



11<sup>th</sup> International Congress on Metallurgy & Materials SAM/CONAMET 2011.

## Talc, Spodumene and Calcium Carbonate Effect as Secondary Fluxes in Triaxial Ceramic Properties

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### Abstract

Triaxial ceramics (clay-quartz-feldspar) represents a significant proportion of traditional ceramic production; art, craft and/or industrial. The use of flux is widely used in order to modify the range of maturation of ceramic pastes.

The objective of the present study is to establish the influence of the addition of different secondary fluxes in the plasticity, sinterability, porosity, contraction, and mechanical properties of triaxial ceramics produced in a wide range of temperatures (800°-1200°C). The samples were elaborated with national raw materials with industrial quality and availability. A traditional clay-quartz-feldspar mixture was employed as a standard; In the present study Talc, Spodumene, and Calcium carbonate were chosen as secondary fluxes and partially replace the feldspar.

All the fluxes enhanced the sinterization, Furthermore the use of talc permitted to obtain a complete dense ceramic at 1200°C. On the other hand the use of spodumene resulted in materials with high elastic modulus.

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Keywords: triaxial ceramics; processing; sintering; mechanical properties.

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## 1. Introduction

Triaxial ceramics (clay-quartz-feldspar) represents a significant proportion of traditional ceramic production; art, craft and/or industrial. The use of flux is widely used in order to modify the range of the firing temperature of these kind of ceramics; Carty and Senapati, 1998, Harvey and Murray 1997 Iqbal and Lee 1999 Zanelli et al. 2011.

In these materials the principal flux is the feldspar, due to its more viscous glassy phase at high temperature, in the present work potassium feldspar (Orthoclase) was employed. This was partially replaced by other secondary fluxes. These introduce alkali and alkaline earth oxides to the formula, all these fluxes will fuse over 1100°C and will influence the evolution of the crystalline phases and consequently the properties of the final ceramic materials. The disappearance and neomineralization of clay and clay containing materials by firing in ceramic kilns has been widely studied; Ptáček et al. 2010A, Ptáček et al. 2010B, Bogahawatta and Poole 1991, Wattanasiriwech and Wattanasiriwech 2011, Grosjean 1985 and Bernasconi et al. L. 2011 . Due to the incorporation of the secondary fluxes the produced glassy phase during firing will have different composition with different properties as well Carty and Senapati, 1998 .

The objective of the present study is to establish and compare the influence of the addition of different secondary fluxes in some properties and behaviour of triaxial ceramics like: plasticity, sinterability, porosity, contraction, and mechanical properties of triaxial ceramics produced in a wide range of temperatures (800°-1200°C). The samples were elaborated with Argentinean raw materials with industrial quality and availability.

## 2. Experimental procedure

The compositions of the modified triaxial formulas studied are shown in Table 1. Prior to sintering, the effect of additives in terms of plasticity (Attenberg method) was evaluated. Subsequently the samples were produced by plastic deformation. Prismatic specimens (10x10x100mm<sup>3</sup>) were produced and let them dried in air for 3 days and in stove at 100°C up to constant weight. Subsequently samples were fired in electric kiln in air atmosphere at different temperatures between 800°C and 1200°C during a 30 minutes soaking. The heating and cooling rate employed was 5 °C/min.; consecutively the sintering parameters were evaluated. Porosity was measured by Archimedes method.

Table 1. Studied Triaxial ceramic formulas (weight basis).

Formula	Clay	Quartz	Feldspar	Talc	Spodumene	Calcium Carbonate
P1	60	15	25			
P2	60	15	10	15		
P3	60	15	10		15	
P4	60	15	10			15

A simultaneous thermo-gravimetric analysis (TG) together with the differential thermal analysis were carried out (DTA) to the four studied compositions in a NETZSCH 409/c equipment. They were completed with a 5 °C/min heating rate up to 1200°C and were cooled at the same rate. Samples of approximately 550 mg were analyzed in PtRh crucible and dynamic air atmosphere, using pure alumina as reference material described elsewhere in , Rendtorff et al. 2011A , Rendtorff et al 2011B .

Finally mechanical properties (flexural strength ( $\sigma_f$ ) and modulus of elasticity (E)) were evaluated. Three-point bending method and impulse excitation technique were employed; equipment was also described elsewhere Rendtorff and Aglietti 2010, Rendtorff et al. 2009.

### 3. Results and discussion

The addition of talc decreased significantly the range of plasticity while the influence of the other two secondary fluxes was not noticeable; this was expected because the clay amount and type was not changed. The values correspond to plastic type clay (ball clay), the actual values can be observed in figure 1A.

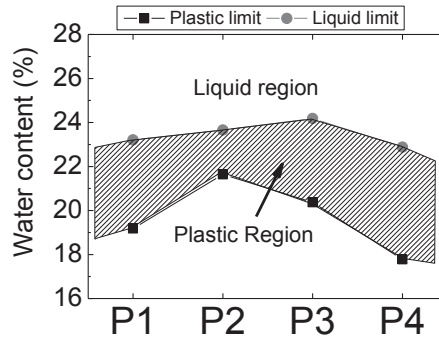


Fig. 1: Plasticity parameters of the studied formulas.

