MICROSTRUCTURAL DEVELOPMENT OF TRIAXIAL CERAMIC BODIES USING ALBITE OR SPODUMENE

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Abstract. *Traditional ceramics are mostly composed of mixtures of clay, quartz and feldspar. Depending on the mineralogical composition of the raw materials and the relative amounts in the triaxial mixture, porcelain, stoneware and such ceramics are produced. In these mixtures, feldspar introduces alkaline oxides such as Na_2O, K_2O or Li_2O, which act as flux, providing a liquid phase at elevated temperatures and thus promoting densification. Commonly, albite and orthoclase are used due their natural abundance. The current work compares the use of the lithium containing feldspar, spodumene, to an albite containing triaxial ceramic mass. Specimen were prepared by dry milling the starting materials, São Simão clay, quartz and feldspar (albite or spodumene) in a ball mill and uniaxial compaction in a rectangular steel die under a pressure of 28 MPa. The feldspar content varied from 15 to 30 wt-%. Sintering of the samples was conducted in an electrical furnace in air at temperatures of 1000, 1100, 1200 and 1250 ^0C, for 1 hour. The microstructure of the sintered samples was analyzed by scanning electron microscopy, SEM, of fractured surfaces. The phase analysis was conducted using X-ray diffraction. The results show that with increasing sintering temperature the amount of glassy phase formed increases and porosity decreases. Phase composition of the sintered samples is very similar. Considering though that the spodumene feldspar used contains less lithium oxide in comparison to the sodium oxide containing feldspar its higher efficiency as flux has been demonstrated.*

Keywords: *Feldspar, Lithium, Triaxial Ceramic, Sintering.*

1. INTRODUCTION

Traditional ceramics are obtained from natural raw materials to fabricate products with pre-defined characteristics (Sacmi, 2003). In Brazil, the participation of the ceramic industry in the gross domestic product (GDP) is estimated in 1%, corresponding to approximately size billion dollars. Due to the abundance of natural raw materials, alternative energy sources and availability of technologies, the Brazilian industry developed vapidly, producing and exporting high quality products (ABC, 2011).

The raw materials used to produce triaxial ceramic materials influence the properties of the final product such as porosity, density, fracture strength and surface quality. In this work, a ceramic mass was formulated based on clay, kaolin, quartz and feldspar. Each of these compounds holds a specific task in the sintering of traditional ceramic materials. Clay forms gradually a liquid phase during sintering of the ceramic articles, producing mechanical strength (Navarro *et al*., 1981 and Brusa, Contoli and Dardi, 1999). Kaolin increases the alumina content and helps to improve the whiteness of the final product (Rodrigues *et al*., 2004). Quartz participates in the fusion of the feldspar and equilibrates the viscosity of the liquid phase (Oliveira, 1998 and Drozda, 2003). Furthermore, feldspar decreases the sintering temperature, acting as flux, forming liquid phase and modifying the porosity of the pieces (Motta *et al.*, 2002 and Llorens *et al*., 2000). The importance of a flux in the in the ceramic wars consists in its capacity to lower the temperature of liquid phase formation during the process of sintering. The liquid formed tends to fill the voids of the ceramic body, reducing or eliminating the porosity (Worral, 1982).

The peculiarity of each of these compounds depending on its origin, require an adjustment of the formulation, or even of the compounds used. In this way it may be possible to explore minerals situated closely to the ceramic industry, reducing transport costs (Verduch and Balmaseda, 1995 and Mussolin and Doneda, 1997).

2. MATERIALS AND METHODS

2.1. Sample Preparation

The specimen was prepared by mixing the starting materials, quartz, kaolin, clay and feldspar in the proportions listed in table 1. The mixture 1 had a total weight of 100 g, the mixture 2 had 105 g, the mixture 3 had 110 g, and the mixture 4 had 115 g, the each mixture were made five specimens of 20 g, the rest of the mass was discarded.

Table 1. Sample composition

Material	Standard Feldspar (g)				Feldspar Containing Lithium (g)			
	Mixture 1	Mixture 2	Mixture 3	Mixture 4	Mixture 1	Mixture 2	Mixture 3	Mixture 4
Clay	40	40	40	40	40	40	40	40
Kaolin	10	10	10	10	10	10	10	10
Feldspar	15	20	25	30	15	20	25	30
Quartz	35	35	35	35	35	35	35	35

The chemical analysis of the Li-feldspar by X-ray fluorescence is listed in table 2, and the other compounds are listed in table 3.

