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Effect of nano addition on the structure and properties of silicon carbide during electroconsolidation.

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The structure and properties of composite materials based on micropowders of silicon carbide and nanopowders of aluminum oxide and zirconium in the process of hot pressing by electroconsolidation are investigated. The results of researches of the influence of structure parameters and phase composition on the microstructure, physical and mechanical properties of silicon carbide-based composites obtained by electrosparking (electroconsolidation) are described. It has been established that the formation of a composite structure due to the introduction of nanopowders of aluminum oxide and zirconium dioxide into the microporous powder of silicon carbide makes it possible to increase some of the physico-mechanical properties of the obtained composite materials.

Keywords: electroconsolidation; silicon carbide micropowder; alumina nanopowder; sintering; structure; density; transcristallic destruction; intercrystalline fracture; eutectic; liquid phase.

В статье исследованы структура и свойства композиционных материалов на основе микропорошков карбида кремния и нанопорошков оксида алюминия и циркония в процессе горячего прессования методом электроконсолидации. Описаны результаты исследований влияния параметров структуры и фазового состава на микроструктуру, физико-механические свойства композитов на основе карбида кремния, полученных методом электроспекания (электроконсолидация). Установлено, что формирование композиционной структуры за счет введения в микропорошок карбида кремния нанопорошков оксида алюминия и диоксида циркония , позволяет повысить некоторые физико- механические свойства полученных композиционных материалов.

Ключевые слова: электроконсолидация; микропорошок карбида кремния; нанопорошок оксида алюминия; спекание; структура; плотность; транскристаллитное разрушение; интеркристаллитное разрушение; эвтектика; жидкая фаза.

В статье исследованы структура и свойства композиционных материалов на основе микропорошков карбида кремния и нанопорошков оксида алюминия и циркония в процессе горячего прессования методом электроконсолидации. Описаны результаты исследований влияния параметров структуры и фазового состава на микроструктуру, физико-механические свойства композитов на основе карбида кремния, полученных методом электроспекания (электроконсолидация). Установлено, что формирование композиционной структуры за счет введения в микропорошок карбида кремния нанопорошков оксида алюминия и диоксида циркония , позволяет повысить некоторые физико- механические свойства полученных композиционных материалов.

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Obtaining a structural material with high hardness and strength based on silicon carbide is an actual task, since dense materials based on silicon carbide can be obtained only at temperatures above 1900°C, which is extremely energyconsuming and laborintensive. The researches carried out by various authors in particular [1] suggest that without the activation of the sintering process, it is practically impossible to lower the sintering temperature. The way when the sintering temperature is lowered, creating a liquid phase due to the addition of magnesium oxide and calcium is perspective [2,3,4]. It is also possible to lower the sintering temperature by using submicronpowders, nanopowders and modern FAST methods [5]. It is known that aluminum oxide and zirconium dioxide at a temperature of 1860°C form an eutectic [4], therefore, by modifying the silicon substrate with carbide alumina and zirconia nanopowders, the sintering temperature can be lowered. In this case, we used the method of electroconsolidation developed by us, i.e. hot pressing with the use of high ampere current [6, 7]. This will allow to obtain a composite ceramic material

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able of long-term operation in an oxidizing environment at a temperature of 2000°C at an oxidizing flow velocity of 350 m/s, at this while maintaining high strength and hardness. A composite ceramic material containing alumina partially stabilized with yttrium oxide, zirconium oxide and silicon carbide, wherein the alumina and zirconia have a dispersion in the range of 30–60 nm. The initial powder mixtures were prepared by mixing powder components containing alumina, zirconium oxide, silicon carbide, granulating them, then hot pressing with direct current transmission (electroconsolidation) was carried out at a temperature of 1600–1860°C and a pressure of 30 MPa.

