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The analysis of strength properties of ceramic preforms for infiltration process

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ABSTRACT

Purpose: The goal of this work is the optimization of sintering process of the ceramic preforms based on Si_3N_4 and Al_2O_3-Ti(C,N) materials. The influence of pore forming additives on porosity, microstructures and compressive strength are investigated. The aim of this study is to obtain the nitrides and carbides base preforms material for the infiltration process of molten aluminium alloys.

Design/methodology/approach: The method of obtaining the silicon nitride and oxide-carbonitride porous preform for the infitration process is the free sintering process. The preforms were produced by the mixing of ceramic powders with organic binders, drying and sintering. Porosity, density were measured for the materials. Microstructure observation was carried out using scanning microscope. The compressive strength of Si_3N_4 and Al_2O_3-Ti(C,N) porous preforms were investigated.

Findings: For sintered porous Si_3N_4 preforms, influence of the porous forming additives on material porosity is observed. Compressive strengths of Si_3N_4 were in the range of 2.9-4.8 MPa. The highest value of the compressive strength was obtained for Al_2O_3-Ti(C,N) preform with 8 wt.% of tylose and 25 wt.% of glykol. For these materials compressive strength were in the range of 13.2 up to 14.3 MPa. In spite of lower value of the compressive strength for Si_3N_4 preforms, this material exhibits high shock thermal resistance.

Practical implications: Pressureless infiltration of molten metals into ceramics is the most cost-effective approach to liquid-metal processing of MMCs. Metal matrix composites are applied widely in aircraft production technologies and defence technology.

Originality/value: Infiltration of molten metals into porous ceramic preforms is the only technique suitable for the fabrication of high volume fraction of ceramic materials in MMCs. Infiltration process generates thermal stresses in the ceramics preforms. The thermal shock resistance of Al_2O_3 is lower than Si_3N_4 or Al_2O_3-Ti(C,N) materials. New kinds of porous materials were obtained.

Keywords: Porous ceramics; Si_3N_4; Alumina carbonitride; Stregth propetries

PROPERTIES

1. Introduction

Cermets could be produced by infiltrating pre-sintered ceramic skeleton bodies with molten metal. There are several approaches for the fabrication of the micro/macro porous ceramics using: the controlling the particle size of ceramics forming pressure or/and sintering time, the mixing of the ceramic powders with bubble former, the mixing ceramic powders with additional organic particles, which vaporize form small homogeneous pores [1,2].

Composite materials reinforced by ceramic materials are characterised by the very good mechanical properties over a wide

range of temperatures, and are applied widely in aircraft production technologies and defence technology [3,4]. Two basic techniques are used in production of the ceramic-metal materials: casting and powders metallurgy [5,6]. Pressureless infiltration of molten metals into ceramics is the most cost-effective approach to liquid-metal processing of metal-matrix composites. Infiltration of molten metals into porous ceramic preforms is the only technique suitable for fabrication materials with the high volume of ceramics material fraction. Molten metal infiltration can be classified into three categories, based on the source of driving force pressure assisting, vacuum driven and pressureless or capillarity driven [7,8,9].

The most popular material for porous preforms is Al_2O_3 because of it low cost. But infiltration process generates effect of thermal stress in the Al_2O_3 preforms [10]. Porous alumina ceramics are the relatively cheap reinforcing material but it does not develop an interface with aluminium liquids, also ceramic preforms are contained more expensive materials than alumina, like SiC, Si_3N_4, AlN, TiB_2, B_4C compounds. For porous material applied for the infiltration process, the important are: resistance for thermal shock, the open pore structure in preform good mechanical properties and good reaction between the infiltrated metal and ceramic preforms.

In spite of excellent properties, the potential of porous ceramics has not been fully realized because of their defects such as lack of pore size control, lack of continuous processing method, and absence of a model relating pore structure to mechanical properties [1].

In these studies, silicon nitride as preforms to the infiltration process, was choose. Si_3N_4 is a highly covalent compound. The strongly covalent bonds of Si_3N_4 produce a number of desirable engineering properties: high strength, thermal stability up to approximately 2120 K, where is decomposed, good oxidation resistance, low coefficient of thermal expansion (good thermal shock resistance), modulus of elasticity greater then many metals. Solid - state free sintering requires additives to achieve full density of this material. Sintering agent like MgO, Y_2O_3, CeO_2, ZrO_2, Al_2O_3 oxides or AlN, TiN, ZrN, CrN, Mg_2N_3 nitrides, makes possible to sinter silicon nitrides at 2150 K to 2400 K and at pressures up to 2 MPa with reported resulting density higher than 99.3% of theoretical [11]. Silicon nitride material has a high hardness at temperatures between 870-1270 K. Generally, for silicon carbide preform, the most important properties are that: Si_3N_4 is shock resistant, with high thermal conductivity and a moderate thermal expansion.

