Journal of Engineering Physics and Thermophysics, Vol. 66, No. 6, 1994

THERMAL EXPANSION OF SILICON CARBIDE MATERIALS

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UDC 539.21

The influence of porosity and sintering additives on the thermal expansion of silicon carbide ceramics with additions of Al_2O_3, $B(B_4C)$, and BeO is investigated in the temperature range 50–700°C.

A ceramics based on silicon carbide is distinguished by high heat resistance owing to low thermal expansion and high heat conduction. In creating ceramic items to be used at high temperatures and loads, it is necessary to have reliable data on the dependence of the thermal expansion of the ceramics on the temperature, porosity, and kind of sintering additives. Moreover, determination of regularities in the change in the properties as a function of the porosity and the sintering additives may facilitate production of SiC-based materials with specified properties.

A number of works [1–5] are devoted to a study of the thermal expansion coefficient (TEC) of silicon carbide materials. However, the reported data correspond mainly to a narrow temperature range or to some temperature close to room temperature. Moreover, these works contain no information on specific features of the behavior of the TEC of porous materials.

The present work reports results of an investigation of the temperature and porosity dependence of the thermal expansion coefficient for silicon carbide-based materials with Al_2O_3, $B(B_4C)$, BeO as sintering additives and for relatively pure silicon carbide powder, grade ZAK, without specially added impurities. The content of the corresponding sintering additives in the ceramics, namely, 1.5% BeO, 0.5%B + 1%C, ~2% Al_2O_3 was the optimum [1–3, 6].

The temperature dependence of thermal expansion of the samples was measured by a quartz dilatometer with a photoelectric recording system.

The samples were characterized by different porosity, which, as our investigations revealed, exerts a pronounced influence on the thermal expansion of these ceramic materials. According to [7], the effect of porosity on the TEC is negligible in materials that represent a solid skeleton with isolated inclusions in the form of pores. The materials of which our samples were made represented complex heterogeneous structures with high intercommunicating porosity (up to 40%). The skeleton of these ceramics consisted of SiC single crystals oriented in a different manner, the cohesion forces between which limited the overall strength of the ceramics. As a consequence of solid-phase reactions of the SiC single crystals with the sintering additives, the intergranular space was occupied by the binder, which, in turn, disrupted the continuity of the main phase. The dependence of the thermal expansion on the temperature and porosity of these ceramics is shown in Fig. 1. This behavior of the TEC is probably associated with the same effect as the decrease in the heat capacity per unit volume, whose value falls with an increase in porosity due to the decrease in the mass of the substance in the volume [7]. The thermal expansion, just like the heat capacity, is proportional to the lattice energy, and therefore it may be expected that the expansion of ceramics with the described structure is determined by the total number of interatomic bonds. In [4], it is reported that the mean TEC tends to fall in silicon carbide ceramics of the same composition and different porosity.

Processing of the results of the investigation of the porosity dependences of the TEC in our samples showned that they are well approximated by the exponent

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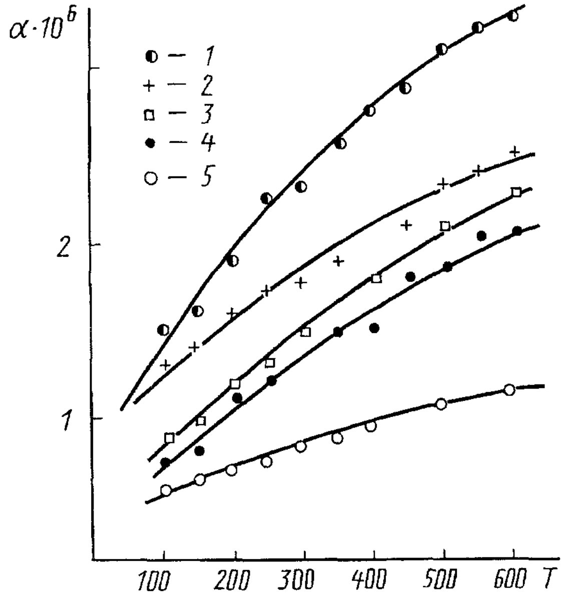
Daghestan State University, Makhach-Kala. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 66, No. 6, pp. 739-741, June, 1994. Original article submitted May 22, 1992.