As the initial powders, submicron SiC powder (0.1–0.3 μm) obtained by SHS method, alumina nanopowder with a grain size of 30–50 nm (Germany), partially yttria-stabilized zirconia was used. The mixing was carried out in a planetary mill, granulated with the addition of polyvinyl alcohol (PVA), and dried at a temperature of 150–200°C. Hot pressing was carried out at 1600–1860°C in a vacuum environment and held at the final temperature for 2–5 minutes. The increased resistance to oxidation of the proposed high-density composite material is achieved by the introduction of oxide components – nanodispersed alumina and nanosized zirconium oxide partially stabilized with yttrium oxide, which during the hot process can form an eutectic at a temperature of 1860°C. At temperatures of 1600–1860°C diffusion processes proceed more actively. This is one of the factors that provides a high-density, durable material with high temperature and oxidation resistance. It is known that ZrO_2 forms an eutectic of 40 wt% ZrO_2 and 60 wt% Al_2O_3 with a melting point of 1860°C. Introduction ZrO_2–3 wt% Y_2O_3 makes it possible to increase the strength and fracture toughness of the composite material due to the transformation hardening and also stimulates the defect formation reaction inside the nanostructured alumina, thereby promoting the formation of a solid solution at the phase boundaries, which in turn dissolves in corundum causes the formation of vacancies for aluminum. Crystals of corundum become isometric. It is known that silicon carbide is a diamond-like wide-gap semiconductor. Among the family of wide-band materials, silicon carbide has a high Debye temperature, which characterizes its resistance to external influences. Extremely valuable quality is also a sufficiently high thermal conductivity of SiC, second only to diamond, but several times greater than the same parameter for cuprum. Silicon carbide refers to compounds with strong covalent bonds, which hinders mass transfer during sintering without the use of activating additives and application of external pressure. Therefore, in our opinion, the use of hot pressing with the use of high-amperage currents will make it possible to significantly activate the process of obtaining a dense material at relatively low temperatures of hot pressing. Electro-consolidation (electrosparking) is a process of hot pressing with direct passing of high-temperature current through a graphite mold [8]. Sintered samples were used to determine the density, Vickers hardness at a load of 100 N, fracture toughness on a indentation when indenter was pressed in. The microstructure was researched by scanning electron microscopy.

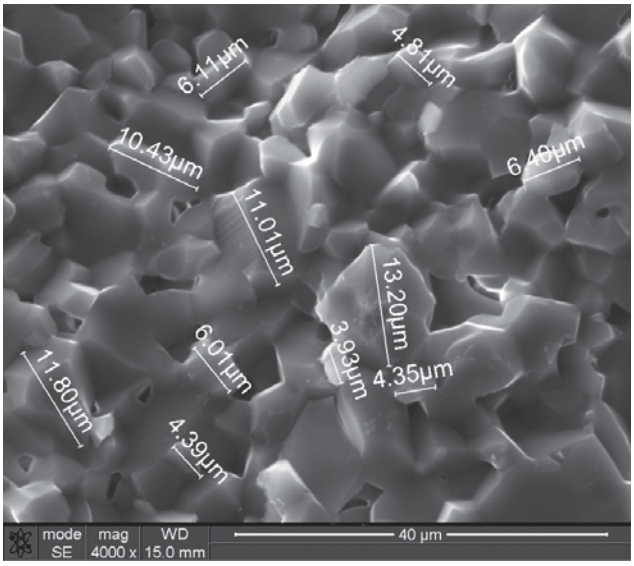
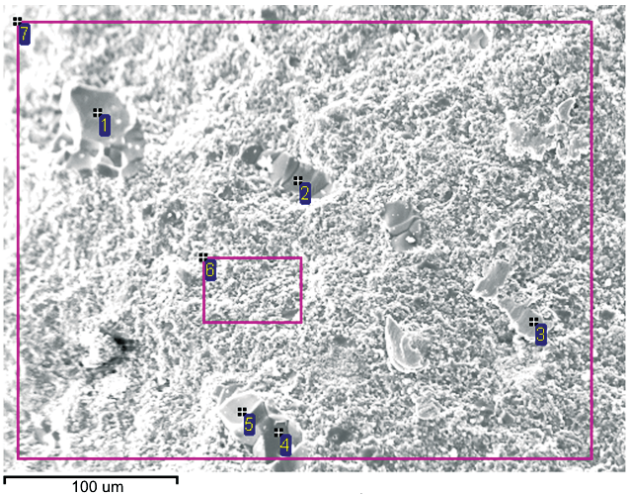


