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izvorima električne energije**

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Electrical Power Sources**



Beograd, 15. oktobar 2021
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**Predsednik Društva za
obnovljive izvore
električne energije
pri SMEITS-u**

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UTICAJ VREMENA SINTEROVANJA NA GUSTINU I SEM ANALIZA KORDIJERITNE KERAMIKE

INFLUENCE OF SINTERING TIME ON DENSITY PROPERTIES AND SEM ANALYSIS OF CORDIERITE-BASED CERAMICS

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Mehanička aktivacija početnih smeša kordierita ($2\text{MgO}\cdot 2\text{Al}_2\text{O}_3\cdot 5\text{SiO}_2$) sa 5.00 mas. % TiO_2 izvršena je u visokoenergetskom mlinu sa kuglama tokom 10-80 min. Primenjeni pritisak presovanja pre procesa sinterovanja bio je 2 t/cm^2 . Proces sinterovanja je izveden na $1350\text{ }^\circ\text{C}$ tokom 2h i 4h u vazdušnoj atmosferi. Skenirajuća elektronska mikroskopija je izvršena u cilju analize mikrostrukture presovanih i sinterovanih uzoraka. Za ispitivanje površine sinterovanih uzoraka korišćen je mikroskop atomskih sila. Ovaj rad istražuje uticaj produženog vremena sinterovanja na gustinu sinterovanih uzoraka zajedno sa električnim svojstvima.

Ključne reči: mehanička aktivacija; gustina; SEM; kordijerit

The mechanical activation of the starting mixtures of cordierite ($2\text{MgO}\cdot 2\text{Al}_2\text{O}_3\cdot 5\text{SiO}_2$) with 5.00 mass % TiO_2 was performed in a high energy ball mill during 10-80 min. The applied compaction pressure before the sintering process was 2 t/cm^2 . The sintering process was performed at $1350\text{ }^\circ\text{C}$ for 2h and 4h in air atmosphere. Scanning electron microscopy was performed to analyze the microstructure of both compacted and sintered samples. Atomic force microscope was used to investigate the surface of the sintered samples. This paper investigates the influence of prolonged sintering time on the densities of the sintered samples, along with electrical properties.

Key words: mechanical activation; density; SEM; cordierite

1 Introduction

Cordierite-based ceramic materials are attracting much interest for their various applications in industry, for manufacturing multilayer circuit boards, catalytic converters, filters, thermal insulation, kiln furniture, components of portable electronic devices, etc. In order to reduce production costs and modify cordierite-based materials, mechanical activation can be used. In this study, microstructural and electrical properties of mechanically activated $\text{MgO-Al}_2\text{O}_3\text{-SiO}_2$ system have been analyzed. Owing to the very low temperature thermal expansion coefficient ($20\times 10^{-7}/^\circ\text{C}$) and low relative dielectric constant (~ 5), these ceramics are also well known by their good thermo-mechanical, chemical and dielectric properties [1, 2]. They can be applicable as materials that are exposed to sudden temperature changes [3-7] and also as a semiconducting bearer [8, 9]. The temperature range of cordierite sintering is very narrow ($1300\text{-}1400\text{ }^\circ\text{C}$).

Our previous investigations showed a significant influence of mechanical activation, as well as compaction pressure on starting mixtures (kaolin, quartz, magnesium oxide) on lowering of sintering temperature [10]. Mechanically activated components increased energy due to induced crystal defects compared to non-activated ones. During mechanochemical treatment, several processes occur: attrition of starting material, crystal lattice destruction, various defects formation, etc. All mentioned processes increase the chance that during heating, processes could be observed at lower temperatures than usual [11]. Furthermore, mechanical activation could affect the final electrical characteristics,

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so it is very important to approach and understand changes that get introduced into the system while milling.

In this paper, the authors used some starting conditions based on the previous investigation and tried to study the influence of sintering time on density, phase composition as well as on the microstructure along with electrical properties of sintered samples.

