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Original scientific paper

XRD ANALYSIS OF ACTIVATED FOUR-COMPONENT CERAMICS

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Abstract: The aim of the presented research was to analyze ceramic material based on cordierite as a function of activation time and sintering temperature. Three-component oxide mixture was prepared (MgO + Al2O3 + SiO2 in the ratio 2:2:5). To decrease the sintering temperature, 10 mass % Bi2O3 was added to this mixture. The mixtures were mechanically activated for 5 and 240 minutes in a ceramic ball mill. Activated mixtures were sintered at temperatures of 1173-1573K. XRD method was used to determine the structural transformations of the obtained products.

Keywords: cordierite, activation, XRD

1 INTRODUCTION

Cordierite ceramics are formed during the sintering process in a narrow temperature "window" (Δt> 283K) at temperatures above 1573K (Obradović et al. 2016; Obradović et al. 2019a). Cordierite has a low coefficient of expansion, good mechanical properties and a low dielectric constant. It is most widely used in places of sudden temperature changes. In order to lower the sintering temperature, bismuth oxide was added to the basic cordierite mixture, and mechanical activation was performed to determine its influence on the formation of this electronic ceramic.

The activation of materials is widely used as a procedure for bringing energy to the system so that it begins to react in a controlled manner. This allows the system to skip over the "energy barrier" on the reaction path (Baláž, 2003; Maciá-Agulló et al. 2004; Wang et al. 2005; Castro et al. 2004). The method of mechanical activation, which, in

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various technical and technological ways, brings mechanical energy to solid materials thus changing and regulating many physico-chemical characteristics related to increasing the system's reactivity, is becoming more and more topical (Berbenni et al. 2001; Xue et al. 2002; Praveenkumer et al. 2008).

Additives can decrease the reaction temperature during sintering. Application of additives should improve the contact between reacting components during sintering process. Additive with lower melting point than reacting components temperature should enable the formation of the liquid phase in the system, and improve the contact between mixture components. It is also important that the additive ionic radius should be high enough not to upgrade itself into the crystal structure of cordierite (Kumar et al. 2014; Maurya et al. 2009; Obradović et al. 2019b).

2 EXPERIMENTAL PROCEDURE

In presented research the technical quality oxides MgO (purity 98.60%, Euro Hemija, Beograd), Al2O3 (purity 99.19%, Aluminijumski kombinat Podgorica), SiO2 (purity 96.10%, Bela Reka) and Bi2O3 (purity 99.98%, Реахим) were used. Bi2O3 have the melting point near 1093˚C. The mixture of MgO+Al2O3+SiO2 content in 2:2:5 ratio, with addition 10% Bi2O3, was mechanochemically activated. Activation periods were 5 and 240 minutes (mixtures designated B1 and B6) in cylindrical ceramic mill with balls (VEB, model 13x10,5''). Table 1. presents the samples designations and corresponding milling times.

Activated samples were prepared as the tablets of radius of 8mm and 4mm high, obtained with pressure of 1 t/cm^2. Samples activated for 5 minutes were sintered at the temperatures of 1273K and 1373K (sample B1). Samples of 240 minutes activation period were sintered at the temperatures of 1173K, 1273K, 1373K, 1473K and 1573K (samples B6).

Sintered samples were analyzed using X-ray diffraction "Philips" XRD model PW-1710, with curved graphite monochromatic and scintillated counter. Intensities of diffracted CuK$_\alpha$ X-ray radiation (λ=11.54178Å) were measured on the room temperature in 0.02 2θ intervals and 0.25s time intervals in range of 5˚ to 85˚ 2θ. Rö tube was working at the 4kV voltage and at 3mA current, with collimator angle of 1˚ and with 0.1mm.

Table 1 Milling times and sample designations

Sample	Milling time
B1	5 min
B6	240 min

3 RESULTS AND DISCUSSION

XRD analysis of the experimental cordierite mixture activated 5 minutes and sintered at 1273K (B1/1273K) did not detect the presence of cordierite. XRD analysis of the same sample sintered at the 1373K (B1/1373K) detected the presence of the indialite (cordierite modification) traces. Larger amounts of indialite were detected in samples activated 240 minutes and sintered at 1373K (B6/1373K). Both XRD diffractograms of the B1, B6 samples sintered at 1373K are presented on the Figure 1.