Table 2. Chemical analysis of Li-feldspar (CIA Industrial Fluminense – MIBRA)

Compounds	wt %
SiO_2	76.36
Al_2O_3	16.09
Fe_2O_3	2.05
CaO	1.42
Na_2O	4.10
K_2O	2.31
MnO	0.15
TiO_2	0.0003
MgO	0.68
P_2O_5	0.14
Li_2O	0.91
LOI	0.35

Table 3. Chemical analysis of other compounds

Compounds	SiO_2 (wt%)	Al_2O_3 (wt%)	Alkalis*(wt%)
Na-feldspar	65.92	19.31	14.77
Kaolin	52.74	45.83	1.44
São Simão Clay	63.76	35.03	1.21
Quartz	99.23	0.50	0.27

* Contents of CaO + Na_2O + K_2O + MgO; the origin of raw material: MINASOLO Comércio e Representação Ltda., (2)UFSCar, (3)TALMAG PP-325 - Magnesita S.A.

The mixing of the powder was done by dry milling of the starting materials in a ball mill for 2h. The specimens were fabricated by cold uniaxial pressing in a steel die, under a pressure of 28.5 MPa for 30 seconds, SIWA Press PH 15 ton. For each composition studied, 20 specimens were pressed with dimensions of 70 x 20 x 7 mm and approximately 20g weight. After pressing, the green density of the samples was determined by measuring their volume and weight.

2.2. Sintering

The specimen were sintered at temperatures of 1000, 1100, and 1200°C, in an electrical heated furnace in air, using a heating rate of 3°C/min and an isothermal soaking time of 1hour at maximum temperature, Lavoisier Furnace Model 4020. After that, the oven was switched off, allowing natural cooling of the samples inside the furnace.

2.3. Characterization of the Sintered Samples

The sintered samples were characterized by the weight loss, linear shrinkage, absorption of water and porosity. Furthermore, the phase composition was determined by X-ray diffraction, and the microstructure observed by scanning electron microscopy, SEM, of fractured surfaces.

2.4. Analysis of the Microstructure

The microstructure of each sample composition and sintering condition was investigated by observation of the fractured surface in a scanning electron microscope, SEM, HITACHI

3. RESULTS AND DISCUSSION

No significant differences of the microstructures have been observed by scanning electron microscopy (SEM) between samples prepared with Li-containing feldspar and samples containing the Na-containing feldspar see Fig. 8 to 11. In all compositions studied, and even after sintering at 1200^{o}C, large faceted particles with a size of up to 70µm have been observed, probably residual quartz. Furthermore, it can be noted that with increasing sintering temperatures a glassy phase is formed, which exhibits a smooth fracture surface. With increasing sintering temperature and increasing feldspar content of the samples higher amounts of glassy phase are detected. The formation of the glass phase is more pronounced in samples sintered at 1200^{o}C or higher, due to the reactions between feldspar, quartz and meta-kaolinite.

With increasing amount of glassy phase formed the fractured surface is smoother, still exhibiting residual porosity, indicating that densification during sintering has not been completed, even when using 30% of feldspar in the triaxial mixture and sintering at 1200^{o}C.

The phase analysis of the sintered samples conducted by X-ray diffraction analysis revealed the presence of quartz in all samples, independent of the composition or sintering temperature. This indicates that the quartz particles used in the starting mixture did not completely react to form a liquid phase during sintering, even after sintering at 1200^{o}C. Furthermore, Mullite has been formed in the samples sintered at 1200ºC or higher temperature by the reaction of feldspar and spinel.

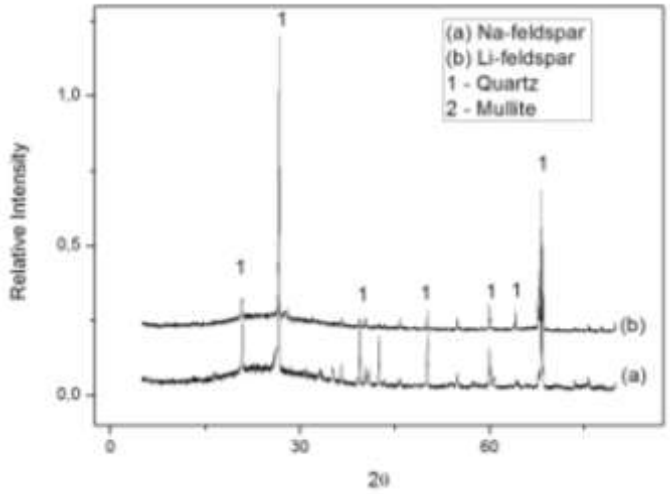


Figure 1. X-ray diffraction diagram of samples sintered at 1000°C, containing 15 % feldspar, (a) Na-feldspar and (b) Li-feldspar

