UDC 666.3-16 + 666.762.852-494

COMPOSITE CERAMIC MATERIALS WITH SiC FIBER

A. V. Galakhov,[1] G. A. Pankov,[1] and G. A. Fomina[1]

Translated from Ogneupory, No. 8, pp. 3 – 6, August, 1995.

Original article submitted February 13, 1995.

The technology and mechanical characteristics of composite ceramic materials strengthened by discrete and continuous fibers of silicon carbide are described. The introduction of an 8% volume fraction of continuous SiC fibers into cordierite increases its strength by almost a factor of 2. Among composites strengthened by SiC whisker the highest density is exhibited by materials with a matrix of $Al_2O_3 - ZrO_2$ (580 MPa). Materials with matrices of Al_2O_3 and synthetic mullite have mean ultimate bending strengths of 460 and 420 MPa, respectively. The microstructures of the materials are described.

At the present time intense research is directed toward the creation of new composite materials and technological processes for producing them. This is associated with the solution of new problems in both traditional and new fields of science and engineering [1 – 3]. Interest in composite materials with a ceramic matrix stems from the need to reduce or eliminate the usual deficiency of ceramic materials caused by their low crack resistance. Reinforcement of ceramics by high-strength fibers is a very promising way to improve their crack resistance. Many works on ceramic composites are devoted to the development of materials that have oxide matrices and are reinforced with silicon carbide fiber. Interest in these materials is due to progress in the development of high-strength silicon carbide fiber, on one hand, and the high functional properties of these materials, on the other.

In the present work we present results of technological investigations of the following ceramic composites: (1) cordierite – continuous SiC fiber of the Nikalon type, (2) composites with matrices of mullite, Al_2O_3, $Al_2O_3 - ZrO_2$ strengthened by discrete SiC whisker.

INITIAL MATERIALS AND METHODS OF INVESTIGATION

We tested two types of fiber produced by the Redkinskoe Research and Production Association "Khimvolokno", namely, SiC fiber of the Nikalon type and silicon carbide whisker. The continuous Nikalon fibers are supplied in the form of strands. The diameter of the fibers is 12 – 15 μm. X-ray phase analysis of the material showed that the fibers do not contain SiC as a crystalline phase. The elasticity modulus and the ultimate tensile strength of the fiber, by data of the supplier, are 140 and 1.5 – 2.5 GPa, respectively. The discrete SiC whisker, by data of the supplier, has a diameter of 5 – 7 μm and a length of 10 – 20 μm. The whisker is highly contaminated by unreacted carbon and residual silica.

The matrix powders of cordierite ($2Al_2O_3 \cdot 2MgO \cdot 5SiO_2$) and mullite ($3Al_2O_3 \cdot 2SiO_2$) were high-purity submicrometer powders specially synthesized by the sol-gel method. The process of their preparation is described in detail in [4, 5]. The matrix powders based on Al_2O_3 were obtained from commercial alumina of grade G-00. The alumina was fired at 1400°C into the α-form with subsequent mechanical crushing to a specific surface of 8 m^2/g. The mass fraction of ZrO_2 in the $Al_2O_3 - ZrO_2$ system was 16%. The powders of ZrO_2 contained a 3% (mole fraction) stabilizing addition of Y_2O_3. The powders in the system $Al_2O_3 - ZrO_2$ were prepared by chemical coprecipitation from mineral salts. Their specific surface was 32 m^2/g.

In order to obtain composites with continuous fibers we used a technology that included preparation of a mat with the requisite configuration from the fibers, impregnation of the mat by a suspension of matrix powders, drying, pressing in a metallic mold, distillation of the binder, and sintering or hot pressing. Before using SiC whisker it was subjected to chemical cleaning from the impurities; the globular particles were separated by flotation. Composites with SiC whisker were prepared by a technology that included mixing of the matrix powders and whisker in the process of wet crushing in a ball mill, drying, and hot pressing.

[1] Moscow State Aviation Technology University, Moscow, Russia.

0034-3102/95/3608-0235$12.50

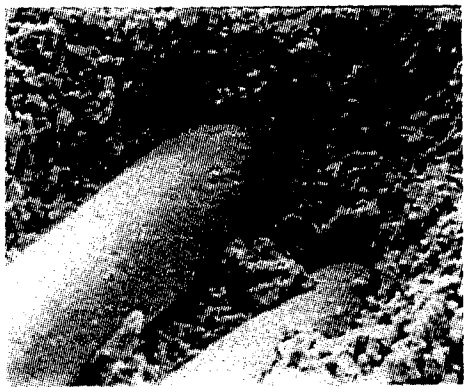

Fig. 1. Electron micrograph of a fracture surface of a hot-pressed composite of cordierite and continuous SiC fiber. × 3000.