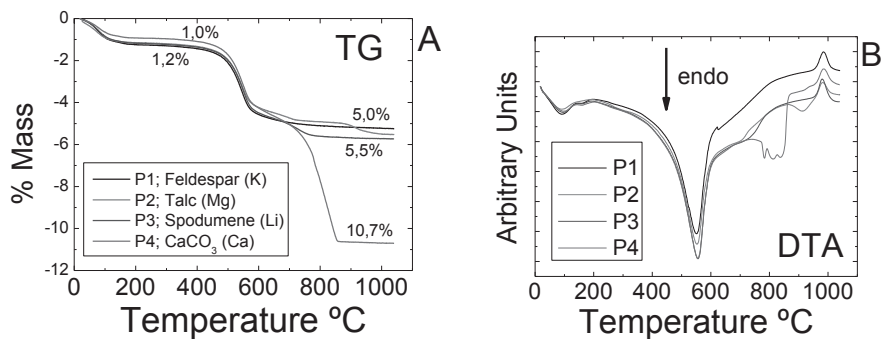


Fig. 2: (A) Thermogravimetric analysis (TG) and (B) differential thermal analysis (DTA) of the studied materials.

Figure 2A shows the results of thermogravimetric analysis of the studied materials. The observed behavior was typical of clayey ceramic bodies, Ptáček et al. 2010A. There is an initial loss of about 1% at 100° C, corresponding to water adsorbed on the surface of the crystals. Then there is clearly a significant mass loss around 600° C (begins at 500 and ends at 600 ° C) which corresponds to the loss of chemical water of the clay fraction of the formulas studied. As the four formulas had the same amount of clay, these losses are equivalent. From this temperature differences can be observed that P4, which contains calcium carbonate, has a significant loss between 600 ° C and 800 ° C. This loss resulting from the decomposition of carbonate was also observed by XRD, showing that even at 800 ° C the carbonate is partially decomposed into oxide. The small differences observed in the other pastes at these temperatures show impurities (low contents of carbonates, etc) in the raw materials.

Figure 2B shows the result of differential thermal analysis. There is a correlation between the DTA and TG. Again the behaviour was typical of clayey ceramic bodies, Ptáček et al. 2010A, Ptáček et al. 2010B, Bogahawatta and Poole 1991. It detects the loss of adsorbed water and chemical to 100-150 ° C and 500-600 ° C respectively. Both endothermic processes, meaning that take energy of the medium, the quartz transformation occurs at 573°C but the dehydroxylation processes is much more energetic than the quartz transformation hence is difficult to detect, in fact the observed peak corresponds to addition of both processes[ref]. Then again there are differences for P4 due to the decomposition of calcium carbonate which occurs in several steps between (750-820°C). An endothermic process at 900 ° C corresponding to the decomposition of talc was observed for P2 Ptáček et al. 2010A, Ptáček et al. 2010B, Bogahawatta and Poole 1991.

Finally there is an exothermic peak for the four compositions studied for the formation of pre-mullite spinel type aluminosilicate which can not be easily detected by XRD to 950°C Ptáček et al. 2010B.

Afterwards, as previously mentioned, contraction, porosity and density of the samples were evaluated at different temperature ranges. The results are shown in Figures 3A-C respectively. All the fired samples were transformed into a ceramic material. Observing the behaviour of these parameters it can be conclude that the sintering started only after 1100°C treatments. For the additive traixial materials (P2-P4) the sinterization was more abrupt. The sinterability follow the subsequent sequence (P2>P3>P4>P1).