Hard particles of titanium carbide and/or nitride in alumina increase the materials hardness at temperature up to 1073 K, when compared to pure oxide ceramics. In the same time, the fracture toughness and bending strength is improved through crack impediment, crack deflection or crack branching caused by dispersed hard particles [12]. The higher hardness together with higher toughness of such composite material increases its resistance to abrasive and erosive wear considerably while its lower thermal expansion and higher thermal conductivity contribute to better thermal shock resistance and thermal shock cycling capabilities when compared to nonreinforced oxide ceramics [12].

The previous authors paper was related to wettability studies for compacts with 90 wt.% α-Si_3N_4, 5 wt.% Al_2O_3 and 5 wt.% Y_2O_3 and 68 wt.% Al_2O_3, 2 wt.% ZrO_2, 30 wt.% of Ti(C,N) and Al, AlSi11 alloy [13,14]. The results obtained for contact angle and shear strength of selected couples are very promising for application of the produced porous preforms in manufacture of composite materials of the Al-Al_2O_3-Ti(C,N) and Al-Si_3N_4 systems since there is strong metal/ceramic bonding. In cases where the molten metal does not wet the porous preform material sufficiently; the molten metal may not completely infiltrate the sintered skeleton under the action of capillary forces. In such situation the pores are not filled completely, even where pressure infiltration in autoclave is used [15].

In this paper, porosity, microstructures and compressive strength of Si_3N_4 and Al_2O_3-Ti(C,N) porous preforms were investigated.

2. Experimental procedure

Mixture of powders for Si_3N_4 porous material is composed of 90 wt.% of α-Si_3N_4 (H.C.Starck, AEE, 1-5 μm), 5 wt.% of Al_2O_3 (Alcoa, A16SG, <0.5 μm) and 5 wt.% of Y_2O_3 (Fluka). Mixtures of powders are milled in the alumina rotary mill in the ethyl alcohol medium with porous forming additives: 15 wt.% and 20 wt.% tylose MH 1000 (Fluka) or mixture of tylose with polyethylene glycol 6000 (PS PARK). For Al_2O_3Ti(C,N) porous preforms mixture of powders was composed of 68 wt.% Al_2O_3 (Alcoa, A16SG), 2 wt.% ZrO_2 (Fluka, <0.5 μm) and 30 wt.% of Ti(C,N) (H.C.Starck).

After milling, the powder mixtures were dried and granulated. Discs of 16 mm diameter and a thickness of 16 mm were formed by single action pressing at pressure of 30 MPa and dried at 498 K during 48 hours. Initial sintering for Si_3N_4 porous preforms was carried out using the furnace MOV Balzers at the heating rate of 600 K/h, at 873 K during 1 h. Samples were free-sintered at nitride boride packing material in nitride atmosphere at 1880 K for 1 h, using the GERO HTK 8/22G furnace. Initial sintering for Al_2O_3Ti(C,N) porous preforms was carried out using the MOV Balzers vacuum furnace at the heating rate of 600 K/h, in the 873 K for 1 h. Next, samples were free-sintered at this same furnace at 1973 K and 10^{-4} T for 1 h.

Densities and porosities of sintered samples were measured according to the Archimedes principles [2]. For samples the compressive strength was measured using INSTRON TT-DM apparatus with initial rate of strain 10^{-4} s^{-1}.

3. Results and discussion

During the initial studies, methocel, polyvinyl alcohol, triethanolamine and glycol 10000 and 3000 were used like pores forming additives due to their decomposition [4]. But these materials are not considered for further examination because of very low porosity of produced Si_3N_4 materials (below 20%). Only tylose and glycol 6000 were choose for sintering process. In the present works 1.1 μm grain size of Si_3N_4 powder was changed and the 1-5 μm grain size Si_3N_4 powder was used. Some selected physical properties of porous Si_3N_4 samples containing 15 wt% up to 60 wt.% porous forming additives and

Al_2O_3-Ti(C,N) with 8 wt.% of tylose and 25 wt.% of glycol are presented in Table 1.

Table 1.
Apparent density, open porosity, percent of theoretical density for compacts with 90 wt.% α–Si_3N_4 (AEE, 1-5 μm) submicro-powders, 5 wt.% Al_2O_3 (Alcoa, A16SG, particles size <0,5 μm), 5 wt.% Y_2O_3 (Fluka) and 68 wt.% Al_2O_3 (Alcoa, A16SG), 2 wt.% ZrO_2 (Fluka, <0.5 μm) and 30 wt.% of Ti(C,N) (H.C.Starck)

Base material	Porous forming additives wt.%	Apparent density, g/cm^3	Open porosity, %	Theoretical density, %
Si_3N_4	Tylose - 15	1.71	46.8	51.9
Si_3N_4	Tylose - 20	1.38	57.9	41.9
Si_3N_4	Tylose - 5 Glycol - 40	1.45	55.1	44.1
Si_3N_4	Tylose - 20 Glycol - 40	1.04	68.1	31.5
Si_3N_4	Tylose - 20 Glycol - 10	1.26	61.4	38.1
Si_3N_4	Tylose - 20 Glycol - 20	1.17	64.2	35.4
Al_2O_3-Ti(C,N)	Tylose - 8 Glykol - 25	1.94	54.5	44.9