The specific surface of the synthezied powders was determined by the BET method, and the temperatures of phase transformations were determined by differential thermal analysis. The morphology and sizes of the particles were studied using scanning electron microscopy. This method was used to study fractures in the composite materials and investigate their structure in microsections. X-ray phase analysis was performed on a DRON-3M diffractometer by the standard method. The apparent density and the open porosity of the specimens were determined by hydrostatic weighing in distilled water. The ultimate bending strength was determined on specimens 6 × 4 × 32 mm in size by the three-point loading technique. The specimens for mechanical tests were cut with a diamond disk and ground over the perimeter without polishing.

RESULTS OF THE INVESTIGATIONS

Composite with continuous SiC fibers. The volume fraction of fibers in the composite material with a cordierite matrix was 8%. The first batch of specimens was obtained by conventional sintering without applying pressure. The sintering was conducted in air. In order to determine the sintering temperature appropriate for preparing a composite with a density of at least 96% of the theoretical value, they were subjected to firing at different temperatures in the 1350 – 1420°C temperature range. It was established that the density of specimens sintered above 1380°C is higher than the 96% limit.

Results of the mechanical tests showed that the ultimate bending strength of specimens of the cordierite – SiC fiber composite obtained at a sintering temperature of 1390°C is 100 – 128 MPa and that of specimens sintered at 1400°C is only 60 – 96 MPa. Reference specimens of matrix powders without fiber were tested together with the specimens of the composite in the same press. The matrix material exhibited an ultimate bending strength of 96 – 104 MPa at all sintering temperatures. Reinforcement of the material with the fiber increased the ultimate bending strength by 20%.

Investigation of fractured surfaces showed that sintering of the composite is accompanied by chemical interaction on the matrix-fiber interface. Traces of this interaction are clearly seen on protruding fiber parts. An interaction zone in the form of a concentric void is formed in the interface region. At 1400°C the fiber dissolves completely in the matrix material. In order to determine the temperature and the nature of the interaction we obtained separate thermograms of the fiber and the mixture of the cordierite powder and the fiber. The beginning of the interaction between the fiber and the matrix material was detected at 890 – 900°C and exhibited an exothermic effect on the thermogram. X-ray phase analysis of specimens passed through a thermoanalyzer did not show the presence of any new phases in either the fiber or the mixture. Up to 1400°C the fiber remained amorphous and the mixture exhibited reflections corresponding to α-cordierite (the matrix material). These data mean that the Nikalon fiber produced by the Redkinskoe Plant does not contain crystalline SiC and seems to consist of free carbon and vitreous SiO_2. At sintering temperatures above 800°C silicon fiber is dissolved in cordierite with a change in the stoichiometric composition of the latter. We did not detect thermal effects on thermograms of the matrix material and the fiber taken separately. Preservation of the fiber in the cordierite matrix and realization of the effect of fiber strengthening in the given system are possible either at a sintering temperature below 900°C or in brief sintering under pressure (hot pressing).

Investigation of the process of hot sintering in the given system showed that the material is completely compacted even at 1200°C. Electron micrographs (Fig. 1) of fracture surfaces exhibit no traces of chemical interaction. The ultimate bending strength of the composite material increased almost twofold compared to the matrix material and was equal to 180 MPa.

Composites with SiC whisker. The volume fraction of whisker in composites with SiC whisker was 12% for any type of matrix material. The composites were obtained by hot pressing. This method is traditionally used for preparing such materials because sintering without pressure does not provide the requisite density for whisker-reinforced materials. All materials were pressed in graphite molds at 1500°C and 20 MPa. The holding time was 1 h.

