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Microarticle

Microstructural and physical properties of magnesium oxide-doped silicon nitride ceramics

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ABSTRACT

Silicon nitride based ceramics with aluminum, yttrium and magnesium oxides were produced by cold isostatic pressing and free sintering. The phase composition of the starting MgO powder obtained by the novel technology has been studied. The effect of magnesium oxide content on the structure of the produced materials has been investigated. It was found, that obtained materials with 1 and 2 wt.% of magnesium oxide and without it have a typical β-silicon nitride structure with elongated grains. Ceramics with 5 wt.% magnesia has a duplex α/β-structure with elongated and equiaxed grains. Ceramics with 2 wt.% magnesium oxide has a maximum density of 2.91 g/cm^3. The increases in magnesium oxide content upto 5% led to decrease in the shrinkage (from 16% to 12%) and density (from 2.88 to 2.37 g/cm^3).

Introduction

Recently, much interest has been generated in Si_3N_4 ceramics due to its excellent combination of properties. In particular, silicon nitride has high strength, hardness, thermal stability, etc. [1]. Ceramics based on silicon nitride are often used in many areas of mechanical engineering, engine building, chemical and aviation industries. However, the most important disadvantage of the ceramics is brittleness [2]. It is well known, that due to the bad compactibility of silicon nitride, oxide additives are used. Thus, such oxide additives as MgO, Al_2O_3 and Y_2O_3 are most commonly used for formation of such phases, as SiAlON, $MgSiO_4$ [3] and $MgSiO_3$ [4]. The investigated ceramics was obtained by cold isostatic pressing and frees sintering in a nitrogen atmosphere. There are many well-known commercial methods of obtaining this type of ceramics such as hot isostatic pressing (HIP), spark plasma sintering (SPS), and gas pressing (GP). Nowadays, one of the most important issues is development of cheaper methods of obtaining structural silicon nitride ceramics. Cold isostatic pressing seems to be very beneficial approach to this situation.

The aim of this work is to develop the influence of the additive of magnesium oxide, prepared by novel technology on the structure and some physical properties of ceramics based on silicon nitride.

Experimental procedure

The initial batch included the silicon nitride 85 wt.% Si_3N_4, aluminum 9 wt.% Al_2O_3 and yttrium 6 wt.% Y_2O_3 oxide additives and various contents of magnesium oxide (chemical compositions with 0, 1, 2 and 5 wt.% respectively, see Supplementary Table 1). Powders were mixed in the vibratory disk mill (RS200 Retsch). Then, the mixture of powders was compacted by cold isostatic pressing (EPSI CIP 400 B-9140). Sintering of green bodies was performed in a vacuum furnace (VHT 8/22-GR) at 1800 °C temperature in a nitrogen atmosphere.

Microstructure characterization of sintered ceramic was performed by electron ion microscope Quanta 600 FEG equipped with integrated microanalysis system Pegasus 2000 (SEM-EDS).

Proposed magnesium oxide powder was synthesized by thermal plasma in a novel thermal DC plasma torch using magnesium nitrate hexahydrate. $Mg(NO_3)_2{\cdot}6H_2O$ was obtained from the serpentinite. The serpentinite consisted mostly of magnesium, silicon and iron in the serpentinite form ($Mg_3Si_2O_5(OH)_4$); lizardite and it was found in the Khalilovsky deposit of the Orenburg region in Russia. Serpentinite was dissolved in 40% nitric acid solution. $MgNO_3$ solution was obtained after the ions like Fe^{3+} and Fe^{2+} were transformed into hydroxide precipitates, and the precipitates were separated by filtration. $MgNO_3$-rich solution was transferred to a

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glass beaker for evaporation of the solution. The solution started boiling at 90 °C and was boiled for 4 h in order to evaporate most of the solvent. The residue of hydrated magnesium carbonate was cooled to room temperature and filtered. $Mg(NO_3)_2{\cdot}6H_2O$ was ground by a vortex jet flow type mill (productivity of 50 g per minute, air pressure of 10 bar, the air volume of 1 m^3/min).

Nanoscale pure magnesium oxide was obtained by a novel thermal DC plasma torch using $Mg(NO_3)_2{\cdot}6H_2O$. The powder is separated during the passage of the gas–dust mixture through a system of cyclones, and the gas mixture is utilized in the venturi scrubber.