Fig. 1. Structure of SiC-ceramic obtained by electroconsolidation of SiC nanopowder and temperature 1800°C and pressure 30 MPa, sintering time 3 min.



a

Range	C	O	Al	Si	Fe	
1	3.33	51.91	44.23	0.53		100.00
2	2.01	52.97	45.03			100.00
3	3.63		0.31	0.31	95.75	100.00
4	3.72	44.50	51.78			100.00
5	3.46	54.93	41.32	0.29		100.00
6	9.90	41.78	30.53	17.79		100.00
7	10.10	37.96	28.49	18.33	5.12	100.00

b

Fig. 2. a – structure of hot-pressed SiC + Al_2O_3 (50% by weight) at a temperature of 1600°C, a pressure of 30 MPa and a time of 4 min., *b* – distribution of chemical elements in the resulting sample

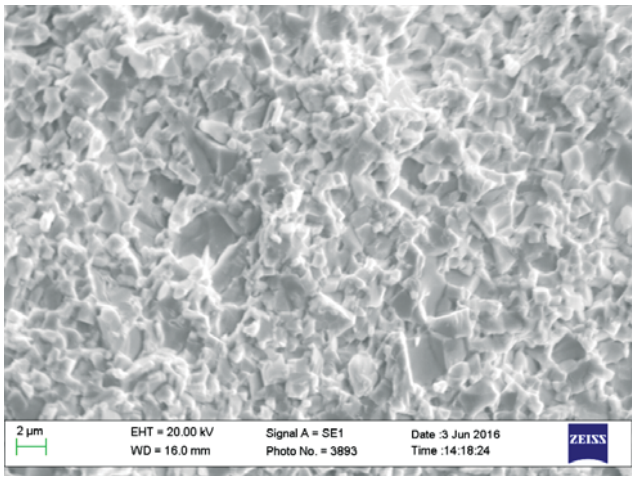


Fig. 3. Fractogram of fracture of composite material SiC–8 wt% ZrO_2–12 wt% Al2O3, obtained at temperature T = 1860°C, pressure P = 30MPa, sintering time 3 min.

From the fractogram of the fracture in Fig. 3, it can be seen that the destruction of the sample is predominantly trans-crystalline, which indicates quite strong bonds at the phase boundaries. This is apparently facilitated by the formation of a liquid phase during sintering.

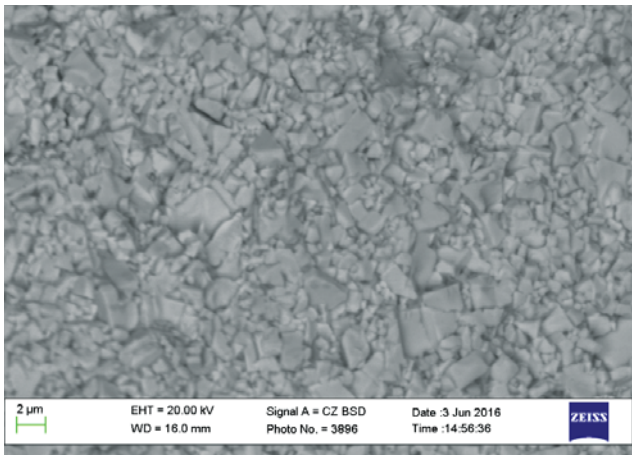


Fig. 4. Fractogram of fracture of composite material SiC – 20 wt% Al_2O_3, obtained at a temperature T = 1790°C, pressure P = 30 MPa, sintering time 3 min.

The microstructure (Fig. 4) shows that the destruction of the sample is also predominantly trans-crystalline, but a small amount of pores is present. It can be assumed that the processes of accelerated surface diffusion on the cleaned surface of powder particles contribute to a more complete compaction in each of the cases shown.

