of precision, slowness of the tests, etc.). This study has therefore set the following objectives:

- Preparing an experimental procedure for assessing plasticity, which is useful for control and characterization of raw materials and clay masses.
- Designing and building a testing assembly, which will allow the method developed to be performed in a cost-effective way.

3. MATERIALS AND PROCEDURE

3.1. MATERIALS

To perform the tests, four clay samples having very different characteristics were chosen. Table 2 and Figure 2 show the chemical analysis, specific surface area and particle-size distribution of each of these materials.

Table 2. Characterization of the clays used.

REF	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	TiO_2	L.o.i.*	S.S.⁺
I	48.15	28.51	9.33	0.10	0.26	0.10	0.51	1.48	11.20	34.4
II	61.24	18.75	6.70	1.50	1.37	0.28	3.80	0.82	5.25	20.5
III	62.50	17.50	6.90	1.39	1.56	0.37	3.11	0.91	5.62	11.7
IV	48.00	37.00	0.65	0.07	0.30	0.10	1.75	0.02	12.50	15.4

* L.o.i. = Loss on ignition

⁺ S.S. = Specific surface area (m^2/g)

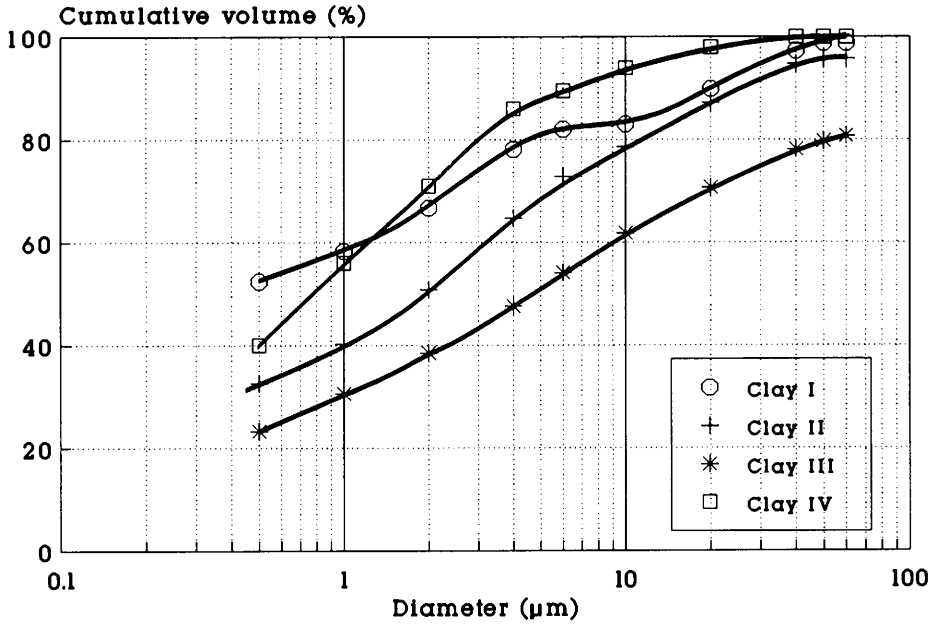

Figure 2. Particle-size distribution of the clays studied.

3.2. EXPERIMENTAL PROCEDURE

3.2.1. Proposed technique

a) Apparatus.

Figure 3 shows a schematic of the apparatus designed. It is made up of the following main parts:

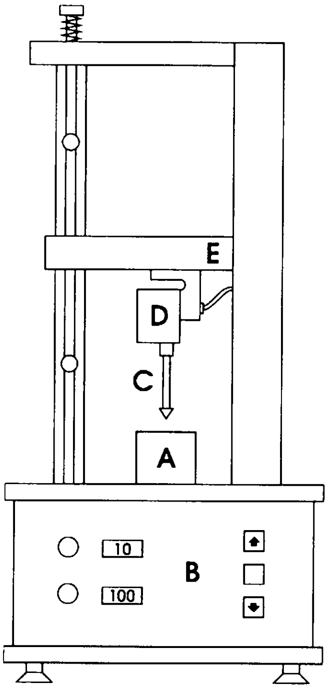

Figure 3.Plasticity apparatus.

A) Sample-holding mould of circular cross-section, 5 cm high with a diameter of 5 cm. These dimensions allow edge effects to be neglected.

B) Control unit.

C) Conical punch for indentation (height: 15 mm, angle:30°).

D) Load cell of 100 N with an accuracy of ±0.01 N.

E) Rising and falling frame, allowing testing to be performed at a constant rate of 10 mm/min.

b) Work method.

The samples were ground in a hammer mill with a discharge screen size of 0.5 mm, tempering with the amount of water needed to start the trials. The masses thus obtained were kept in closed receptacles for 24 hours, in order to achieve uniform moisture content.

To perform the study, the mass was transferred to the cylindrical mould. The excess material was removed by using a spatula, so that the body completely filled the mould, with a smooth top surface. Several measurements were effected with each sample, using varying water contents.

The value of the force was recorded at the moment the conical part of the punch completely entered the plastic mass. As the penetrating punch surface remained constant in all the trials, the value of the indentation force can be taken as a relative value of shear strength, or consistency [1].

The plasticity indices were computed according to the procedure described in Section 1.3, considering the liquid limit and plastic limit to lie at consistencies of 1 and 46 Newtons respectively.

In order to confirm the improvement in precision of the proposed procedure in respect of the traditional one, a study was carried out on the reproducibility and repeatability of the findings using both methods [25]. Repeatability, r, was determined as the error associated with the results obtained under nominally identical conditions (that is, identical clay samples, same equipment, working in the same laboratory with the same worker). On the other hand, reproducibility, R, was determined as the error associated with the results obtained by different workers, while holding the other conditions.

