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ON REACTION-SINTERING PRODUCTION OF SILICON CARBIDE MATERIALS

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The effect of the production process of the initial charge and conditions of reaction sintering on the structure and mechanical properties of silicon carbide materials is studied. The kinetics of reaction sintering of silicon carbide materials and the mechanical properties of the sintered material are investigated.

The creation and use of effective materials that remain reliable under extreme operating conditions are vitally important for the acceleration of progress in science and technology.

Materials based on silicon carbide are most promising, and their main advantages are substantial high-temperature and breaking strengths, low density, and high erosion and chemical stabilities.

It is known that the serviceability of products made of silicon carbide materials depends on their density and phase composition. With this in view, we have investigated the interrelation between the structure, phase composition, and properties of reaction-sintered materials.

Green silicon carbide powders M40, M28, and M10 manufactured by Zaporozhe Abrasive Works were used in the study.

Since in this country industry does not produce silicon carbide powders with a wide range of grain sizes and a high density of compaction can only be achieved by combining certain amounts of powders of the various fractions, initial silicon carbide powder M40 was ground by a vibratory mill for 15, 30, 45, and 60 min. In order to obtain finer fractions, silicon carbide powders M28 and M10 were ground for 30 and 60 min and 30, 60, 90, and 120 min, respectively.

Carbon black in amounts of 15, 20, 25, and 30% was used as an additive. More carbon and better compactability were provided by the introduction of either a phenolformaldehyde binder or a plasticizer in the form of a 10% polyvinyl alcohol (PVA) or polyvinylpyrrolidone (PVP) solution. The silicon carbide charge was produced by mixing powders of the various fractions. The charge was compacted to obtain specimens of $d = 10 \times 10$ mm and $5 \times 5 \times 35$ mm at pressures of 0.8, 1.2, 1.6, and 2 ton for investigation of the structure and bending strength.

Reaction sintering was carried out under vacuum; the maximum sintering temperature was 1700°C.

The structure of the material was studied on unetched microsections with an MEF-3 metallographic microscope (Austria); the phase composition was determined with x-ray analysis on a Dron-3 device under Cu-K_α radiation.

The strength properties (bending strength) were calculated from a four-point static bending failure diagram.

A study of the grinding kinetics for the powders showed that the most intense grinding is observed at the initial stages (Fig. 1). Moreover, this is accompanied by changes in the shape of the particles. Electron microscopy studies showed that they becomes more rounded (Fig. 2). The particle size of the finely disperse powder decreases insignificantly in the grinding process: in 120 min the particle size of powder M10 decreases from 11.8 μm to 7μm.

Initial powder M28 that was premixed with 20% of carbon black was ground in a vibratory mill for 10, 30, and 60 min. After 10-min grinding the average particle size was 17 μm, after 30 min, 15 μm, and after 60-min grinding the average diameter remained $d_{av} = 15$ μm.

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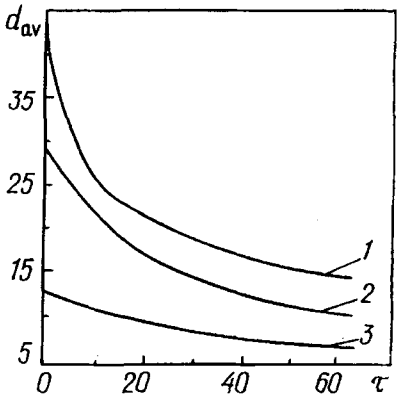

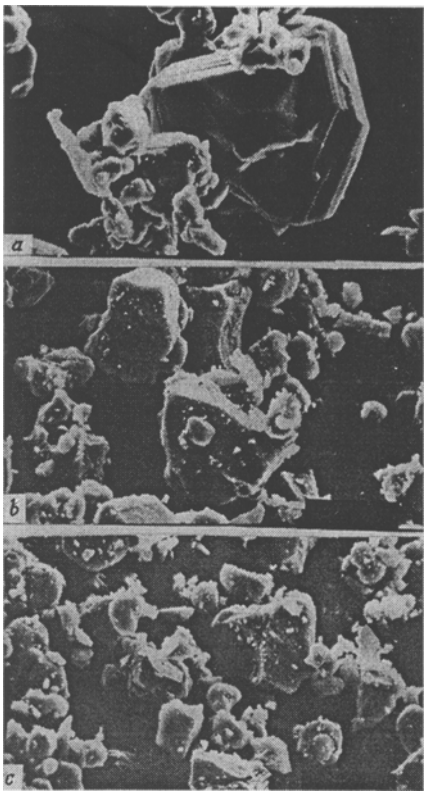

Fig. 1. Grinding kinetics of silicon carbide powder: 1) M40; 2) M28; 3) M10. d_{av}, μm; τ, min.