As can be seen from Fig. 5, the amount of pores grows and intercrystalline fracture is noticeable, which indicates an insufficiently strong coupling at the phase boundaries. Obviously, this hot pressing temperature is not sufficient to obtain a high density. The increase in the amount of nanopowders of alumina apparently does not greatly affect the activity of the sintering process at a given relatively low temperature.

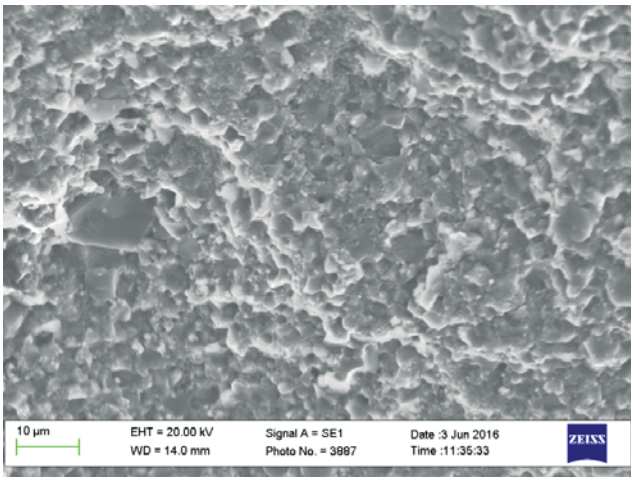


Fig. 5. Fractogram of fracture of composite material SiC – 40 wt% Al_2O_3, obtained at temperature T = 1600°C, pressure P = 30 MPa, sintering time 3 min.

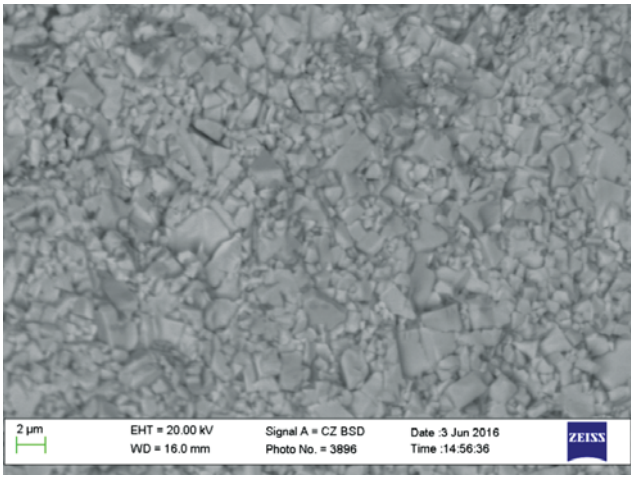


Fig. 6. Fractogram of fracture of composite material SiC – 50 wt% Al_2O_3, obtained at temperature T = 1600°C, pressure P = 30 MPa, sintering time 3 min.

Almost the same is also true for composite materials with a higher content of aluminum oxide nanopowders (Fig. 6). Researches of some physicomechanical characteristics were carried out on samples measuring 5×5×35 mm. Properties of the composite SiC (micropowder)–15 wt% (ZrO2–3wt% Y2O3)–wt% Al2O3:

- flexure toughness – 600-900 MPa;
- fracture toughness – 5-7 MPa·m0,5;
- hardness – 91-93 HRA;
- thermal conductivity – 20-25 Вт/м·К;
- heat resistance – 2000°C.

Thus, the additions of aluminum oxide and zirconia nanopowders partially stabilized with yttrium oxide make it possible to obtain a dense material with high physicomechanical properties at a relatively low temperature, below the eutectic temperature of 1860°C, which is characteristic of Al2O3–ZrO2 materials. The products of the proposed material can be used for the manufacture of heat-stressed parts operating at temperatures

up to 2000°C under conditions that require high strength, hardness and oxidation resistance, as well as in thermal shock conditions, for example thermocouple covers, continuous monitoring of the temperature of metal melts, metalworking industry for the manufacture of cutting tools, in the oil and gas industry (valve devices and pump sealing rings), the tips of mouthpieces for welding, nozzles for sandblasting machines and atomizers of chemical solutions.

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