Available online at www.sciencedirect.com
SCIENCE DIRECT®
Journal of the European Ceramic Society 25 (2005) 801–807
www.elsevier.com/locate/jeurceramsoc

Effect of AlN-content on the microstructure and fracture toughness of hot-pressed and heat-treated LPS–SiC ceramics

K. Strecker [a,*], M.-J. Hoffmann [b]

[a] *Department of Materials Engineering, Faculty of Chemical Engineering of Lorena, Polo Urbo Industrial, Gleba AI-6, s/n, 12600-000 Lorena, SP, Brazil*
[b] *Institute for Ceramics in Mechanical Engineering, University of Karlsruhe, Haid-und-Neu-Str. 7, D-76131 Karlsruhe, Germany*

Received 28 October 2003; received in revised form 26 January 2004; accepted 30 January 2004
Available online 20 June 2004

Abstract

The influence of additive content on the microstructural development of hot-pressed and heat-treated LPS–SiC has been investigated using $AlN–Y_2O_3$ mixtures at a molar ratio of 80:20, varying the total amount from 5, 10, 15 to 20 wt.%. Specimen were hot-pressed at 1900 °C for 1 h in nitrogen atmosphere under an applied pressure of 25 MPa and subsequently heat-treated at 2000 °C for 1, 2, 4 and 8 h.

It has been found that the transformation rate of β- into α-SiC is retarded by higher AlN-contents and the formation of the 6H α-SiC polytype is favored. Furthermore, grain growth during annealing is also effectively inhibited. While hardness remained almost unchanged, fracture toughness varied with additive content and/or duration of the heat-treatment. Fracture toughness increased during the first 1 or 2 h of annealing,depending on the AlN-content, and diminishing for more prolonged treatments. The maximum fracture toughness has been determined for samples containing 10 wt.% of additives, hot-pressed and annealed during 1 h.

Keywords: SiC; Liquid phase sintering; Microstructure-final; Phase analysis; Fracture toughness

1. Introduction

Silicon carbide is a compound of relatively low density, high hardness, elevated thermal stability and good thermal conductivity, resulting in good thermal shock resistance. Because of these properties, SiC materials are widely used as abrasives and refractories.

However, it has not been possible to sinter SiC to theoretical density without additives or external applied pressure, because of its covalent bonding character. The conventional densification route of SiC via solid-state sintering using small quantities of B or Al and C or compounds thereof[1–3] is widely applied today to produce components on a commercial base. The driving forces for densification are volume and surface diffusion which are enhanced by the addition of boron and aluminium. In order to increase the diffusion coefficient, sintering temperatures in the range of 2050–2200 °C are applied.

An innovative approach to solid-state sintering of SiC has been initiated in the 1980s by Omori and Takei,[4] who sintered SiC to high densities via liquid-phase sintering using Al_2O_3 and Y_2O_3 as additives. Since then a growing interest for liquid-phase sintered SiC ceramics is noted, because this type of material offers the opportunity of increasing fracture toughness by controlling the microstructure.[5–10] In general, the microstructural control is achieved making use of the β- to α-SiC phase transition. The formation of platelet shaped α-SiC grains is thereby influenced by the α/β-SiC ratio of the starting powders, the additive system beside the sinter parameters temperature, time and atmosphere. Fracture toughness values of 6–7 MPa $\sqrt{m}$ have been reported, signifying an almost 100% increase when compared to solid-state sintered SiC ceramics.

Rixecker et al.[11–13] investigated the pressureless sintering with oxynitride additives, starting from $AlN–Y_2O_3$ powder mixtures. They report that using AlN as additive and a nitrogen sintering atmosphere the weight loss during sintering can be effectively minimized, because the decomposition of AlN is avoided by the nitrogen atmosphere. On the other hand, it is known that a nitrogen atmosphere re-

* Corresponding author. Tel.: +55-12-5533174; fax: +55-12-5533006.
E-mail address: strecker@demar.faenquil.br (K. Strecker).

0955-2219/$ – see front matter © 2004 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jeurceramsoc.2004.01.024

duces the sintering rates and also hinders the β- to α-SiC transformation.[14–16]

This work investigates the influence of the A1N–Y_2O_3 additive content on the β to α-SiC phase transition in hot-pressed and heat treated LPS–SiC ceramics, on the microstructure and its influence on fracture toughness and hardness.