Fig. 2. Form of silicon carbide powder: a) initial powder M28; b) ground for 10 min; c) ground for 20 min.

A study of the compaction process revealed that without preliminary grinding in a vibratory mill the powders have no moldability. Irrespective of compaction pressure, a loose briquette is formed and its shape is not preserved. Use of PVP as a plasticizer increases the moldability of the charge, however, it is impossible to obtain a quality briquette from the initial powders. Additional treatment with a vibratory mill and introduction of a 1% PVP solution result in a briquette with the required density of 1.6–1.8 g/cm^3. It should be noted that an increase in compaction pressure has almost no effect on the density of the specimens.

Moreover, this process has a positive effect not only on the moldability and compactability of the charge, but also on the sintering quality of silicon carbide materials, since in the grinding process the carbon-black particles are spead over the silicon carbide particles, which results in more uniform and intense interaction with the silicon during sintering.

The reaction sintering of silicon carbide materials can be divided into three stages.

The first stage is heating to 1100°C to remove the binder introduced into the charge and random impurities. It should be emphasized that the charge should be heated at a low rate; otherwise, the specimens will crack due to intense gas emission in this temperature range.

The second stage is silicon melting at 1300–1700°C. In this range, the heating rate should be about 50°C/min. At a temperature above 1600°C the viscosity of the silicon drops abruptly and the contact angle between the carbon and silicon carbide and liquid silicon decreases, and, consequently, the formation of secondary silicon carbide is accelerated.

The third stage is isothermal holding at 1700–1750°C. The holding time is determined by diffusion processes associated with silicon–carbon interaction, recrystallization of the initial silicon carbide, and formation of secondary silicon carbide. Therefore, the holding time depends on the size of the specimens and should be sufficient for full completion of the diffusion processes.

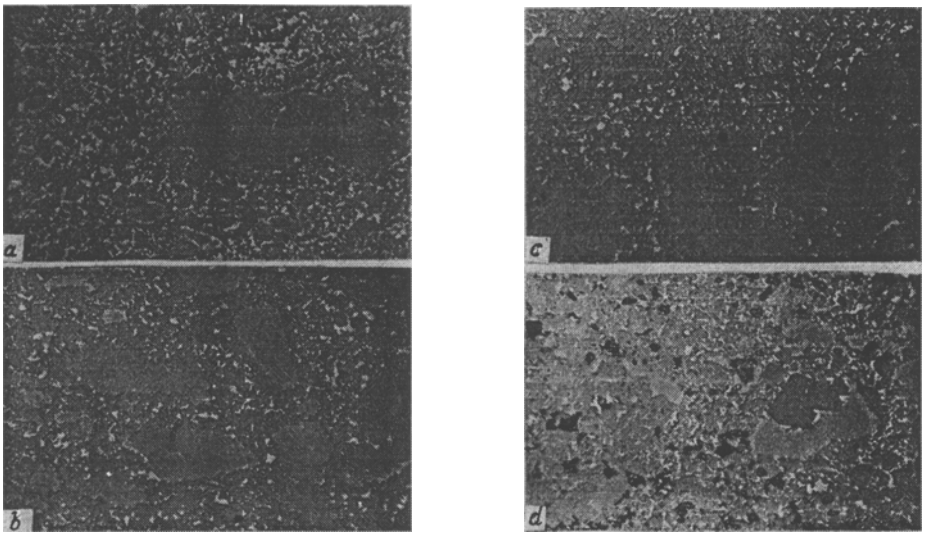

Fig. 3. Microstructure of reaction-sintered silicon carbide material from powder M28 with various contents carbon black in charge: a) 15%; b) 20; c) 25; d) 30%. ×200.