2 Experimental procedure

Mixtures of Mg(OH)₂, Al₂O₃, SiO₂ and TiO₂ (all p.a. purity) were used in these experiments. The cordierite ceramics starting mixture, MgO+Al₂O₃+SiO₂ in the 2:2:5 ratio, with the addition of 5.00 mass% TiO₂, were mechanically activated by grinding in a high-energy planetary ball mill. ZrO₂ vessels and balls were used with the powder to balls mass ratio of 1:40. The milling process was performed in air atmosphere for 10, 20, 40 and 80 minutes.

The pressure used in our experiments was 2 t/cm² (approximately 200 MPa). The pressure was performed in a double-sided tool 6 mm in diameter (Hydraulic press RING 14, VEB THURINGER). The density of the specimens was calculated from measurements of their diameter, thickness, and mass. The theoretical density (TD) of mixture is 3.078 g/cm³ and is calculated based on the following equation (eq. 1):

$$\rho = \frac{m}{V_1+V_2+V_3+V_4} \quad (1)$$

where: m – mass of the mixture (30.00 g), V_1 , V_2 , V_3 and V_4 – volumes of component 1 – Mg(OH)₂, component 2 – SiO₂, component 3 – Al₂O₃ and component 4 – TiO₂ in the mixture, calculated by usage of TD of each component.

The compacts were sintered isothermally at 1350 °C, in air atmosphere for 2h and 4h, with heating rate of 10 °C/min.

3 Results and discussion

Mechanically activated samples of cordierite with the addition of 5mass% TiO₂ were sintered for 2 and 4 hours at 1350 °C. The density changes of the samples before and after sintering process were investigated. Density values of samples before and after sintering are shown in Table 1 for samples sintered for 2 hours, and in Table 2 for samples sintered for 4 hours. The results shown in Tables 1 and 2 are consistent with the expected results. The density of the samples was higher than the density of the starting material, also the density increased with prolonged sintering time.

Table 1. Mass and densities of green bodies and 2h sintered cordierite samples with addition of 5 mass % TiO₂

Activation time (min)	* $m_o(g)$	$m_s(g)$	** $\rho_o(g/cm^3)$	$\rho_s(g/cm^3)$
0	0.2980	0.2845	2.001	1.990
10	0.2991	0.2845	2.070	2.145
20	0.2978	0.2839	2.048	2.178
40	0.2986	0.2845	2.039	2.184
80	0.2982	0.2856	1.962	2.168

In order to compare the results of changes in the density of samples before and after the sintering process, density variation $\Delta\rho$, ($\Delta\rho = \rho_s - \rho_o$), as a function of mechanical activation time for both sintering time, 2 and 4 hours, was analyzed.

In the first 20 min of mechanical activation, the major changes are taking place in the starting powder material. It causes significant fragmentation and homogenization of the starting ingredients. Thus the activated samples during the sintering process show faster chemical reaction in the solid state and the density curve of the sintered samples as a function of activation time shows that the length of time of mechanical activation significantly increases the density of the obtained material.

Samples sintered for 4 hours have a higher change in density than samples sintered for 2 hours. Samples sintered 2 and 4 hours exhibit changes in density sharply in the first 20 minutes of mechanical activation. After 20 min of activation time, density changes are still moving to higher values, but slower.

Table 2. Mass and densities of green bodies and 4h sintered cordierite samples with addition of 5 mass % TiO₂

Activation time (min)	*m _o (g)	m _s (g)	**ρ _o (g/cm ³)	ρ _s (g/cm ³)
0	0.2980	0.2840	1.975	2.011
10	0.2978	0.2835	2.072	2.160
20	0.2980	0.2843	2.035	2.204
40	0.2982	0.2842	2.050	2.202
80	0.2983	0.2853	1.968	2.178

