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ScienceDirect

Ceramics International 41 (2015) 4308–4311

CERAMICS INTERNATIONAL

www.elsevier.com/locate/ceramint

Effect of AlN content on properties of hot-press sintered Sialon ceramics

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Received 4 November 2014; received in revised form 19 November 2014; accepted 21 November 2014
Available online 29 November 2014

Abstract

This article concerns a hot-press sintered Sialon ceramic using Si_3N_4 and AlN as raw materials, Y_2O_3 and La_2O_3 as additives. The influences of AlN adding amount (5～30 wt.%) on the sintered densification, phase compositions and microstructures are discussed. Results showed that the β-Sialon ceramic could be obtained by hot-press sintering of 95～80 wt.% Si_3N_4, 5～20 wt.% AlN and extra 5 wt% Y_2O_3-La_2O_3 additives, while the excess of 25 wt.% AlN would lead to the transformation from β-Sialon to α-phase. The relationships of AlN contents with bending strength and thermal conductivity of the sintered composites were also involved in the study. For the composition of 80 wt.%Si_3N_4-20 wt.%AlN, the sintered sample showed the fracture strength of 408 MPa and the highest thermal conductivity of 34 W/(m · K) which is 44.7% higher than that of the sample at an adding AlN content of 5 wt.%.

Keywords: AlN; Sialon ceramics; Hot-press; Thermal conductivity

1. Introduction

The discovery of Si-Al-O-N system, a solid solution of Si_3N_4 and Al_2O_3, by Oyama [1] and Jack [2] in the early 70 s has attracted the gaze of people on the new high-temperature structural ceramics. It has been well acknowledged that Sialon ceramics have the similar physical performance with Si_3N_4 and excellent chemical properties parallel to Al_2O_3[3–5]. Generally, the sintered Sialon ceramics can be divided into two categories: β-Sialon in accordance with the common formula $Si_{6-z}Al_zO_zN_{8-z}$ with $0 < z < 4.2$ and α-Sialon in accordance with the formula $Me_{m/val}(Si, Al)_{12}(O, N)_{16}$ where Me can be vacancies or metal such as Y, Ca, Mg, Li and lanthanides [6].

Zhen-Kun Huang [7] fabricated β-Sialon and α-Sialon ceramics with incorporated Y and La by pressureless sintering Si_3N_4 using $(Y, La)_2O_3$-AlN as additives. In order to obtain dense materials, Ilmars Zalite et al.[8] prepared α- and α-/β-Sialon composites using five plasma-synthesized nanopowders such as Si_3N_4, the mixture of 90%Si_3N_4-10%AlN, 90%AlN-10%Y_2O_3, Si_3N_4-AlN-Al_2O_3-Y_2O_3 and Y_2O_3 with three different starting compositions of Si_3N_4-(9AlN · $1Y_2O_3$), Si_3N_4-AlN and Si_3N_4-AlN-Al_2O_3-Y_2O_3.

It has been reported that β-Sialon exhibited enhanced fracture toughness and strength as well as high thermal conductivities compared to α-Sialon due to their rod shape grain structure [9,10]. In addition, β-Sialon showed non-toxicity, good electrical insulation and thermal shock resistance properties, which enabled it to be a promising candidate for high-speed circuits and power devices as heat dissipation and packaging material [11]. Although the preparation of Sialon ceramics with additives such as Y_2O_3 and other rare-earth oxides has been researched, few studies were conducted on the influence of AlN addition on the phase evolution, densification behaviors, and thermal and physical properties of Sialon ceramics including the β-Sialon phase.

In this article we explored the effect of AlN content on the microstructures and properties of Sialon ceramics. The AlN powders were added from 5 wt.% to 30 wt.% with Si_3N_4 as the

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http://dx.doi.org/10.1016/j.ceramint.2014.11.118
0272-8842/

raw materials, extra 5 wt.% Y_2O_3-La_2O_3 (La_2O_3:Y_2O_3=3:2) as sintering additives. The mixtures were sintered at 1750 °C by hot-pressing method.

2. Experimental procedures

Commercially available AlN powders (D_{50}=0.5 μm, AlN > 99%, specific surface 0.92 m^2/g, Toyo Aluminium Co. Ltd., Japan.) and α-Si_3N_4 powders (D_{50}=0.518 μm, α-Si_3N_4 > 93%, a small amount of oxygen and free silicon, Beiing Tsinghua Unisplendor Founder High-Tech Ceramics Co. Ltd.) were used as the starting materials. Y_2O_3 and La_2O_3 in high purity were used as sintering additives in amounts of 5 wt.% (La_2O_3:Y_2O_3=3:2). The mixtures were ball milled for 2 h using alcohol as a mixing medium with the mass ratio of 1:2:0.8 for the powders, balls and alcohol. After drying and pelleting, the mixtures were sintered by verticle vacuum hot-pressed furnace. The sintering under uniaxial pressing was carried out with a 10 °C/min heating rate up to a temperature of 1750 °C for 0.5 h under pressure of 30 MPa in N_2 atmosphere.