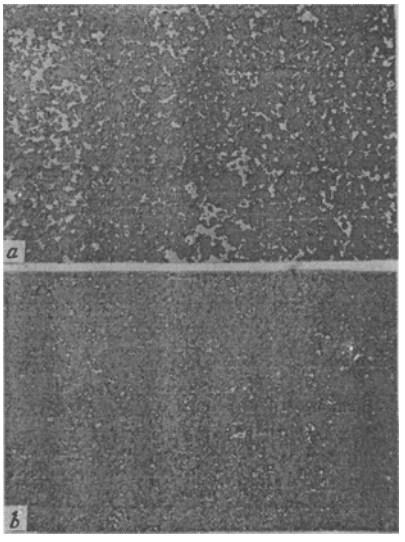

Fig. 4. Microstructure of sintered silicon carbide material from powder M28 with addition of 20% carbon black ground for 10 min (a) and 60 min (b). ×200.

Investigation of the structure of silicon carbide materials with various carbon contents in the charge has shown the presence of large amounts of silicon (up to 40%) in the specimens with 15% carbon black after sintering (Fig. 3a). This results in a decrease in strength and embrittlement of the material.

When the carbon-black content was increased to 25–30%, some of it did not react: less at 25% (Fig. 3c) and more at 30% (Fig. 3d). An optimum amount of carbon black introduced into the charge is 20% (Fig. 3b). In this case, the content of silicon in the sintered material is 10–15%.

In this work the effect of carbide particle size on the structure and density of the sintered material was also investigated.

After reaction sintering at 1700°C following the above scheme, the density of specimens from a charge ground for 10 min was 2.97 g/cm^3 (the density after compaction was 1.86 g/cm^3) and the density of specimens ground for 60 min was 2.92 g/cm^3 (the initial density was 1.96 g/cm^3).

The study of the microstructure of the sintered specimens has shown that the material obtained from a charge ground for 10 min has a coarser-grained structure with larger and nonuniform inclusions of free silicon (Fig.

TABLE 1. Mechanical Properties of Silicon Carbide Materials

Charge composition	Grinding time, min	Density, g/cm^3	Bending strength, MPa
M28+20% of carbon black	10	3.03	330
	30	3.06	390
	60	3.04	290
M40+20% of carbon black	10	3.03	278
	30	3.07	299
	60	3.07	274

4a). The content of free carbon black is very low and does not exceed 3–5%. An increase in the grinding time results in a fine-grained structure with a more uniform distribution of silicon inclusions (Fig. 4b).

Depending on the initial composition, sintered silicon carbide materials have densities of from 2.9 to 3.06 g/cm^3. The content of free silicon in the sintered materials from powder M28 compacted at 15 MPa is 19%, at 48 MPa, 16.5%, and at 64 MPa, 13%.

The phase composition of the material also depends on the amount of the carbon addition. When 15% carbon black is introduced, up to 40% silicon is observed in the sintered material; when the content of added carbon is above 25%, some of the carbon does not react.

The grain size of the sintered material depends on the initial particle size of the silicon carbide powders. Materials from powder M28 that was ground beforehand for 10 min have a grain size of 35–50 μm, and those ground for 60 min have a grain size of 25–30 μm. Materials from powder M40 preground for 30 min have a grain size of 60–100 μm, and those preground for 60 min, 30–60 μm.

The study of the strength of the sintered materials revealed that it depends on the density and phase composition of the material (Table 1).

A tendency to decreasing strength of the specimens from powders ground for 60 min is observed in the materials from both silicon carbide M28 and M40. It can probably be attributed to changes in the phase and chemical composition of the material. Since the powders were ground by steel balls in a steel mill, the charge is contaminated with iron in the grinding process. For example, if the initial silicon carbide contains 0.0005% iron, after 60-min grinding it contains 0.31%. An increase in the iron content of the material impairs its mechanical properties.

Use of ceramic milling bodies will prevent contamination of the charge with iron and improve the mechanical properties of silicon carbide materials.

REFERENCES

1. G. G. Gnesin, Silicon Carbide Materials [in Russian], Moscow (1977).
2. Yu. P. Dyban', Solid-Phase Sintering of Silicon Carbide [in Russian], Kiev (1988).