Experience of ceramic production from silicon carbide with the addition of SiC nanopowder

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Abstract. The effect of silicon carbide nanopowder additives on the properties of armor ceramics from this compound has been investigated, and the results are presented. The study reveals that the powder introduction in an amount from 5 to 10 wt. % leads to an increase in hardness and crack resistance when density is maintained. It enabled us to obtain the material which can be used in structural and armored ceramics.

1. Introduction

Oxygen-free refractory compounds are characterized by high melting points, chemical bond strength, thermal conductivity, dielectric properties and chemical resistance. Silicon carbide (SiC) occupies an important place among them. Consequently, SiC is a promising material finding application as structural ceramics to manufacture parts of gas turbine engines, cutting tools and ceramic bearings. High strength of the chemical bonds in this compound allows it to be also used as lightweight armor, because most of the kinetic energy is consumed on breaking the chemical bonds of the armor material under dynamic impact.

Due to the covalent structure of the chemical bond in silicon carbide, ceramics based on this compound with high relative density can be obtained only by means of the solid-phase sintering mechanism at elevated temperatures (from 2050 to 2200 °C) \cite{1}. To obtain dense ceramic from SiC at lower temperatures, it is necessary to apply liquid phase sintering with the use of modifying additives such as oxides in sufficiently large quantities, which leads to porosity formation and a decrease in strength \cite{2, 3}. Another method appears to be an alternative to this ceramics manufacturing technique. This method considers reaction-sintered ceramics (Si-SiC) fabrication for a short period of time and at a lower temperature \cite{4} via penetration of the liquid phase (liquid silicon) into the porous crude SiC preform. The most common current methods of dense SiC ceramics production are: pressureless sintering (PS-SiC), reaction sintering (Si-SiC), and hot pressing (HP-SiC). Silicon carbide-based ceramics are mainly obtained by reaction sintering (Si-SiC) and liquid phase sintering (LPS-SiC) methods.
The use of LPS-SiC ceramics as armor materials with the mechanical properties outweighing advantages of Si-SiC materials allows either increasing the armor resistance class of such ceramics with the same armor thickness or reducing the armor weight under maintaining the armor-defeating performance. However, all these methods have certain disadvantages. For instance, S-SiC materials are resistant to oxidation when being exposed to high temperatures, but their crack resistance is rather low (from 3 to 4 MPa-m$^{1/2}$) [5, 6]. Moreover, an abnormal growth of SiC grain is observed in ceramics owing to high-temperature sintering and relatively long exposures at these elevated temperatures. Si-SiC fabricated by reactive sintering cannot be used at high temperatures due to the low melting point of its silicon constituent. Along with this, the hot pressing method (HP-SiC) is energy-consuming, expensive, and inefficient, that’s why it has not found wide application. The complete compaction can be achieved via high-temperature sintering during short exposure periods, which is accomplished by the method of spark plasma sintering (SPS). The use of additives from metal and ceramic nanopowders as modifiers is considered to be one of the ways to the problem solution. SiC nanopowder modifier is known to be the most promising additive that allows the efficient control over the ceramics structure based on silicon carbide.

The study objective is to propose approaches to increasing strength, hardness, and crack resistance of silicon carbide ceramics by means of SiC nanopowder introduction into its composition and the use of the SPS method to consolidate experimental samples.

2. Materials and experimental procedure

To conduct the experiments, we used 64C green silicon carbide produced by Volzhsky Abrasive Works. Its specific surface was 7.5 m$^2$·g$^{-1}$ after grinding in a jet mill followed by sieving on a classifier. The average particle size of the base powder was equal to 1 μm after grinding. Silicon carbide nanopowder was obtained by mechanochemical activation in the AGO-2C ball planetary mill. Deagglomeration of the nanopowder was carried out by means of ultrasound with 20 kHz oscillation frequency and the processing time of 120 min. Its specific surface was 30 m$^2$·g$^{-1}$. The average particle size of the silicon carbide nanopowder ranged from 75 to 100 nm after deagglomeration. The presence of predominantly hexagonal modification of silicon carbide in the sample (α-SiC) was determined by X-ray diffraction analysis with the use of an ARL X’TRA diffractometer and CuKα radiation. A Technai G2 FEI transmission electron microscope was used to determine the particle size of nanopowders, which ranged from 12 to 200 nm, but the particles with sizes up to 90 nm make up the largest volume fraction.

