The influence of ZrB_2-SiC powders mechanical treatment on the structure of sintered ceramic composites

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Abstract. The effect of mechanical treatment by planetary ball milling on the properties of hot pressed ZrB_2 - SiC ceramics was studied. It was shown that material densification after mechanical treatment is finished at initial stages of sintering process. Addition of SiC leads to an essential increase of sample density to 99% of theoretically achievable for powder with 2% of SiC, as compared with ZrB_2 with the density less than 76%. It was demonstrated that all defects that were accumulated during mechanical treatment are annealed during hot pressing, and there are no changes of CDD values in sintered ceramics.

1. Introduction

The refractory compounds are the basic components of materials used in high-temperature engineering, such as thermal protection of space vehicles, electronics, etc. Among materials with high melting temperature, a special attention is paid to ceramic ZrB_2-based composites; however, strong covalent bonds are the reason of low diffusion mobility of an atom, which appreciably impedes the manufacturing of products.

It is known [1], that addition of SiC to ZrB_2 increases the density of sintered materials due to smaller melting temperature of SiC. Besides, ZrB_2-SiC ceramics are characterized by high thermochemical stability, including high stability to oxidation in conditions of super-high temperatures.

The ZrB_2-SiC composites are usually obtained by sintering powders under pressing at temperatures higher than 2000 ºC [2], and to lower the sintering temperature, the powders undergo mechanical treatment in high-energy (planetary) mills. In this case, subsequent sintering will be activated due to increased number of defects, acceleration of diffusion processes and simplification of plastic flows during sintering [3], so sintering can be carried out under pressing, i.e. it is possible to realize hot pressing or SPS-process [4].

Unfortunately, data on the influence of mechanical treatment on the properties of powders and the process of subsequent hot pressing are poorly investigated. Therefore, the main aim of this paper is to study the influence of mechanical treatment of ZrB_2-SiC powders on their properties and properties of ceramic composites sintered by hot pressing.

2. Materials and Experimental Procedure

The research was carried out using powder mixtures of ZrB_2 (d_{50} = 2.5 μm) and SiC (d_{50} = 4.2 μm) with SiC content of 10, 15 and 20 vol.%. The powders were mechanically treated in a planetary mill at the centrifugal acceleration of approximately 30g. The duration of processing was up to 20 minutes.

Hot pressing of ceramic composites was carried out at the temperature of 1800 °C and pressure of 50 MPa with isothermal sintering for 30 minutes. According to [5], X-ray with CuKα source was used to measure raw density of mixtures, phases, structure and coherently diffracting domains (CDD). Scanning electron microscope observations operated at 20-30 kV were used to determine the structure and average grain size using Tescan VEGA-3SBH.

3. Results and Discussion

Figure 1 shows the dependence of relative density ρ_r/ρ_t (ρ_r raw density, ρ_t theoretical density) vs. treatment time. Evidently, for any powder, the increase of treatment time was accompanied by increase of its raw density; in addition, the morphology of particles has appreciably changed, from separate particles in the beginning state up to the formation of agglomerates in the end of treatment (Figure 2). Before the treatment, ZrB_2 powder had the smallest raw density; however, after processing, its density has increased essentially and exceeded the raw densities of powder mixtures with SiC. These mixtures in initial state had very close values ρ_r/ρ_t, but after the treatment, the powder with the maximal content of SiC had higher raw density. The X-ray phase analysis of mixtures has shown that during the treatment there were no changes; addition of SiC to the mixture leads to the occurrence of its peaks. With increased treatment time, we have found a broadening of peaks due to increasing number of lattice defects and decrease of CDD or grain size from 46 down to 37 nm (Figure 3).