2. Experimental procedure

Powder batches were produced using β-SiC powder (Grade B10, H.C. Starck), with an average particle size of 0.79 μm and a specific surface area of 13 m^2/g. The additives were AlN (Grade C, H.C. Starck) and Y_2O_3 (Grade Fine, H.C. Starck) with average particle sizes of 1.05 and 1.07 μm and specific surface areas of 7 and 15 m^2/g, respectively. Furthermore, α-SiC powder (FCP-15, Norton, AS) was used. Four powder batches were prepared, each containing 1 wt.% of α-SiC to act as seeds. The additive content varied from 5, 10, 15 to 20 wt.% of the total mixture, maintaining a constant molar ratio of AlN:Y_2O_3 of 80:20. The compositions prepared are listed in Table 1.

Mixing was performed by attrition milling using Si_3N_4 balls as grinding media and isopropylic alcohol as vehicle. After mixing, the powder batches were dried in a rotary evaporator, crushed and screened through a 325 mesh sieve. After attrition milling, the powders had almost identical particle size distributions (see Fig. 1), with average particle sizes of 0.66 μm.

Samples were hot-pressed at 1900 °C for 1 h under an applied pressure of 25 MPa in nitrogen atmosphere. The heating rate was 10 °C/min and the cooling rate about 20 °C/min, until the inertia of the furnace prevailed. The final shape of the hot-pressed samples were discs of approximately 25 mm diameter and 7 mm height. Samples were cut from the discs and further heat-treated at 2000 °C under 0.2 MPa flowing nitrogen atmosphere during 1, 2, 4 and 8 h. Bulk densities were measured by the Archimedes' method. Observation of the microstructural development has been performed by scanning electron microscopy on fracture surfaces and polished and plasma etched surfaces. The phase composition of the hot-pressed and heat-treated samples was determined by X-ray diffraction of crushed specimen using Cu-K_α radiation, a step width of 0.01° with an exposure time of 5 s per position. Quantitative phase analysis of the SiC polytypes was conducted using a programme based on the work of Ruska and Gauckler.[17] For computation the intensities of the peaks at 2θ 33.7, 34.2, 34.9, 35.7, 38.2 and 41.5° were determined.

Table 1
Designation and composition of powder mixtures

Sample designation	β-SiC (wt.%)	α-SiC (wt.%)	AlN (wt.%)	Y_2O_3 (wt.%)
A80-5	94	1	2,103	2,897
A80-10	89	1	42,065	57,934
A80-15	84	1	63,099	86,901
A80-20	79	1	84,130	115,868

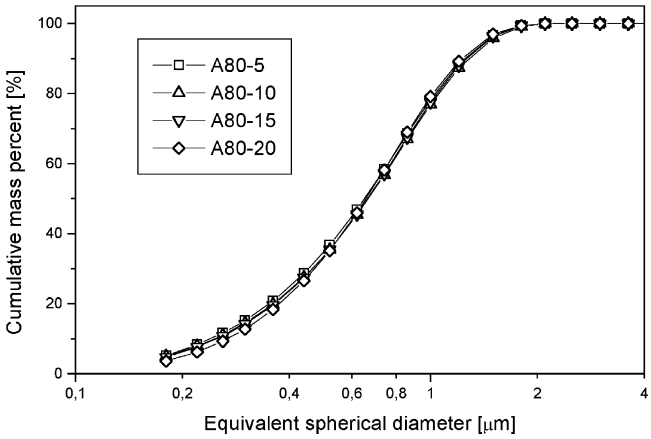

Fig. 1. Particle-size distributions of attrition milled powder batches.

Hardness was determined by Vickers indentation under a load of 9.81 N. The fracture toughness was calculated by the length of the cracks originating from the edges of the indentation marks, using the equation described by Niihara et al.[18]:

$$K_{I_c} = 0.018 HV\sqrt{a}\left(\frac{E}{HV}\right)^{0.4} \times \left(\frac{c}{a} - 1\right)^{-0.5} \quad (1)$$

where K_{I_c} is the fracture toughness of the material, HV the Vickers hardness, E the Young's modulus (for LPS–SiC a value of 400 GPa was assumed), c the crack length and a the half indentation diameter.

3. Results and discussion

All samples reached final relative densities higher than 98.5% t.d. after hot-pressing. A slight decrease of the relative densities of the further heat-treated samples was observed with increasing duration of the treatment, but limited to less than 0.5% for all compositions studied.

Fig. 2 shows fracture surfaces of the hot-pressed samples. While the overall grain sizes remained very fine for all compositions investigated, it can be noted that the grain sizes decrease with increasing additive content. Furthermore, the grain shape remained globular, only sample A80-5 exhibits some hexagonal, platelet like shaped grains, indicating that under the hot-pressing conditions, 1900 °C during 1h under nitrogen atmosphere, the β- to α-SiC phase transformation did not occur and that high AlN-contents retard the phase transition, in agreement with observations published by Nader et al.[9]

With increasing duration of the heat-treatment, grain growth occurs, as demonstrated in Fig. 3, using specimen of composition A80-10 as example. Furthermore, the