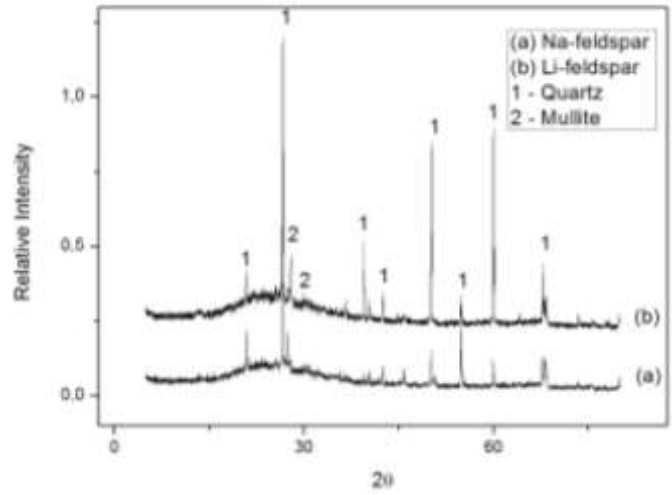


Figure 2. X-ray diffraction diagram of samples sintered at 1000°C, containing 30 % feldspar, (a) Na-feldspar and (b) Li-feldspar

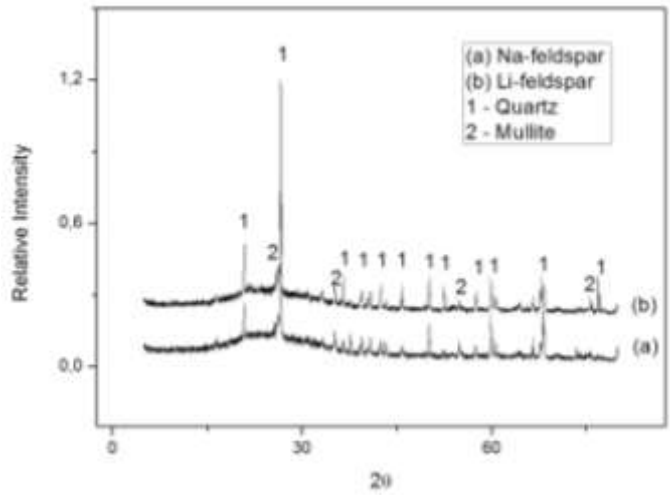


Figure 3. X-ray diffraction diagram of samples sintered at 1200°C, containing 15 % feldspar, (a) Na-feldspar and (b) Li-feldspar