3.2.2. Traditional testing technique

The experimental procedures adopted in determining the Atterberg limits were as specified in the UNE standards [5][6].

4. RESULTS AND DISCUSSION

It was indicated in Section 1.3 that the variation in consistency with moisture content of clay bodies in the plastic behaviour interval was of a parabolic kind, which allowed the Atterberg limits to be calculated, whenever there were enough available data in this interval.

Figure 4 shows a plot of consistency on logarithmic coordinates versus moisture content of the bodies prepared with the four selected clay materials. The experimental points are observed to satisfactorily fit straight lines, substantiating the validity of the assumptions.

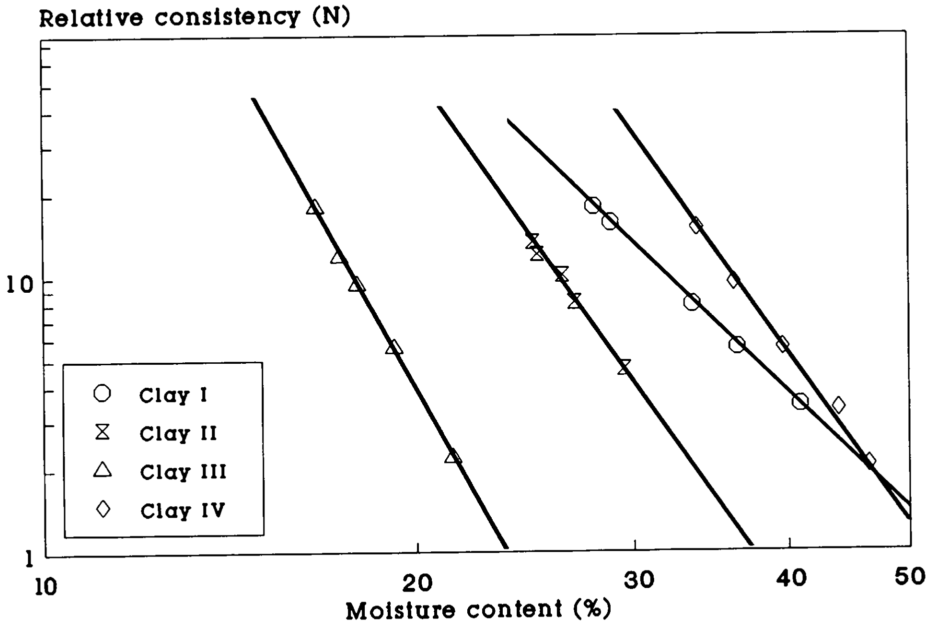

Figure 4. Variation of consistency versus moisture content.

The values of the liquid and plastic limits, as well as the resulting plasticity index, found from the previous experimental data, are detailed in Table 3. The results obtained by the traditional method are also included, besides showing the percentage differences exhibited by the proposed procedure compared to the traditional one, ϵ. These differences were computed for the sole purpose of comparing the two experimental techniques qualitatively, not a quantitatively.

Table 3. Overview of the results obtained by both techniques.

	TRADITIONAL			PROPOSED					
REF.	LL (%)	PL (%)	PI (%)	LL (%)	ϵ (%)	PL (%)	ϵ (%)	PI (%)	ϵ (%)
I	58.5	23.0	35.5	53.9	7.9	22.5	2.2	31.4	11.6
II	38.0	20.0	18.0	38.1	-0.3	20.1	-0.5	18.0	0.0
III	23.3	13.7	9.6	23.8	-2.1	14.7	-7.3	9.1	-5.2
IV	52.0	28.0	24.0	52.7	-1.4	28.0	0.0	24.7	-2.9

An examination of the results obtained allows the following aspects to be highlighted:

- The results obtained by the indentation procedure are quite similar to those obtained by the traditional method, as the percentage differences, except for clay I, never exceed 6%.
- The difference observed in the liquid limit value in sample I, is likely to be due to the great adhesion of this material, of fine particle size, to the Casagrande spoon bowl, which entails a shift in the liquid limit to higher water contents. Contrariwise, materials with coarse particles adhere only slightly to the bowl and the values of the limit, obtained by the traditional procedure will be lower than those determined by the proposed technique (sample III).

Using a method as a control tool requires its repeatability, r, and reproducibility, R, to be suitable. Table 4 details the precision study performed, confirming that reproducibility and repeatability of the results obtained have clearly improved with the new experimental technique.

Table 4. Precision of the techniques

	TRADITIONAL			PROPOSED		
	LL	PL	PI	LL	PL	PI
r (%)	4.0	4.9	3.7	1.1	1.1	1.2
R (%)	4.0	5.9	5.0	1.1	1.4	1.6

5. CONCLUSIONS

The following inferences may be drawn from this study:

- The method proposed by Atterberg is currently the most widely used one owing to the advantages involved in respect of other methods, but its use in quality control is quite restricted, especially as a result of its lack of precision.
- A new experimental technique has been developed to calculate the Atterberg limits, based on the determination by indentation of shear strength of a plastic mass at varying water contents, substantiating that the values obtained are quite similar to those determined by the traditional procedure.

- The precision of the results obtained by the new experimental technique has improved appreciably with regard to the traditional procedure, which allows its use in quality control.
- An assembly has been designed and built, which enables indentation trials to be performed at a constant rate in plastic masses with a wide range of consistencies.

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