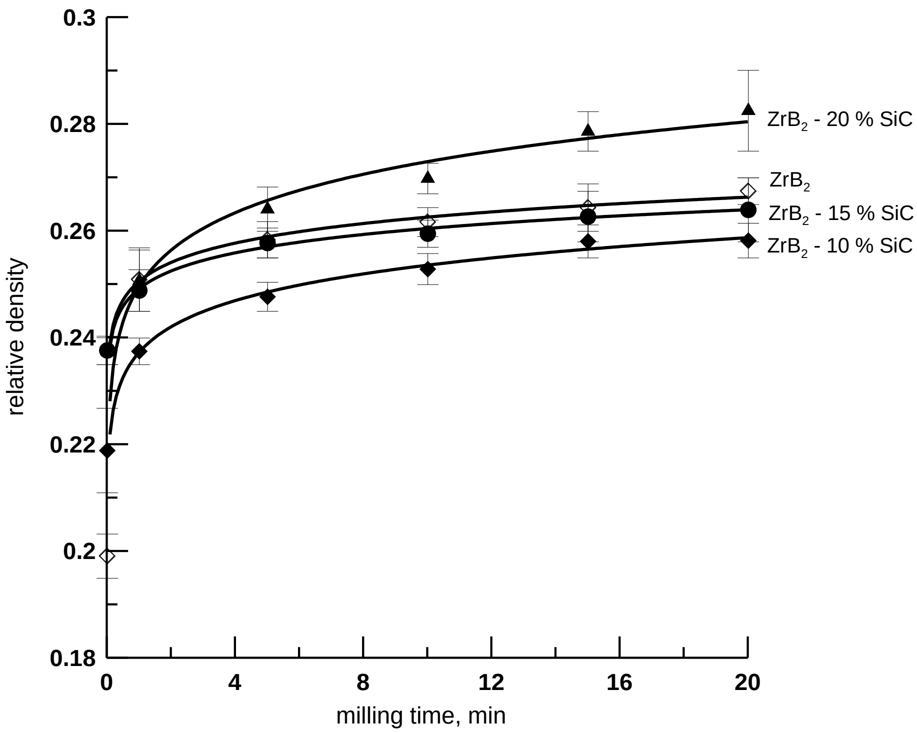
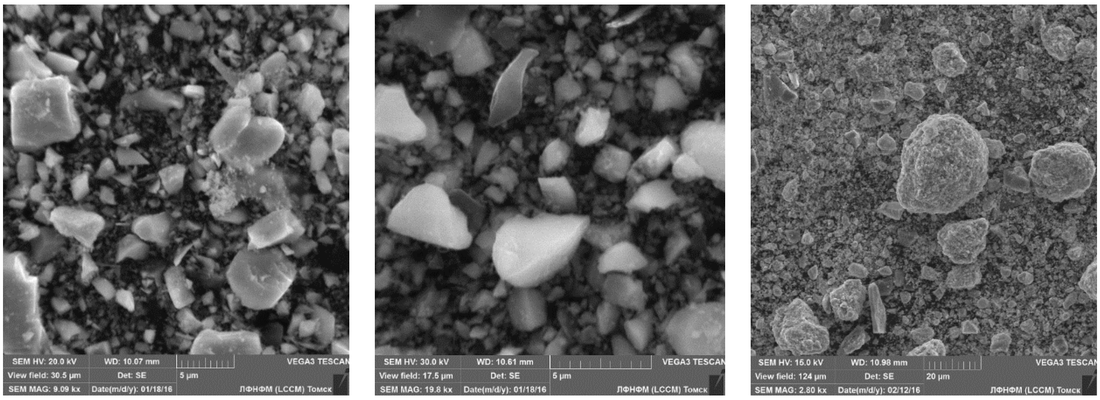


Figure 1. The dependencies of raw density of ZrB_2–SiC powders after mechanical treatment in planetary mill

After hot pressing of ceramics, the phase content did not change, and the increase of treatment time in the planetary mill before sintering have no effect on CDD of sintered materials (Figure 3). This means that all defects are annealed during hot pressing process. In Figure 4, the densities of hot pressed samples are shown. Obviously, addition of SiC leads to essentially increased sample density: its value goes up to 99% of a theoretical one for a powder with 20% of SiC, as compared to ZrB_2 going not higher than 76%. This agrees well with data of [3].

IOP Conf. Series: Materials Science and Engineering **140** (2016) 012006 doi:10.1088/1757-899X/140/1/012006

The dependencies of relative density change in sintered materials are well described by a simple function like $Y = aX^n$, where parameter n characterizes the speed of density change. This can be easily determined by re-plotting these dependences in log-log coordinates. Then, the inclination of a straight line will be equal to this parameter. Thus determined parameter "**n**" is shown in Figure 5, and as one can see, the addition of SiC to the mixture leads to up to four-fold decrease of its value. Hence, beside the sample increased density after hot pressure, the material densifies at initial stages of the sintering process.