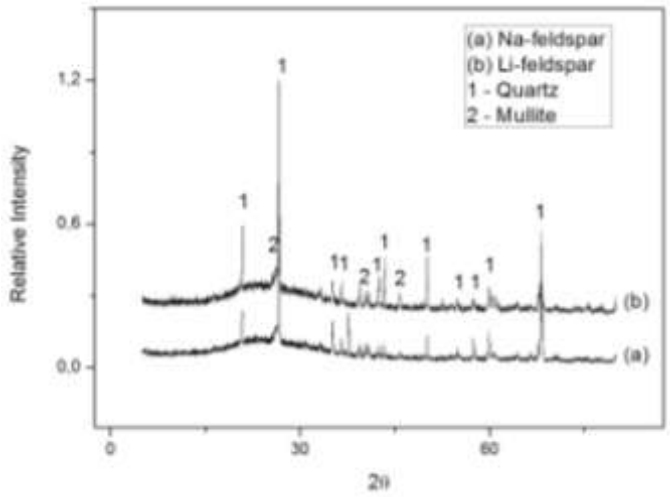
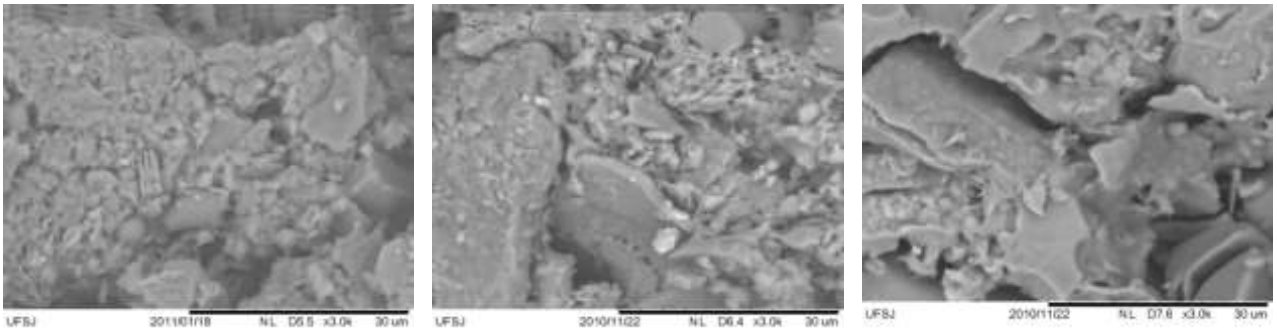
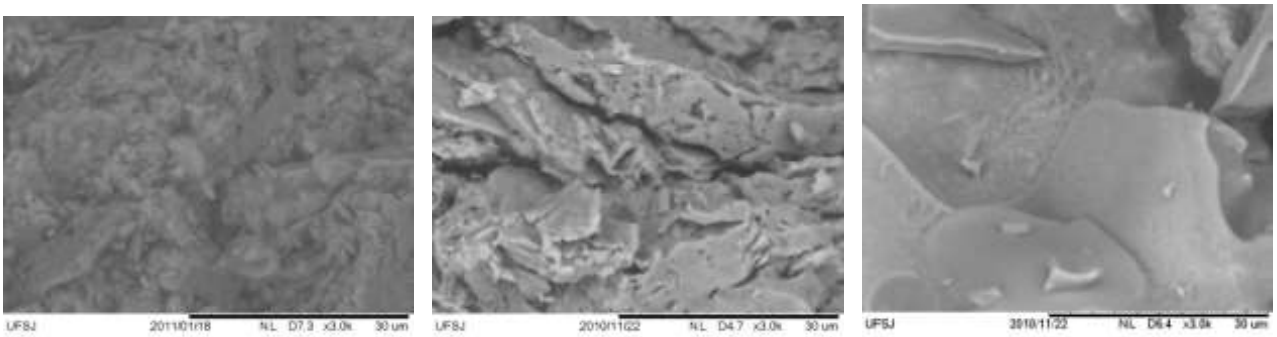


Figure 4. X-ray diffraction diagram of samples sintered at 1200°C, containing 30 % feldspar, (a) Na-feldspar and (b) Li-feldspar



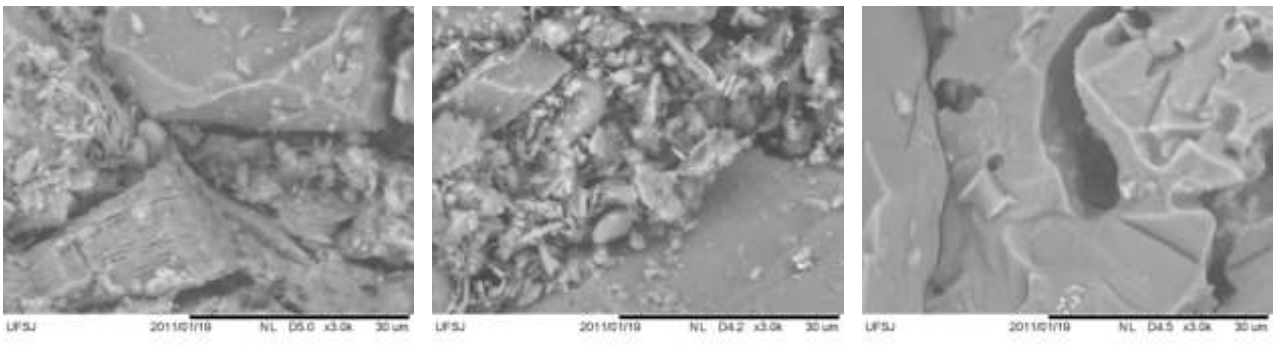
i) ii) iii)

