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New Shaped Ceramics Based on Silicon Carbide

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Abstract—New techniques have been developed for producing inexpensive shaped SiC ceramics with certain structure and porosity for a wide variety of applications. These techniques are based on the interaction of silicon melt with carbon from a previously pressed blank of definite composition (carbon, silicon carbide, organic bond) and porosity.

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Silicon carbide (SiC) based ceramics features high mechanical stability at high temperatures, wear resistance, low thermal expansion coefficient, high oxidation resistance at temperatures below 1500°C, high oxidation resistance, biocompatibility, corrosion resistance, radiation stability, high hardness, and thermal conductivity [1–3]. Because of unique physicochemical properties, SiC ceramics is widely useful in mechanical engineering, as well as nuclear power, defense, metallurgical, food, chemical, and petroleum industries. Possible applications include friction pairs, dry gas-dynamic seals, plain journal bearings rated for use in severe conditions of abrasive and chemically active environments at high temperatures, heating elements, die blocks, spray nozzles, thermocouple cases, and structural elements of rotary engines and turbocharged engines.

There are various ways of obtaining SiC materials: interaction of a silicon melt with porous graphite to produce the so-called siliconized graphite [4, 5], agglomeration of SiC powder at high (>2100°C) temperatures in the presence or absence of activators [6, 7], hot pressing (application of pressure in sintering speeds up significantly the material compaction) [7, 8], and production of a reaction-sintered (self-bonded) SiC [9]. Each of these approaches features nevertheless a number of processing constraints (complexity, high power consumption, impossibility of obtaining complex shapes, etc), due to which the characteristics of resulting SiC ceramic materials often fail to satisfy the up-to-date requirements.

For the most part these materials are heterogeneous compositions in which individuals SiC grains are cemented by adhesives of different composition and physicochemical properties [9]. These adhesives are necessary to facilitate the production of materials and articles or reach intended physical properties or operational performance. In the latter case the properties of the material depend on the amount of the phase components, their sizes, type of distribution, and nature of interphase interactions. In turn, the above factors depend on the process particularities in production of particular materials.

We have developed an economical method for producing multifunctional shaped SiC ceramics. This method relies on the interaction of silicon melt with carbon (siliconization), which is contained in a previously shaped blank of certain geometry, composition (carbon, silicone carbide and organic adhesive), and porosity (Fig. 1). The deviation of siliconized article sizes from preset values does not exceed 1%; i.e., the mechanical treatment necessary for refining articles is minimized, additionally reducing their cost.

This method has an important advantage: the content and porosity of primary blank can be varied in a wide range, depending on the available raw materials, ratio of the components, and the ways for their mixing and compaction. To prepare the input components, we have developed a dry-mixing technique. This approach is more flexible and environmentally friendly, because it excludes toxic phenolic adhesives.

Varying the gas environment composition and the temperature and duration of thermal treatment of siliconized articles, one can control the content of residual silicon and carbon in the articles, which in turn helps to obtain articles with widely controllable ther-

Fig. 1. Sequence of structural SiC ceramics production: (left) SiC–carbon blanks, (middle) siliconized blanks, and (right) final articles after mechanical treatment.

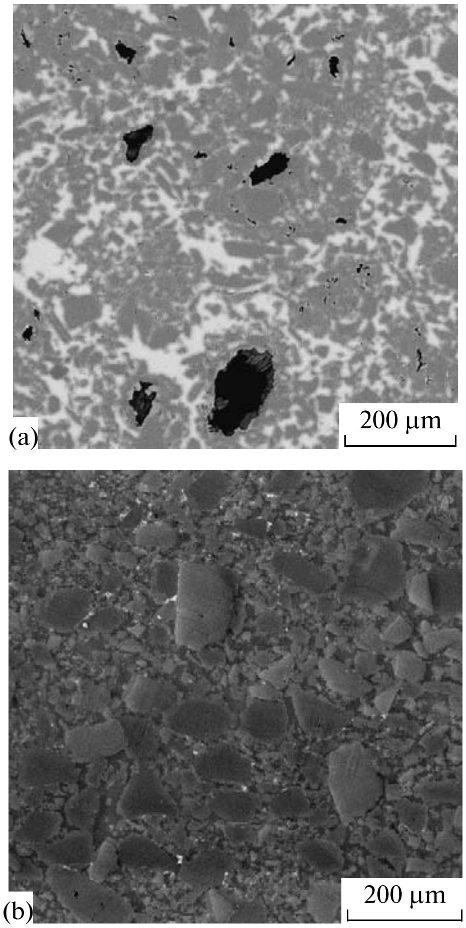

Fig. 2. Structure of SiC ceramics (a) with high content of residual silicon and presence of carbon (black inclusions) and (b) with low content of residual silicon (bright areas).