Original 10 min 20 min

Figure 2. Change of powder morphology during milling

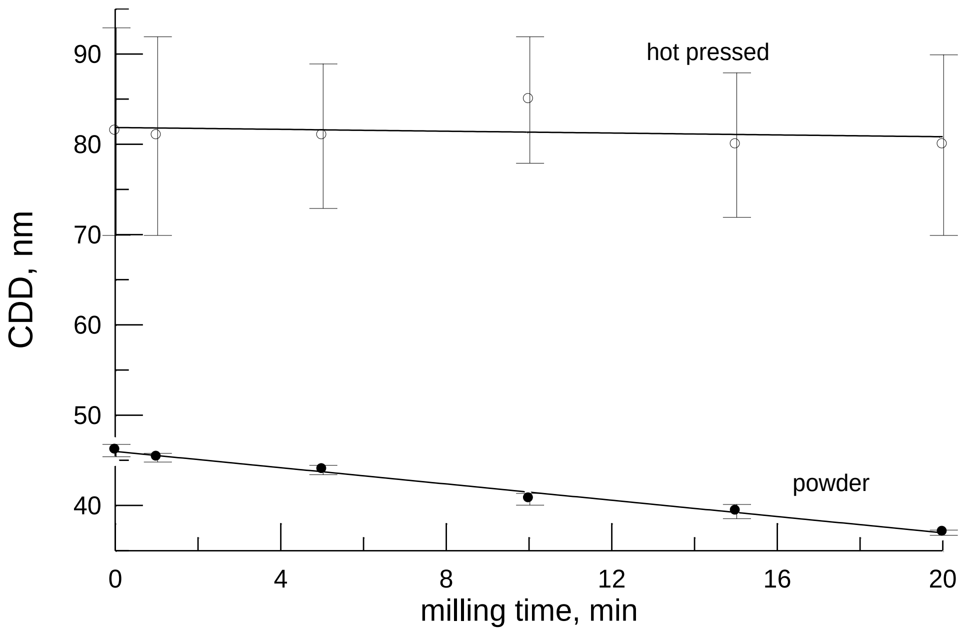


Figure 3. CDD vs. milling time of powder and sintered ceramics.

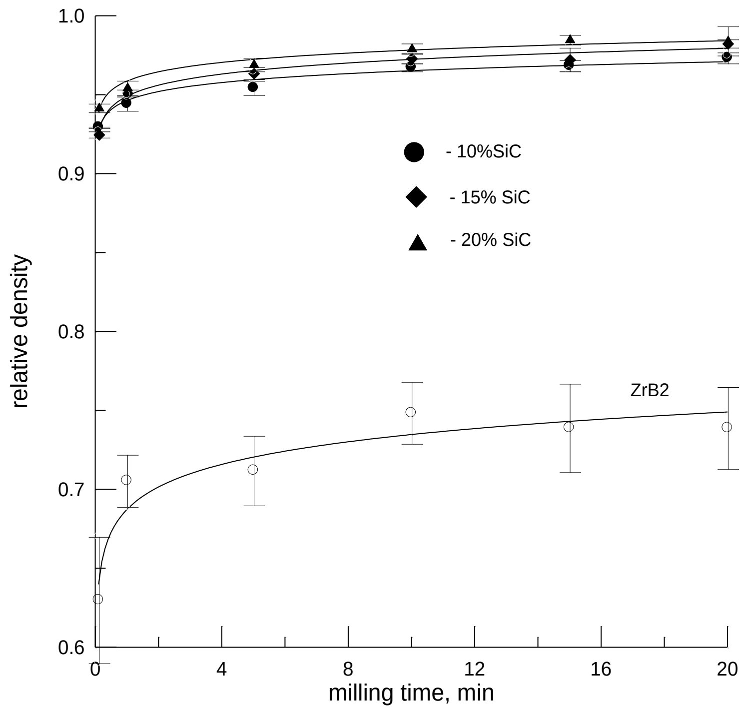


Figure 4. Relative density ρ/ρ_t of hot pressured ceramics vs. milling time of powder.

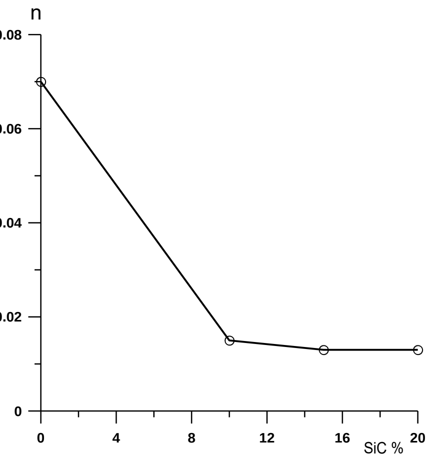


Figure 5. The dependence of parabolic index n of sintering process on SiC content in powder

4. Summary

It was demonstrated that material densification after mechanical treatment is finished at initial stages of sintering process. Addition of SiC leads to essentially increased sample density up to 99% of theoretical value for powder with 20 % SiC, as compare to ZrB_2 with the density not higher than 76%. It was shown that all defects that were accumulated during mechanical treatment are annealed during hot pressing process, and there are no changes of CDD values in sintered ceramics.

Acknowledgement

This work was financially supported by the Ministry of Education and Science of the Russian Federation in the framework of Agreement No. 14.607.21.0056 (RFMEFI60714X0056).

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