Fig. 5: Micrograph of fracture observed by scanning electron microscopy of samples sintered at i) 1000, ii) 1100 and iii)1200°C, containing 15 % Na-feldspar



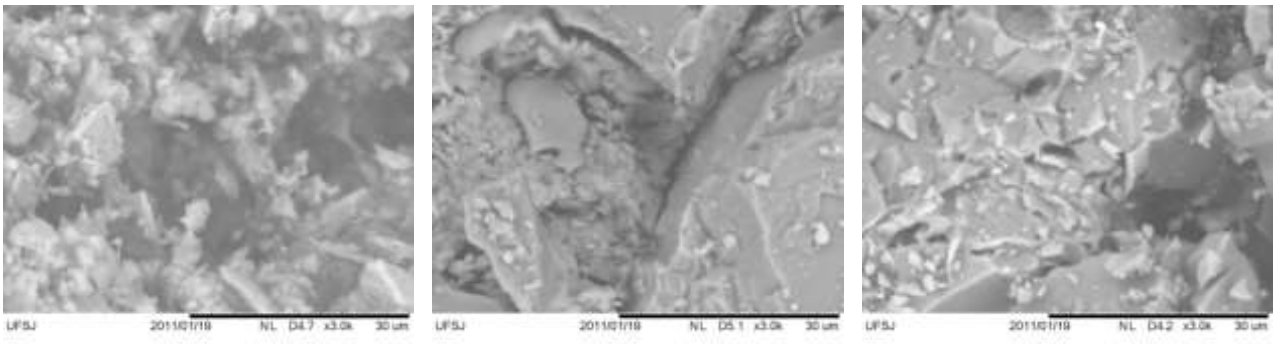
i) ii) iii)

Figure 6. Micrograph of fracture observed by scanning electron microscopy of samples sintered at i) 1000, ii) 1100 and iii) 1200°C, containing 15 % Li-feldspar



i) ii) iii)

Figure 7. Micrograph of fracture observed by scanning electron microscopy of samples sintered at i) 1000, ii) 1100 and iii) 1200°C, containing 30 % Na-feldspar



i) ii) iii)

Figure 8. Micrograph of fracture observed by scanning electron microscopy of samples sintered at i) 1000, ii) 1100 and iii) 1200°C, containing 30 % Li-feldspar