The sintered density was measured by the Archimedes method using water. The phase analysis was performed by X-ray diffraction (XRD, X'pert Pro MPD, Japan). The microstructures including the crystal morphology and pore distribution were characterized by scanning electron microscope (SEM, JSM-6308LA, Japan). The measurement of flexural strength was performed by three-point bending method, in which 3 mm × 4 mm × 30 mm test pieces were applied. 2 mm × 4 mm × 30 mm specimen pieces were prepared for measurements of fracture toughness by single edge notched beam test with a cross head speed of 0.05 mm/min. The laser flash method was applied to determine the thermal diffusivity at ambient temperature by JR-3 laserflash thermal analyzer in which a disk-shaped specimen was fabricated and placed in the middle of the analyzer furnace.

3. Results and discussion

3.1. Phase formation and microstructure analysis

Fig. 1 shows the X-ray diffraction patterns of the sintered samples with different AlN addtions. It revealed that two kinds of Sialon phases, α- and β-Sialon, were detected. When the adding contents of the AlN were 5 wt.%, 10 wt.% and 15 wt. %, the dominant phase of the samples was β-Sialon, while both α-Sialon and β-Sialon were detected for the samples at an adding AlN content of 20 wt.%. With the continuing increase of AlN, the dominant phase transformed from β-Sialon to α-Sialon. For the samples with AlN content of 25 wt.% and 30 wt.%, the main peaks in Fig. 1(E) and (F) belong to α-Sialon, together with very weak peaks of AlN.

According to the isothermal phase diagram of the Si-Al-O-N system by Gauckler et al.[12], the compositions of 95～80 wt.% Si_3N_4 and 5 wt.%～20 wt.% AlN leads to the formation of β-Sialon for a temperature slightly below 1760 °C while other phases such as α-Sialon would be detected instead out of the compositions. The present results by X-ray diffraction were in accordance with the phase diagram perfectly. The transformation from β-Sialon

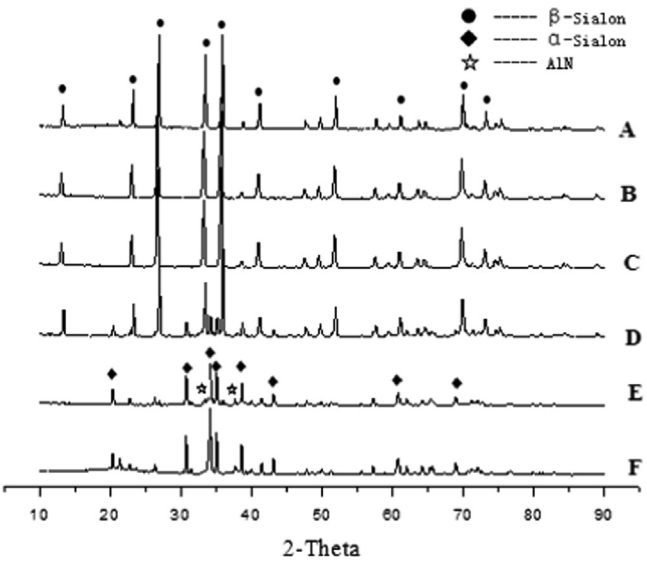

Fig. 1. XRD patterns of the sintered samples with different AlN contents. (A: 5 wt.%; B: 10 wt.%; C: 15 wt.%; D: 20 wt.%; E: 25 wt.%; F: 30 wt.%).

to α-Sialon with the increasing AlN content could be explained as following: When α-Si_3N_4 was used as starting material, α-Sialon can be formed directly from α-Si_3N_4. The thermal stability of α-Sialon was so weak that it tended to transform into β phase at 1300 °C or higher temperatures [13]. However, the compositions of 20～30 wt.% AlN addition in the starting powders favoured the stability of α-Sialon or the conversion of β-Sialon to α phase so much. It indicated that the phase evaluation of Sialon ceramics was greatly dependent on the proportion of AlN in the starting powders and the pure β-Sialon can be obtained conditionally at 1750 °C.

Fig. 2 displays the SEM images of the fracture surfaces for samples, showing their crystal morphology and microstructural features. Yet the microstructures of these samples are so unlike that they change from elongated grains to equiaxed grains. When the AlN contents were 5 wt.%, 10 wt.%and 20 wt.%, many grains in the shape of long rods which stacked or overlapped each other were observed. It can also be found that the sample with 10 wt.% of AlN has a larger number of elongated β-Sialon grains in good crystallization than those samples with 5 wt. % and 20 wt. % of AlN addition. When the AlN content increases, the β-Sialon tended to transform to α-Sialon shown in the Fig. 1 and smaller grains which were homogeneous in structures were found in the Fig. 2d and e, indicating that an excess of AlN addition is responsible for the formation of α-Sialon phase in Sialon ceramics.

3.2. Densification behavior

Fig. 3 is the relative densities of the samples with different amounts of AlN, ranging from 84.7% to 96.1% as the AlN content varied from 5 wt.% to 30 wt.%. The relative density slightly increases with increasing AlN content up to 10 wt.%, and keeps almost constant with 5 wt.% additions at 10 and 20 wt.% of AlN, then decreases as the AlN content further increases. The relative density of 96.1% is shown for the sample with AlN of 10 wt.%, indicating a nearly densified behavior.