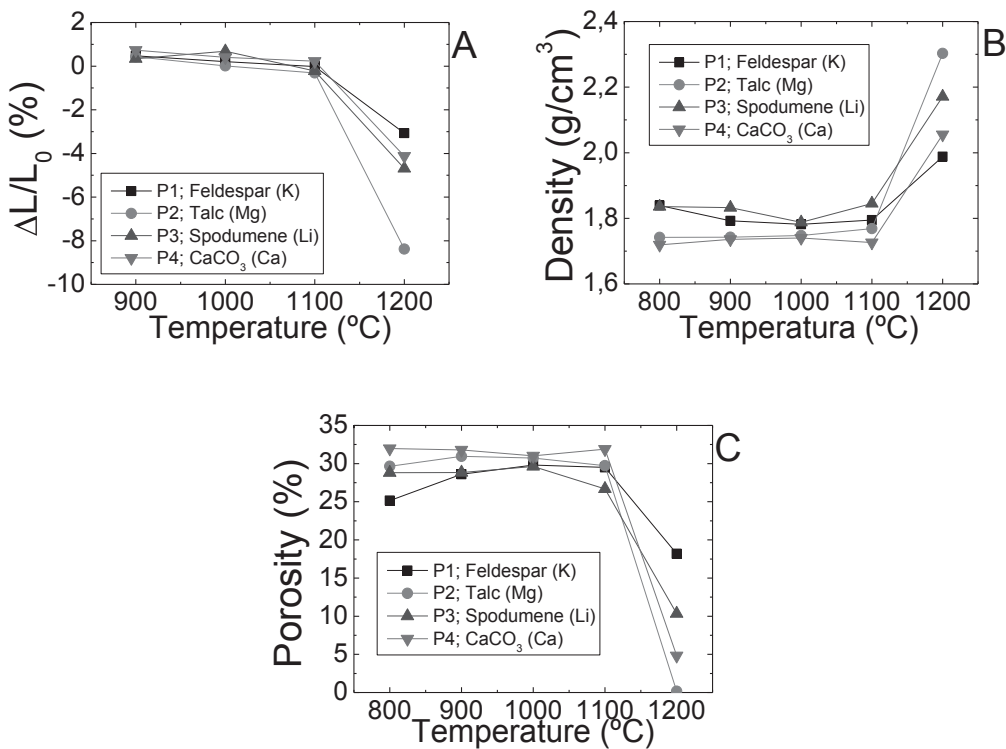


Fig.3. Thermal evolution of the sintering parameters: (A) Contraction (B) Density (C) Porosity.

It was found that below 1100° C the replacement of up to 15% of the flux does not vary with the sintering and mechanical properties. Therefore, the four materials studied can be still classified as earthenware if ware were fired at 1050 °C.

As shown in fig. 3 all additives selected favored the triaxial ceramic sintering. The sinterability found for this series could be ordered as follows:  $Mg \geq Ca \geq Li \geq K$ . Even zero porosity material obtained with the addition of talc sintered at 1200° C. P2 and P4 sintered at this temperature could be considered stoneware (porosity  $\leq 5\%$ ), while P3 would require a heat treatment a little higher and the composition of P1 reach porosities below 5% only treatments of 1280 ° C-1300 ° C.

Particularly, the addition of spodumene lowered the maturity temperature, the material prepared at 1100°C showed a slight decrease in porosity and with an increase in the mechanical properties ( $\sigma_f$  and E).

The mechanical properties ( $\sigma_f$ , E) thermal evolution is shown in fig. 4. As mentioned the mechanical properties of ceramics are strongly correlated with the degree of sintering, Carty and Senapati, 1998 (Fig. 1F and 1E), and the materials studied are not significantly different after heat treatment below 1000 ° C. While the mechanical strength are not significantly different for the materials prepared with different fluxes across the temperature range studied, the materials developed with the addition of spodumene (Li) and Talc (Mg) showed an increase in the modulus of elasticity after treatment 1100 ° C and 1200 ° C reaching double the value of triaxial porcelain used as a pattern (P1). This change in the modulus of elasticity could come to represent an improvement in thermal shock behaviour of these materials Rendtorff et al 2008A, Rendtorff et al 2008B. Studies of the thermal shock resistance of these materials are being carried out in our laboratory.

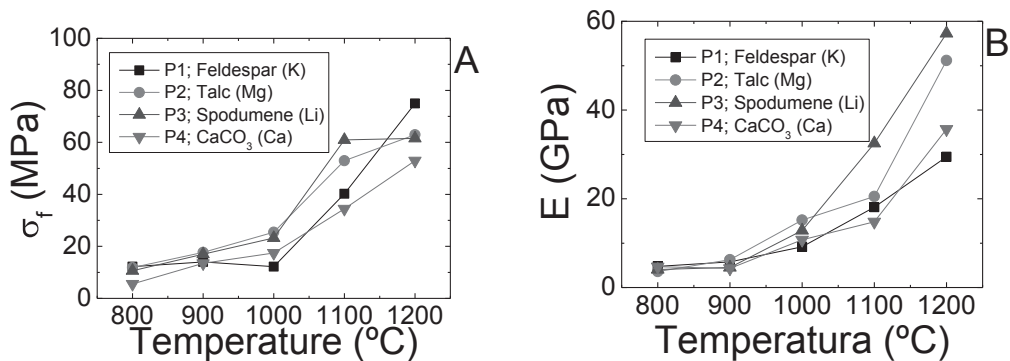


Fig. 4: A) Flexural Strength B) Elastic Modulus (E)

#### 4. Conclusions

The Triaxial ceramic were sinterized in a wide range of temperatures. These were characterized and compared in terms of sinterability, and mechanical properties. The influence of the chosen secondary flux was established.

The three studied secondary fluxes (Talc, Spodumene and Calcium carbonate) enhanced the sinterization of these materials. At Earthenware temperatures, below 1100°C, no effect was detected, showing that both the feldspar and other fluxes act only as filler at these temperatures. Above this temperature the addition of secondary flux influenced the sinterization and properties of the resulting material. The influence of the additives in the sinterization was established.

Showing that, the talc strongly enhanced the sinterization of these materials. Furthermore completely dense stoneware was achieved after 1200°C thermal treatments with this feldspar replacement. The effect of

CaCO<sub>3</sub> and spodumene was also enhancing the sinterability of the materials.

The observed drawback of talc was the influence in the plasticity behaviour of these materials, considerably shortening the plastic zone of the triaxial formulas. On the other hand the other two additives did not influence this important property.

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