

PAPER • OPEN ACCESS

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To cite this article: S N Perevislov *et al* 2019 *IOP Conf. Ser.: Mater. Sci. Eng.* **525** 012073

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Liquid-sintered SiC based materials with additive low oxide oxides

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Abstract. By the method of free liquid-phase sintering, dense materials based on silicon carbide ($P \leq 0.5\%$) were obtained. In the preparation of charge powders, the method of co-precipitation of oxides on silicon carbide powder was used. As a sintering additive, a eutectic mixture of the components MgO, Y₂O₃, and Al₂O₃ was used, corresponding to the composition on the garnet-spinel line. The resulting materials have a high level of mechanical properties: $E = 410$ GPa; $\sigma = 680$ MPa; $K_{IC} = 6.0$ MPa·m^{1/2}; $HV = 21.5$ GPa, approaching the level of properties of hot-pressed silicon carbide materials.

1. Introduction

The classical method of liquid phase sintering of SiC with oxide additives includes mechanical mixing micron powders, molding the preforms and high temperature sintering materials, what It has several disadvantages: uneven mixing powders of oxide between the silicon carbide particles, the complexity of a homogeneous distribution of small amounts (≤ 5 wt. %) of oxides, The impossibility of a uniform distribution of the oxide components in strictly eutectic ratio. Alternative mechanical components may be kneaded mortar mix techniques (sol-gel method, precipitation and co-precipitation from salt solutions, colloidal activation method, and others.). Which was investigated by some researchers [1–4].

The purpose of this paper is to obtain on the basis of materials SiC with additives of oxides MgO, Y₂O₃ and Al₂O₃, deposited on the silicon carbide surface by means of co-precipitation from salt solutions. Basic work task - to achieve a high uniformity of distribution of oxide between the SiC particles and, hence, a high density material SiC–MeO.

2. Starting materials and methods

The problem of obtaining SiC–MgO–Al₂O₃ homogeneous mixture compositions, SiC–Y₂O₃–Al₂O₃ and SiC–MgO–Y₂O₃–Al₂O₃ can be solved as follows: depositing on 1/3 of SiC powder of magnesium hydroxide as a precipitant using NaOH solution; depositing on 2/3 of SiC powder and aluminum



hydroxides of yttrium, using as the precipitant NH_4OH solution. For the preparation of solutions of magnesium nitrate salt was used, the yttrium and aluminum.

As activators during sintering SiC-material using a mixture of oxides of yttrium and aluminum, forming the step sintering aluminum-yttrium garnet (YAG) and magnesium, yttrium and aluminum, forming a ternary mixture of oxides corresponding to the eutectic composition on the line garnet-spinel ternary system diagram of state $\text{MgO}-\text{Y}_2\text{O}_3-\text{Al}_2\text{O}_3$ (MYA) [5]. The solutions were prepared based on the weight percent based on the material composition 80–95 wt. %. SiC + 5–20 wt. % MeO.

Additional studies to obtain fine-grained oxide powders systems $\text{Y}_2\text{O}_3-\text{Al}_2\text{O}_3$ and $\text{MgO}-\text{Y}_2\text{O}_3-\text{Al}_2\text{O}_3$, held at heat treatment hydroxide at a temperature of 600–700 °C (for $\text{Y}_2\text{O}_3-\text{Al}_2\text{O}_3$ system) and 400–500 °C (for $\text{MgO}-\text{Y}_2\text{O}_3-\text{Al}_2\text{O}_3$ system). The obtained X-ray amorphous synthesized powders have a large specific surface area – $57 \pm 3 \text{ m}^2/\text{g}$ (for $\text{Y}_2\text{O}_3-\text{Al}_2\text{O}_3$ system oxides) and $61 \pm 5 \text{ m}^2/\text{g}$ (for $\text{MgO}-\text{Y}_2\text{O}_3-\text{Al}_2\text{O}_3$ system oxides), with increasing temperature and time of heat treatment specific surface area powders decreases.