Investigations of the phase composition of the MgO powder were carried out by Rigaku Ultima IV X-ray powder diffractometer with CuKα radiation. Crystalline phases were identified by the ICDD PDF-2 (2008) powder diffraction database. The microstructure of the starting powders of magnesium oxide and silicon nitride was investigated by the transmission electron microscope (TEM) JEM 2100 (JEOL Ltd., Tokyo, Japan).

Results and discussion

It is well known that, such characteristics of the starting powder as purity, morphology, particle shape and size have a significant influence on the properties of sintered ceramics. At the same time, the most important effect on these properties provides a method of powder production. XRD of the initial magnesium oxide powder (Supplementary Fig. 1) clearly demonstrates the purity of this material. In particular, it has been identified only one phase of magnesium oxide (so-called periclase) with a cubic crystal lattice of the Fm-3 m space group with a 4.215 Å lattice constant.

Supplementary Fig. 2a shows the starting commercial silicon nitride powder image, obtained by TEM. It may be noted that, the starting silicon nitride powder has 1 μm agglomerates. In turn, particles of magnesium oxide powder obtained by the above-described technology shown in Supplementary Fig. 2b. The particles size of this powder varies from 100 to 150 nm. Observed particles are equiaxial and have a globular shape (Supplementary Fig. 3).

Analysis of the structure by SEM showed that the produced material has a moderately dense structure with a uniform distribution of pores on the surface. It was determined, that the obtained material is characterized by an elongated structure typical of β-silicon nitride. In all three cases 0, 1 and 2 wt.% was the observed structure with elongated grains (Supplementary Table 2). An average grain size of Si_3N_4 with 1 wt.% magnesium oxide was 2.2 μm, while the grain size of the ceramics with 2 wt.% magnesium oxide was 3.4 and 3.1 μm without magnesium oxide, respectively. Duplex α/β-Si_3N_4 structure was observed for the 5 wt.% MgO-doped composition. In the case of 5% magnesium oxide the average grain size was 2.4 μm for elongated grains and 1.2 μm for equiaxial grains (Supplementary Table 2). The manufacturing process of the similar 1650 °C sintered material without magnesium oxide, as well as its mechanical and physical properties are described in our previous works and it was characterized by the submicron equiaxed structure and moderately high strength [5–7].

Supplementary Fig. 4 shows some properties of the produced ceramic. In particular, in Supplementary Fig. 4,a shows the linear dependence of the shrinkage on the magnesium oxide content wt.%. Particularly, an increase in the content of magnesium oxide from 1% to 5% is accompanied with a decrease in the shrinkage from 16 to 11 wt.%. Maximum weight loss (8%) as well as the lowest density, 2.37 g/cm^3, has ceramics with 5 wt.% magnesium oxide (Supplementary Fig. 4b and c). Nearly the same density (2.88–2.91 g/cm^3) and weight loss (2.6–5.2%) were observed for ceramics with a small amount of magnesium oxide and without magnesia (1, 2 and 0 wt.%, respectively).

Conclusions

Silicon nitride based ceramics was produced by cold isostatic pressing and free sintering with different contents of magnesium oxide. Al_2O_3–MgO–Y_2O_3 oxide additive system was used. Nanoscale pure magnesium oxide was obtained by a novel thermal DC plasma torch using $Mg(NO_3)_2{\cdot}6H_2O$ which were obtained from serpentinite ore distributed in the Halilovskiy array (Russia, Orenburg region). The test results suggest that the obtained magnesium oxide nanopowder with high purity has the periclase structure and very homogeneous structure without any observable pores. From our present work, it is concluded that the serpentinite ore as a cheap source material and a novel thermal DC plasma torch could be used for the production of MgO nanopowders. Compositions with 0, 1, and 2 wt.% magnesium oxide have a typical β-silicon nitride structure with elongated grains, whereas the composition with 5 wt.% magnesium oxide is characterized by the duplex α/β structure. The increases in magnesium oxide content upto 5% led to a decrease in the shrinkage and density and increasing mass loss. Ceramics with 2 wt.% magnesium oxide has a maximum density of 2.91 g/cm^3.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.rinp.2016.01.005.

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