Figure 2*a* presents an electron micrograph of a fracture surface of a material of the Al_2O_3 – SiC whisker system. The density of the material was 92% of the theoretical value. The mean ultimate bending strength was 460 MPa. The material with a matrix of $Al_2O_3 - ZrO_2$ had a higher density and higher strength characteristics. Its mean ultimate bending strength was 580 MPa at a density of 98% of the theoretical value. The denser structure of this material is shown in Fig. 2*b*. It is obvious that in this case an important role is played by both the addition of active ultradispersed ZrO_2

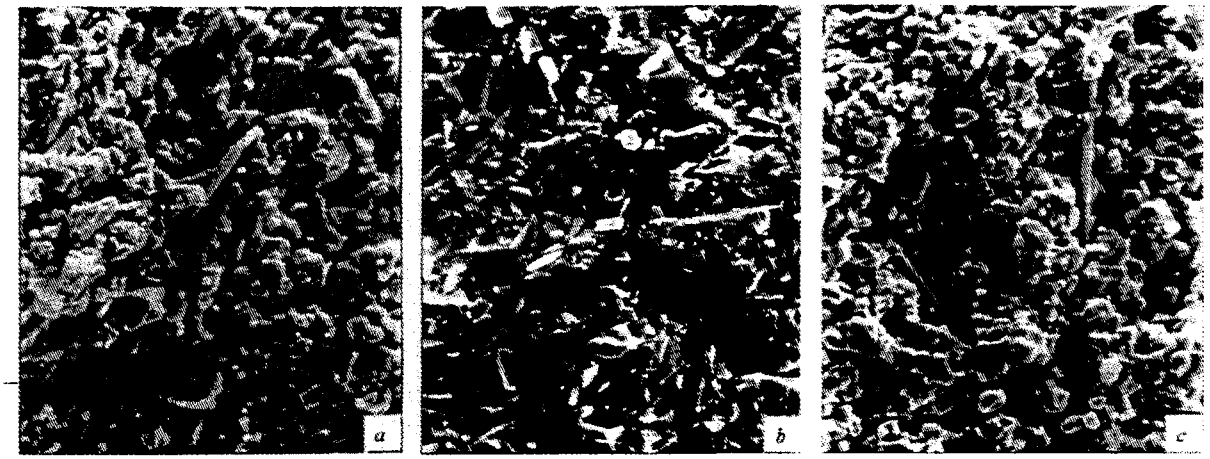

Fig. 2. Electron micrographs of fracture surfaces of hot-pressed composites strengthened by SiC whisker: *a*) Al_2O_3 – SiC whisker; *b*) Al_2O_3 – ZrO_2 – SiC whisker; *c*) mullite – SiC whisker. × 5000.

powder to the matrix Al_2O_3 and the additional contribution of the mechanism of transformation strengthening to the strength that occurs in ceramics containing inclusions of metastable tetragonal particles of ZrO_2. The material with a matrix of synthetic mullite has a mean ultimate bending strength of 420 MPa despite its relatively high density (97% of the theoretical density). A fracture surface of this material is shown in Fig. 2*c*.

In conclusion we should note that the temperature of hot pressing used in our experiments was 150 – 250°C lower than that commonly used for similar materials[2] [6, 7]. However, the strength obtained was at the level of data presented in the domestic and foreign literature for these materials. This indicates that the prospects of the presented technological approaches for further improvement of the mechanical characteristics are quite good.

[2] U. S. Patent 4657877.

REFERENCES

1. A. G. Evans, "Perspective on the development of high-toughness ceramics," *J. Amer. Ceram. Soc.*, 72(2), 187 – 206 (1990).
2. A. V. Galakhov and V. Ya. Shevchenko, "Composite ceramic materials," *Ogneupory*, No. 6, 53 – 58 (1990).
3. D. Desnoyer, "Les materiaux composites a matrice ceramique: des presurseurs de percee technologiques," *Mater. et Tech.*, 79(5 – 6), 57 – 64 (1991).
4. A. V. Galakhov and V. Ya. Shevchenko, "Effect of the conditions of gel formation on phase transformations in polymer aluminosilicate gels of mullite composition," *Ogneupory*, No. 1, 8 – 11 (1991).
5. A. V. Galakhov, V. Ya. Shevchenko, and A. A. Stebunov, "Phase transformations in Al – Si – Mg gels. Effect of Ti, Ce, and Zr additives," *Ogneupory*, No. 6, 11 – 13 (1991).
6. J. Homeny, W. L. Vaughn, and A. K. Farber, "Processing and mechanical properties of SiC-whisker – Al_2O_3-matrix composites," *Amer. Ceram. Soc. Bull.*, 66(2), 333 – 338 (1987).
7. E. Luccini and S. Maschio, "Processing and mechanical properties of SiC-whisker – reinforced Al_2O_3 – ZrO_2 composites," *J. Mater. Sci. Lett.*, 9(4), 417 – 419 (1990).