The samples heat treated in a vacuum furnace at a temperature of 700 °C. To obtain a molding powder SiC+MeO agglomerates crushed pieces in the cone inertial crusher and ground in a planetary mill to size $d_{0.5} = 1 \text{ }\mu\text{m}$. Feedstock composition SiC+MeO, obtained by co-precipitation, kneaded 2 % solution of an organic binder and pelletized. From the powders were compressed samples of size of $6 \times 6 \times 60 \text{ mm}$ at a pressure of 100 MPa, was dried and sintered in a vacuum furnace in the Ar atmosphere at $1850 \pm 10 \text{ }^\circ\text{C}$ (materials with additive YAG) and $1800 \pm 10 \text{ }^\circ\text{C}$ (MYA materials with additive).

X-ray phase analysis was performed on diffractometer «Rigaku Smartlab 3» with $\text{CuK}\alpha$ -radiation and a Ni-filter; density and porosity of the sintered samples was determined by a hydro-static weighing; Study of the microstructure of the sintered body and fraktogrammy, determining the size of silicon carbide grains and crystals synthesized oxides as well as X-ray fluorescence analysis was performed on an electron microscope «Quanta 200». The grain size distribution was studied using a program «Thixomet Pro» program. The flexural strength was studied in the installation «ShimadzuAG-300kNX». The equipment provided «Engineering Center» SPbSTI (TU).

3. Results of the study

The morphology of the composite powder composition of 85 wt. %. SiC + 15 wt. % MYA obtained by co-precipitation, shown in Fig. 1, where larger particles ($\approx 1 \text{ }\mu\text{m}$) is SiC, small (50–200 nm) oxides. The distribution of magnesium oxide, yttrium and aluminum in the composite powder under stirring from the solution is more uniform. Uniform distribution of MYA and YAG oxides over the surface of SiC powder helps to reduce the melting point of the oxide melt and reduce the liquid-phase sintering temperature of the ceramic while maintaining the high level of physical and mechanical properties of SiC-based composite materials.

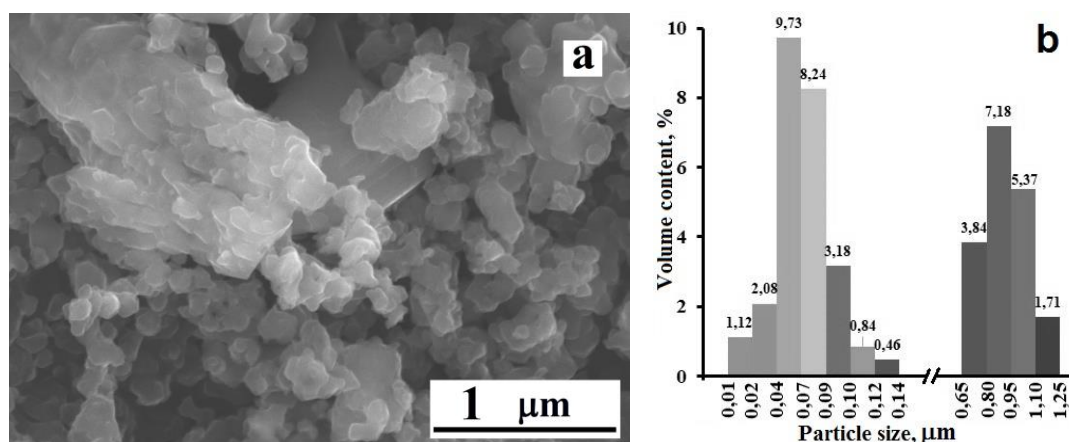


Figure 1. Composite powders 80 wt. % SiC + 20 wt. % MYA (a). Histogram distribution of powder particle size (b)