mal conductivity, electrical conductivity, chemical resistance in aggressive environments, morphology, and other important engineering characteristics. Thus, for specific applications, the density of the new ceramics described here may vary from 1.8 to 3.15 g cm^{-3}. The ceramics with the residual carbon and silicone removed by deliberate thermal treatment features better chemical resistance at high temperatures. On the other hand, articles containing residual carbon exhibit the best antifriction properties.

Examination of the morphology and elementary composition of specimen was carried out using a modern Carl Zeiss Supra 50 VP field-emission scanning microscope with a high resolution (1.3 nm at an accelerating voltage of 20 kV and working distance of 2 mm). The microscope is equipped with several secondary electron detectors: a standard Everhart–Thornley secondary electron detector, a low-vacuum secondary electron detector to examine nonconducting specimen, and an In-lens detector to obtain high-resolution images.

Figure 2 shows the microstructure of SiC ceramics with high (>20%) and low (<2%) residual silicon contents. The SiC ceramics with a high free silicon content (Fig. 2a) was prepared from the initial specimen obtained by wet mixing of carbon and silicon carbide particles with sizes from few microns to 100 μm. To obtain the ceramics with a low silicon content (Fig. 2b) with a small deviation from stoichiometric SiC, we used dry mixing of nanosized powders with subsequent compressing blanks to a density of 1.95 – 2.05 g cm^{-3}. The blank siliconization yielded SiC-ceramics specimen with a density up to 3.15 g cm^{-3}, having a much smaller grain size with a higher structural uniformity.

A significant advantage of the SiC ceramics developed is its high resistance to thermal shock; in this parameter it is considerably superior to the commercial SiC ceramics. This was verified by comparative tests of the articles made of three types of SiC ceramics: the hot-pressed Hexoloy ceramics (Saint Gobain (France, USA) Production) [8], silit produced in Russian, and the ceramics in question. The specimen were heated in air in a resistor furnace to 1200°C and then immediately quenched into cold water. For Hexoloy and silit exhibited crack formation in water in the first cycle, whereas the new-ceramics articles showed no signs of failure after ten heating–cooling cycles. The combination of thermal shock resistance and high-temperature chemical resistance substantively widens the possibilities for practical applications of new ceramics. For example, the replacement of conventional ceramic heaters in high-temperature (1500°C or more) air furnaces will make unnecessary energy- and time-consuming stages of smooth heating and cooling these furnaces. Indeed, for conventional ceramic heaters the heating and cooling rates should not exceed 150 K/h to prevent them from cracked. At the same time, the electric heaters made of the novel SiC ceramics can withstand repeated heating and cooling cycles from room temperature to 150°C for times of 3–5 s, with subseguent instantaneous switching off the feed current.

The possibility of controlling crystallite sizes and pores in articles made of the novel SiC ceramics in a wide range allows one to substantially increase their radiation resistance. One of the reasons for this increase is that the time of radiation defect emergence to the grain surface for small grains (which decreases proportionally to the square of the grain linear size) turns out to be smaller that the time interval between two successive events of trapping of ionized particle by a given grain. As a result, various nanostructured ceramics can stably operate in reactor cores with radiation flows of 10^{13} or more particles per square centimeter per second.

The SiC materials obtained far surpass in basic physical and mechanical characteristics the available siliconized graphite. Their bending resistance (~400 MPa) and Young modulus (up to 430 HPa) are the same or better than the corresponding characteristics of hot-pressed silicon carbide of Hexoloy type, the latter being produced by a much more complicated and expensive technology (the price of hot-pressed silicone carbide articles is approximately an order of magnitude higher than that of similar articles made of our ceramics).

CONCLUSIONS

We have developed methods for producing shaped articles from multifunctional and inexpensive SiC ceramics. Our approach is based on interaction of silicone melt with carbon from a previously pressed blank of definite composition (carbon, silicon carbide, organic adhesive) and porosity.

The indisputable merit of the developed ways for producing SiC ceramics is the simplicity and low cost of equipment, availability of raw materials, and capability of controllable variation of the ceramics composition and structure in a wide range, depending on the particular application. As a result, one can obtain performance specifications significantly exceeding those of commercial SiC ceramics.

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