For the porous materials on the Si_3N_4 base, the highest porosity value was obtained for the tylose participation of 20 wt.%. Increasing of the tylose content has higher influence on porosity of material than the glycol content increasing from 10 wt.% to 40 wt.%. For the application of additives, tylose and glycol together there is the increasing of the preforms water absorbability. Comparison of open porosity for Si_3N_4 preforms depend on the type of porous forming additives are presented in the Fig. 1. Compressive strength were measured for six samples of Si_3N_4 preforms obtained with 20 wt.% of tylose and 20 wt.% of glycol. Second group of material was $Al_2O_3Ti(C,N)$ preforms with 8 wt.% of tylose and 25 wt.% of glycol addition (six samples was measured). The typical curves of compressive strength measurements for Si_3N_4 and for Al_2O_3-Ti(C,N) porous preforms are presented in the Fig. 2. Stress-strain curves porous ceramic preforms are other that for non porous brittle ceramic compacts. The strength is not so high but curves are similar to plastic material deformation. Al_2O_3-Ti(C,N) preform characterized by the few step deformation, Fig. 2. During the compressive test layers are deformed and on the curves the stress decreasing are occurred (Fig. 2).

Compressive strengths of Si_3N_4 preforms were in the range of 2.9-4.8 MPa while for Al_2O_3-Ti(C,N) preforms, it were in the range of 13.2 up to 14.3 MPa. The reason of the low value of the compressive strength for Si_3N_4 preform could be sinterability of large 1-5 μm Si_3N_4 particles.

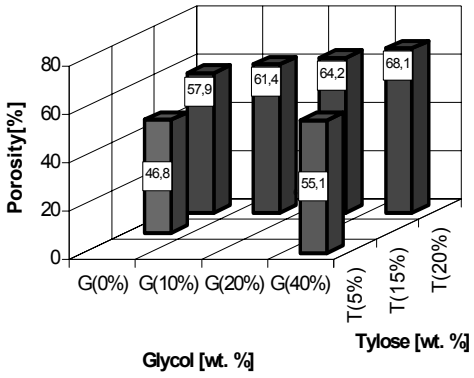

Fig. 1. Influence of porous forming additives (T-tylose; G-glycol 6000) on open porosity of Si_3N_4 preforms

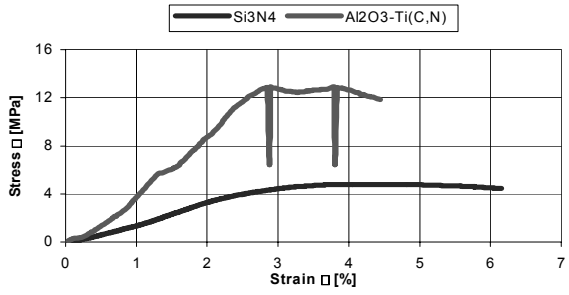

Fig. 2. Stress-strain curves for the Si_3N_4 preform (64.2% open porosity) and for Al_2O_3-Ti(C,N) preform (54.5% open porosity)

Previous works confirm the grain size influence on the porosity of materials. The porosity increases in proportion to the grain size increasing. The grain size increasing have bad influence on sinterability of material and it is inversely proportional to the strength of materials. For the sintered ceramic materials the best sinterability show approximately 1 μm powders but porosity of preforms for this particle size is low (below 20%). The sintering process of Si_3N_4 with grain size over 10 μm is not possible because of low diffusion on the grains boundaries during the sintering process.

Materials are characterized by open porosity. For all samples, the infiltration process in vacuum by fuchsine ($C_{20}H_{20}N_3Cl{\cdot}4H_2O$) was carried out, showing complete infiltration in all bulk of each sample.

4. Conclusions

For sintered porous Si_3N_4 preforms, influence of the porous forming additives on material porosity is observed. Tylose and glycol application cause the water absorbability increasing. Compressive strength of the porous Si_3N_4 preforms is low, due to the large grain size of powders used for the sintering process. Large grain size of powders preserves higher porosity of material but have the influence on the material sinterability and strength properties. Compressive strength of Si_3N_4 was in the range

of 2.9-4.8 MPa. The highest value of the compressive strength was obtained for Al_2O_3-Ti(C,N) preform with 8 wt.% of tylose and 25 wt.% of glycol. For these materials compressive strength were in the range from 13.2 up to 14.3 MPa. In spite of lower value of the compressive strength for Si_3N_4 preforms, this material exhibits high shock thermal resistance. For Si_3N_4 porous preform the optimization of grain size is needed. Application of particles with smaller size should result in the compressive strength increasing.

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