Oxides of Y_2O_3 and Al_2O_3 obtained by co-precipitation, react to form aluminum yttrium garnet at a temperature significantly lower ($\approx 1000\text{--}1050\text{ }^\circ\text{C}$) [4], than the temperature of fusion of micron-sized YAG powders ($\approx 1500\text{--}1550\text{ }^\circ\text{C}$) [6]. For the system $\text{MgO--Y}_2\text{O}_3\text{--Al}_2\text{O}_3$ characteristic similar dependence reduce synthesis temperature to $900\text{--}950\text{ }^\circ\text{C}$ ceramics based on the SiC deposited by co-precipitation oxides sintered to a dense state at a temperature on the $80\text{--}100\text{ }^\circ\text{C}$ below the temperature of preparation of standard liquid-phase-sintered SiC-material powders of micron-sized – $1930\text{ }^\circ\text{C}$ (with added YAG) [7, 8] and $1860\text{ }^\circ\text{C}$ (with addition MYA) [5]. μm

X-ray phase analysis of the sintered samples showed that the main phase is the $\alpha\text{-SiC}$ (6H modification), the secondary phase - $\text{Y}_3\text{Al}_5\text{O}_{12}$ and the mixture $\text{Y}_3\text{Al}_5\text{O}_{12} + \text{MgAl}_2\text{O}_4$. In this case there was no individual phases MgO , Y_2O_3 or Al_2O_3 , which means that the phase constituents MYA and YAG completely synthesized.

Analyzing the mechanism of liquid phase sintering in accordance with the data [6], we may conclude that during sintering stoichiometric YAG and MYA synthesized at temperatures ≈ 1000 and $900\text{ }^\circ\text{C}$, a shrinkage of the material due to the sliding and rotation of particles SiC relative to each other (at a temperature $\approx 1500\text{--}1600\text{ }^\circ\text{C}$), the material is compacted due to mass transfer and contact sintering of SiC particles ($\approx 1800\text{--}1850\text{ }^\circ\text{C}$).

Densities, porosity, grain size of the SiC and the bending strength of liquid-phase-sintered SiC-materials are given in Table 1. The density obtained SiC-materials increases with increasing content of the oxide additive. The minimal porosity of the material is fixed SiC + 20 wt. % MYA, sintered at ($1800 \pm 10\text{ }^\circ\text{C}$): $P = (0,4 \pm 0,1)\%$, at a density $\rho = 3,36 \pm 0,02\text{ g/cm}^3$ (Table 1). By comparing the properties of standard materials SiC+ YAG (MYA), sintered at 1950 and $1860\text{ }^\circ\text{C}$, with new materials prepared at 1850 and $1800\text{ }^\circ\text{C}$, it can be noted that at the lower temperature sintering process is completed totally. At lower temperatures, mass transfer processes are slowed down more that defines a smaller grain size silicon carbide for material obtained using the co-precipitation method (Table 1). Materials consisting of small grains SiC between which thin layers of oxides are formed, it has a high level of strength properties (Table 1).

When increasing oxide content SiC-material flexural strength (σ_f) increases monotonically. The maximum values of the flexural strength attained in a material of composition 80 wt. % SiC + 20 wt. % MYA, Sintered at $1800\text{ }^\circ\text{C}$ – $\sigma_f = 750 \pm 10\text{ MPa}$ (Table 1). By comparing the obtained result with values of standard materials [5, 6], may be mentioned improve-ment of σ_f by 20–30 %.

Table 1. Physical and mechanical properties of liquid-phase-sintered SiC–MeO materials

Oxide content	Sintering temperature ($^\circ\text{C}$)	Density $\rho \pm 0,02\text{ (g/cm}^3\text{)}$	Porosity $P \pm 0,1\text{ (}\%\text{)}$	Grains size of SiC $d_{0,5} \pm 0,1\text{ (}\mu\text{m)}$	Flexural strength $\sigma_f \pm 10\text{ (MPa)}$
5 wt. % YAG	1850	3,19	2,7	1.4	450
10 wt. % YAG		3,27	2,0	1.6	510
15 wt. % YAG		3,35	1,4	2.2	600
20 wt. % YAG		3,43	0,9	2.7	660
5 wt. % MYA	1800	3,20	1,9	1.1	510
10 wt. % MYA		3,21	1,0	1.2	570
15 wt. % MYA		3,32	0,8	1.4	640
20 wt. % MYA		3,36	0,4	1.6	730