1062-0125/94/6606-0660$12.50



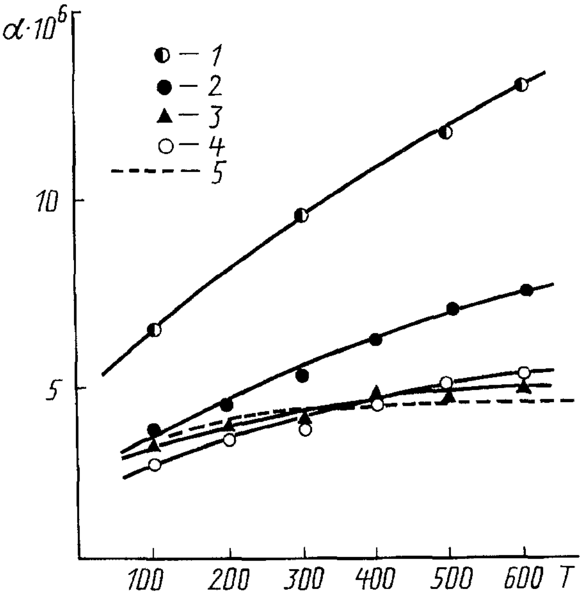


Fig. 1. Temperature dependences of the TECs of the samples $SiC-B(B_4C)$ ceramic of various porosity: 1) 20%; 2) 25: 3) 29; 4) 36; 5) 41.2%. α, K^{-1}, T, °C.

Fig. 2. Temperature dependences of the TECs of nonporous silicon carbide materials: 1) $SiC-Al_2O_3$; 2) $SiC-B(B_4C)$; 3) SiC ceramics; 4) SiC–BeO; 5) SiC single crystal [8].

$$\alpha = \alpha_0 \exp(-bn) \qquad (1)$$

where α, α_0 are the TEC of the porous material and the skeleton material, respectively; n is the porosity; b is a constant. For all the samples, $b \approx 4.2 \pm 0.2$ and it hardly depends on the temperature. Approximation (1) is indicative of the fact that the thermal expansion of the investigated ceramics depends on both the nature of the interatomic interaction forces and the number of bonds per unit volume. Extrapolation to zero porosity enabled consideration of the influence of the type of sintering additives on the absolute TEC values and their temperature dependences (Fig. 2).

The coefficients of thermal expansion of a single-crystal sample [8] and our (SiC-based polycrystalline) samples are close. At the same time, as seen from Fig. 2, polycrystallinity removes the TEC "anomaly" typical of SiC single crystals of α- and β-modification at the temperatures from 300 to 1000 K. The thermal expansion of the polycrystalline SiC increases in the temperature range 300–1000 K, tending to saturation upon approaching the region of Debye temperatures. The addition of BeO decreases (in absolute value) the thermal expansion coefficient up to ~ 600 K but increases its temperature coefficient. Therefore, at $T > 600$ K, the TEC of this ceramics is already higher than that of polycrystalline silicon carbide. This is probably caused by the fact that at high temperatures the TEC of pure beryllium is substantially higher than that of silicon carbide. Our data on the mean TEC of the SiC-BeO ceramics in the temperature range 80–200°C comprise $\sim 2.8 \cdot 10^{-6}$ K^{-1} and are in good agreement with the value $2.6 \cdot 10^{-6}$ K^{-1} reported in [1] for a SiC ceramics with 2% BeO.

The addition of Al_2O_3, on the other hand, substantially increases the thermal expansion of the silicon carbide ceramics. The same effect is described in [5] and is explained by the fact that the alumina, occupying the space between SiC single crystals and possessing high α values, favors an increase in the thermal expansion of the material. According to the data of [2] the alumina at the grain boundaries of SiC forms an amorphous phase, i.e., the binding forces in this part of the ceramics skeleton are weakened, thus causing an additional increase in α. Boron impurities also increase, but much more weakly, the thermal expansion of the silicon carbide ceramics in the high-temperature range. Thus, from our studies it may be inferred that porosity and insignificant amounts of impurities not only exert a pronounced influence on the nature of the temperature dependence of the thermal expansion of the silicon carbide ceramics but also change its value within wide limits. Consequently, by changing the porosity and choosing the sintering additive properly, one may obtain materials with thermal expansion coefficients that meet the requirements of concrete applications.

NOTATION

α, thermal expansion coefficient; b, constant in the exponent; n, porosity; T, temperature.

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