*m_o, m_s – mass of sample before and after sintering process

**ρ_o, ρ_s – densities of sample before and after sintering process

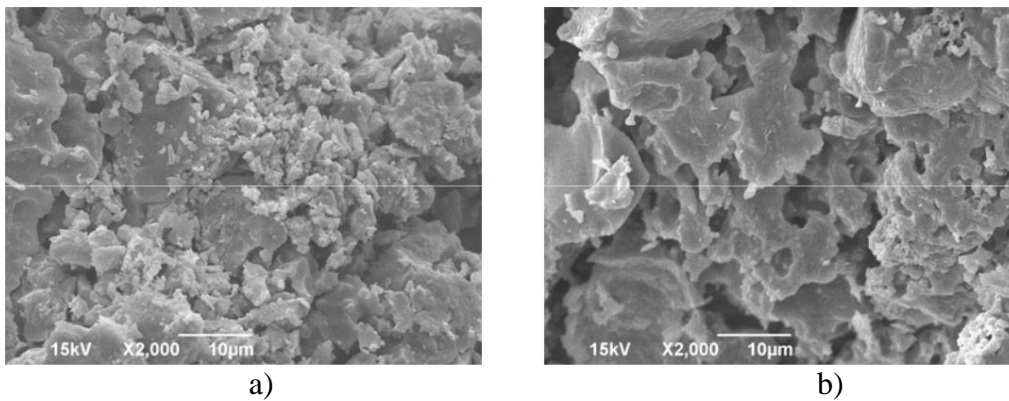


Figure 1. SEM of non-activated samples sintered at 1350 °C for a) 2h and b) 4h

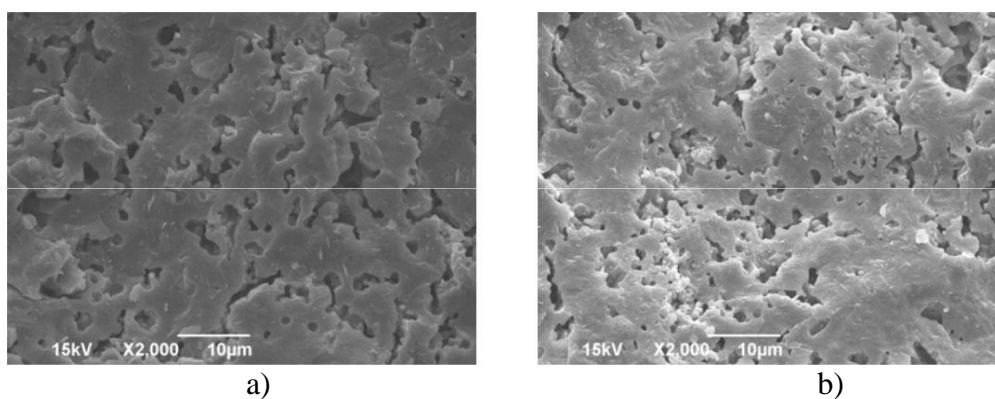


Figure 2. SEM of mechanically activated samples for 80 minutes sintered at 1350 °C for a) 2h and b) 4h

Figure 1 represent scanning electron micrographs of sintered powders. Non-activated powder sintered for 2 hours (Figure 1a)) shows fine-grained structure, defined grain sintered material, and high porosity between grains. The surface is rough. The particles of the starting material were homogenized, but the extension of time of sintering (Figure 1b)) at a temperature of 1350 °C leads to further convergence of grain, and the reaction sintering process allows better contact of the starting particles. This leads to the formation of larger agglomerates. The porosity of the obtained product is

lower than the previous sample sintered for 2h, but still very high. Defined grains are much less conspicuous. Blocks of agglomerates are formed, due to a longer sintering time. Samples which were mechanically activated for 80 minutes showed a significantly smoother texture.

Porosity was significantly smaller, and was practically a negligible difference in the appearance between the surface of samples sintered for 2 and 4 hours (Figure 2a) and b)). The defined grain of the starting powder material flow is not visible. Based on the SEM images, we can see a significant influence of mechanical activation on the sintering process, and the impact of the length of the sintering process on the cordierite sample set.

4 Conclusion

Investigation results shows that sintering time, as well as mechanical activation, has influence on the final characteristics of the sintered material. The results showed that longer time of sintering have effect on final product.

Longer sintering time increases the density variation of the sintered samples. The time of mechanical activation has also influence on the density changing of the samples, and increases with increasing sintering time.

SEM results shows that samples which were mechanically activated for 80 minutes showed a significantly smoother texture. Porosity was significantly smaller than non-activated samples surface, and was practically a negligible difference in the appearance between the surface of samples sintered for 2 and 4 hours.

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