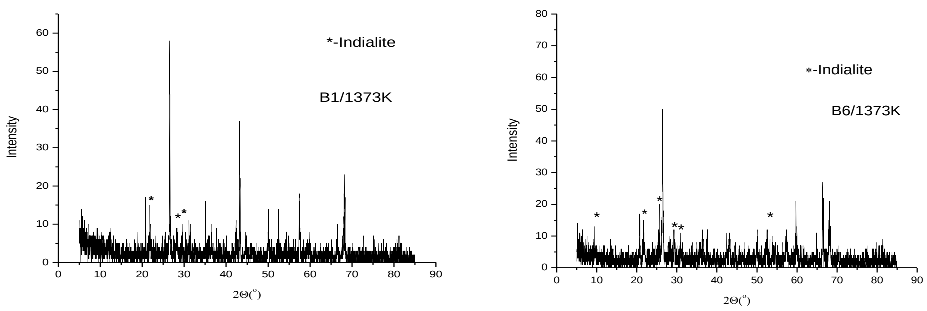

Figure 1 XRD analysis of samples B1(5 min) / 1373K and B6 (240 min) / 1373K

Cordierite is formed at the sintering temperatures above 1573K with no additives, so the influence of the mechanochemical activation and additive due to the obtained results is clearly visible. The activation decreases the size of the mixture particles and increases the free active particle surface. This has direct influence on the kinetics of sintering.

The kinetics of cordierite forming in the experimental samples obtained at higher temperatures was analyzed. XRD diffractogram of sample sintered at temperatures 1473K to 1573K, where even more amounts of indialites were detected, are presented at the Figure 2 and 3.

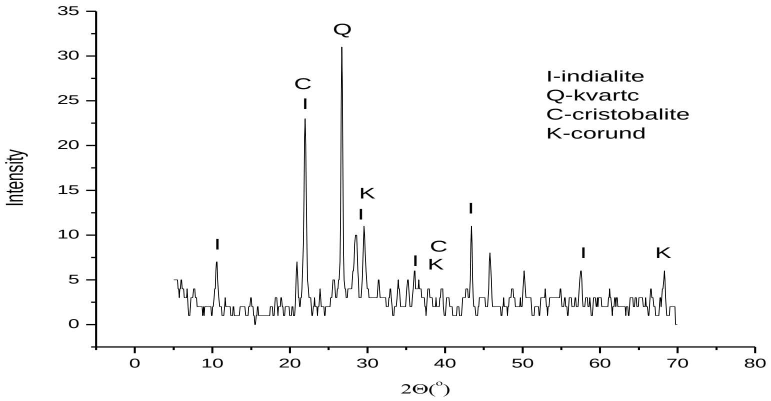

Figure 2 XRD diffractogram B6 (240 min) / 1473K

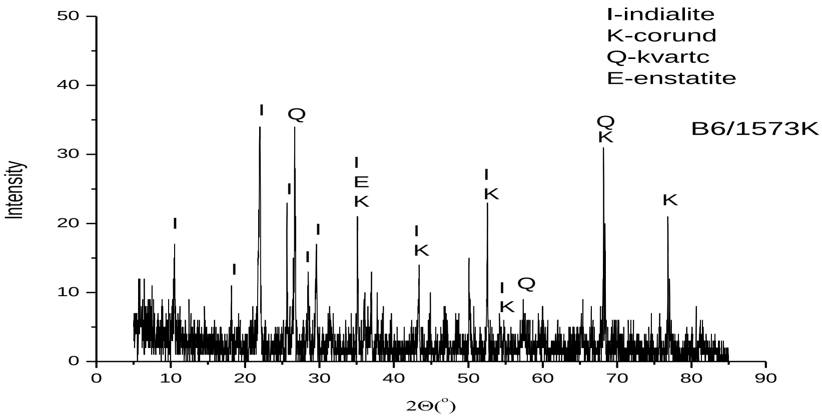

Figure 3 XRD diffractogram B6 (240 min) / 1573K

The performed experiments proved the stated assumption that mechanochemical activation has influence on cordierite forming temperature.

4 CONCLUSION

Presented research was done in purpose to investigate the effect of mechanical activation and sintering temperature on cordierite ceramics. Analysis presents the traces of intialite (cordierite modification) at the sintering temperature of 1373K. Higher amounts of intialite were detected at the same sintering temperature and activation period of 240 minutes at the same sintering temperature for the same activation time. Amount of produced inialite increase due to sintering temperature increase, and the highest gradient is between 1373K and 1573K. The obtained results prove the existence of the activation time influence on the formation temperature of cordierite ceramics.

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