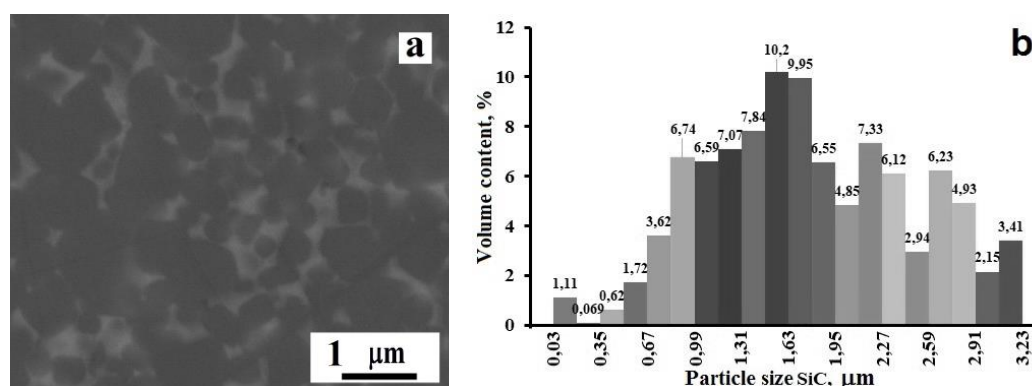


Figure 2. The microstructure of the liquid-phase-sintered SiC-material, supplemented 20 wt. % MYA (a), the histogram distribution of SiC grain size (b)

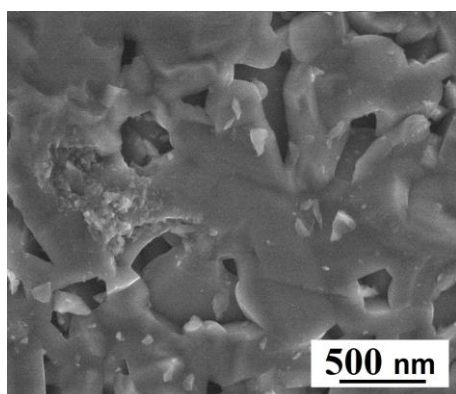


Figure 3. The microstructure SiC ceramics containing 5% by weight. MAY

Figure 2 shows the microstructure of a liquid phase sintered SiC-material comprising 20 wt. % MYA. Grains of SiC, mainly correct (round) form, with uniformly distributed between the oxide phase. Oxides well moisten the surface of the silicon carbide grains materials do not contain SiC particles in the sintered agglomerate.

Fraktogramma destruction SiC-ceramics, sintered with the addition of 5 wt. % MAY at 1800 °C is shown in Figure 3. The crystal size of the oxide phase is much smaller SiC grains, which contributes to a more uniform distribution between the grains of silicon carbide and has a positive effect on the compressibility of liquid-phase-sintered materials.

Elemental analysis of X-ray fluorescence (EDS) showed that silicon carbide grains include phase Si, Mg, Y, Al and O, which indicates that the SiC has a structure «core-shell» where «shell» is a solid oxide solution carbide silicon (containing small amounts of solute elements Mg, Y, Al and O), which confirmed earlier results obtained in [6]. Intergranular phase is characterized by the elements Mg, Y, Al, O and Si, appear due to the dissolution of the SiO₂ film, on the surface of the SiC particles, in the oxide melting with the formation of a siloxisilicate melt.

Application of the method of co-precipitation of the salt solution for a uniform deposition of oxides on the SiC powder to reduce their total number and to receive the high-density SiC-materials with high mechanical characteristics.

The results are a prerequisite for further research in the field of producing high density materials based on SiC, in particular, using as activating additives oxides fusible eutectic systems CaO–Al₂O₃, CaO–TiO₂, MnO–TiO₂ et al.

Acknowledgments

The reported study was funded by RFBR project No 18-38-00383.

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