A powder mixture from base powder particles and a silicon carbide nanopowder was ground in a ball planetary mill. The mass ratio of the dry processed material (the nanostructured powder mixture) to the mass of the planetary mill balls was 1:1, the number of revolutions - 900 rev and the grinding time - 1 min. After grinding, the average particle size of the powder mixture was 0.9 μm. A suspension of a nanostructured mixture of silicon carbide powder was prepared with ethanol used for mixing substances with 1:5 volume-to-volume ratio of “ethanol-mixture”. When the silicon carbide powder was mixed with the silicon carbide nanopowder, the mass ratio of the dry processed material to the balls mass was 1:1. To provide the uniform distribution of nanopowder particles, the number of the planetary mill revolutions was 900 rpm, and the duration of the suspension preparation was 5 minutes. The suspension was dried in an oven at 100 °C for 10 hours, then, the mixture was rubbed through a sieve. The relative humidity of the nanostructured powder mixture amounted to 0.2 wt. % after drying. To determine the optimal amount of the introduced nanopowder, its content had to vary from 1 to 10 wt. % at a constant sintering temperature of 2200 °C. To determine the optimal quantitative content of the SiC nanopowder additive in the powder-based matrix of silicon carbide, model samples of ceramic armor elements with different content of the nanopowder additives were made in the form of 2.5 mm-thick disks with a diameter of 30 mm. The beams with standard sizes were also made for testing of bending strength. The method of spark plasma sintering (SPS) was applied to fabricate the model samples. SPS was carried out in the synthesis mode at a temperature of 2200 °C, under prepress pressure of 85 MPa, and 5 min exposure time at a maximum temperature. X-ray phase analysis and an
energy dispersive elemental analysis were jointly applied with scanning electron microscopy (SEM) to control the phase composition of the powders and ceramics and to check the presence of extraneous phases.

3. Results and discussion

The obtained results confirm that the initial powder additives and fabricated ceramics satisfy the requirements for phase composition and chemical purity. The analysis of the porosity nature was carried out by SEM images (see figure 1) and showed the presence of small intergranular pores in the control sample (without nanopowder additives). Rather large pores are observed in the ceramics with the introduction of 1 % nanopowder (figure 1b), on the contrary, the samples with 5 % (figure 1c) and 10 % (figure 1d) nanopowder additives practically do not contain pores. Based on the research results given in table 1, we can come to conclusion that the use of SiC nanopowder additives leads to a noticeable increase in the intergranular boundaries strength of ceramics. A significant decrease in residual porosity is apparent for samples with 5 wt. % and 10 wt. % nanopowder additives.

![Figure 1. Morphology of cleaved SiC ceramics: (a) control sample; (b) 1 % nanopowder; (c) 5 % nanopowder; and (d) 10 % nanopowder samples.](image)

| The content of the SiC nanopowder additive (wt. %) | Relative density (%) | Fracture toughness, $K_{1C}$ (MPa·m$^{1/2}$) | Vickers hardness (GPa) |
|-------------------------------------------------|----------------------|------------------------------------------|----------------------|
| 1                                               | 97.6                 | 5.0                                      | 28.0                 |
| 5                                               | 98.8                 | 5.0                                      | 24.5                 |
| 10                                              | 98.8                 | 6.4                                      | 28.0                 |

4. Conclusion

The use of silicon carbide nanopowder additives can significantly improve the strength characteristics of SiC-based ceramics. These properties are significantly enhanced for the ceramics containing 5 wt. % and 10 wt. % SiC nanopowder additives. Meanwhile, basic ceramic properties remain almost unchanged. The use of silicon carbide nanopowder additives leads to the formation of a less porous and more uniform microstructure of the material that, consequently, allows increasing the crack resistance of ceramics. Further optimization should be carried out with the ceramics containing 5 wt. % to 10 wt. % nanopowders.
References

[1] Riedel R and Chen L-W 2015 *Ceram. Sci. and Techn.* **2** 3–37
[2] Baud S, Thévenot F, Pisch A and Chatillon C 2003 *J. Eur. Ceram. Soc.* **23** 1–8
[3] Samanta A K, Dhargupta K K and Ghatak S 2001 *Ceram. Int.* **27** 123–33
[4] Grinchuk S *et al.* 2018 *J. Eur. Ceram. Soc.* **38** 4815–23
[5] Swain M 1994 *Materials Science and Technology* vol (New York: VCH Publishers Inc.) p 842
[6] Minori M, Said J, Hamid M, Makio N, Keizo U, Rolf W and Roger M 2002 *Improved Ceramics through New Measurement, Processing and Standards* (American Ceramic Society. Westerville, OH) p 282