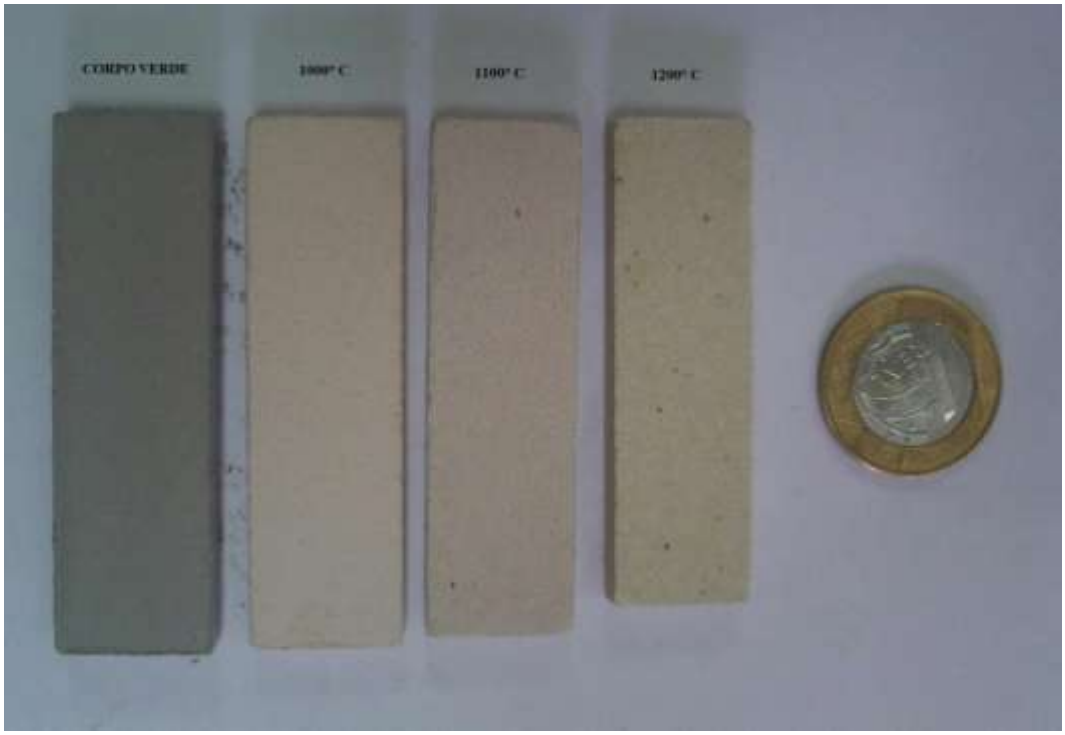


Figure 9. Photographs of samples containing Li-feldspar, before and after sintering at 1000, 1100 and 1200°C, showing coloring and retraction.

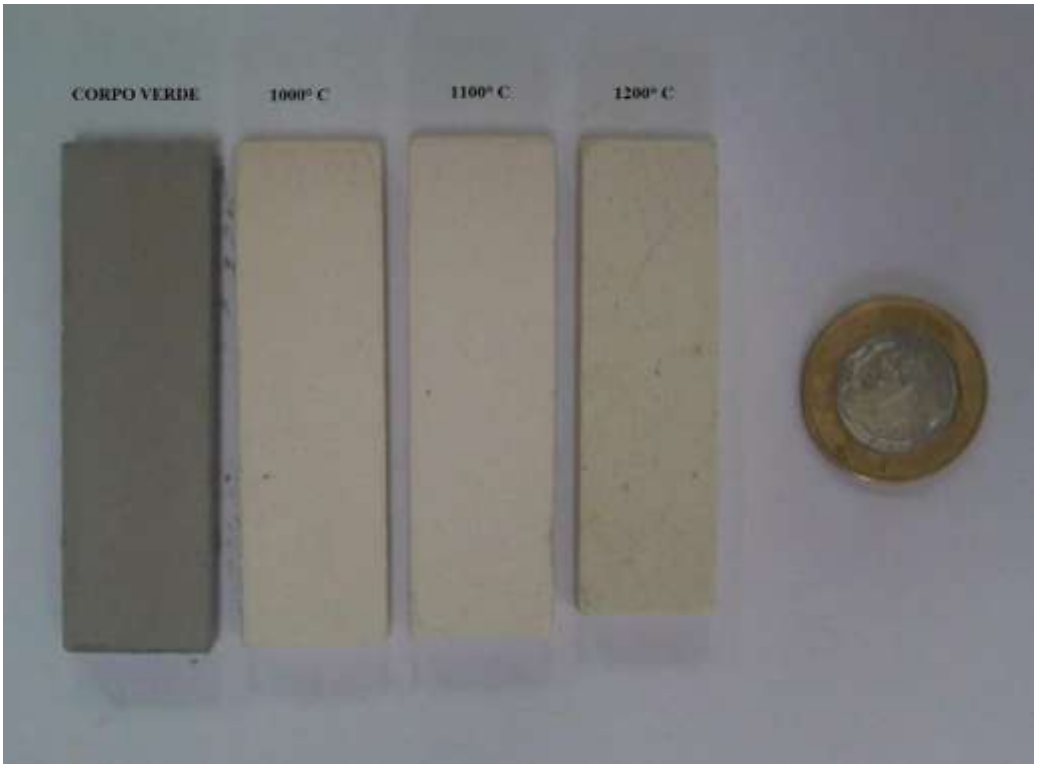


Figure 10. Photographs of samples containing Na-feldspar, before and after sintering at 1000, 1100 and 1200°C, showing coloring and retraction.

Comparing the samples after sintering, as shown in the photographs shown in Fig. 12 and 13, samples containing Na-feldspar exhibited a brighter coloring in comparison to the samples prepared with Li-based feldspar, specifically at higher temperatures of 1200C.

4. CONCLUSIONS

In order to improve the densification during sintering we suggest to use smaller quartz particles and/or increase the sintering temperature to 1300°C or higher. In this way the diffusion paths are shortened and the diffusion coefficients increased, which should result in a higher densification.

No significant differences between the samples prepared with Na-feldspar and the samples with Li-Feldspar have been observed, confirming the possibility to substitute Na-feldspar by the Li containing feldspar in the triaxial mixtures, without prejudice for the properties of the sintered materials.

5. REFERENCES

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