Microstructure Control of Porous In-situ Synthesized Si_2N_2O-Si_3N_4 Ceramics

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Abstract

Using 6wt%Y_2O_3-2wt%Al_2O_3 as sintering additives and Si as a raw powder, the continuously porous in-situ Si_2N_2O-Si_3N_4 bodies were fabricated by multi-pass extrusion process and their microstructures were investigated depending on the addition of carbon (0-9wt %) in the mixture powder. The introduction of Si_2N_2O fibers observed in the unidirectional continuous pores as well as in the pore-frame regions of the nitrided bodies can be an effective method in increasing the filtration efficiency. In the case of no carbon addition, the network type Si_2N_2O fibers with high aspect ratio appeared in the continuous pores with diameters of 150-200 nm. However, in the case of 9wt% C addition, the fibers were found without any network type and had diameters of 200-250 nm.

Keywords : extrusion, porous materials, Si_2N_2O ceramic, fiber

1. Introduction

Reaction-bonded Si_3N_4 (RBSN) is one of the most attractive ceramics because of some of its advantages such as the low price of raw powders, the easy control of dimensions, low cost of production as well as their outstanding characteristics such as good thermal and chemical stability at high temperatures [1]. On the other hand, Si_2N_2O ceramic has also been found attractive, compared to Si_3N_4 due to its high oxidation resistance, and its chemical resistance at high temperatures [2]. However, the in-situ synthesized Si_2N_2O-Si_3N_4 ceramics showed combined properties such as oxidation resistance as well as strength.

Recently, porous ceramics have been used widely as environmental filters for the purification of water and air [3]. Ceramic Diesel Particulate Filters (DPF) used for the purification of air in automobiles need to have an excellent thermal shock resistance, thermal and chemical stability and coatability of catalyst. However, the environmental filters should have a high surface area as well as high porosity with a desired mechanical strength to increase the filtration efficiency in their application area. Moreover, the introduction of whiskers or fibers in the porous ceramics can be an effective effort to increase the filtration efficiency, as well as the mechanical strength through fiber reinforcement.

In this works, continuously porous Si_2N_2O-Si_3N_4 ceramics were fabricated using the multi-pass extrusion process. Efforts were made to modify the microstructures of the Si_2N_2O-Si_3N_4 ceramics depending on carbon addition for the application of environmental filters. The Si_2N_2O fibers which were frequently observed in the nitrided bodies were investigated depending on fabrication conditions, and their beneficial effect as a filter was also discussed.

2. Experimental and Results

As starting materials, the Si powder (Permascand, Sweden, d50=7μm, BET=1.2 m^2/g), sintering additives (6wt%Y_2O_3-2wt%Al_2O_3, Y_2O_3, Daejung Chemicals & Metals Co., Korea; Al_2O_3, AKP-50, Sumitomo Chemical Co., Japan), different weight percentages (0-9wt%) of a pore-forming agent carbon (10-15μm, Aldrich Chemical Co., USA) were ball milled in ethanol and using Si_3N_4 as a milling media. The dried mixture powder with a polymer binder such as ethylene vinyl acetate (EVA) (ELVAX 210 and ELVAX 250, Dupont, USA) and stearic acid ($CH_3(CH_2)_{16}COOH$, Daejung Chemicals & Metals Co., Korea) with a volume ratio of 55/38/7 was shear mixed at 120ºC for 30min using a shear mixer (Shina Platec. Co., Korea). The homogeneous mixture was used to produce a tube by warm press. On the other hand, the pore-forming agent carbon, polymer and stearic acid with a volume ratio of 48/45/7, were also shear mixed and the mixture was extruded as a rod. The rod was inserted into the tube to prepare the feed roll and was extruded at 90°C to make the 1st passed filaments with a diameter of 3.5mm [4]. The 2nd passed filaments were produced by passing the bundle of the 1st passed filaments through extrusion.

Binder burn-out was employed in a tube furnace at 700°C in flowing N_2 gas. After that, a 2nd burn-out was carried out at 1000°C in air to remove the pore-forming agent. To make

the unidirectional continuous porous Si_2N_2O-Si_3N_4 ceramics, the nitridation process was performed at 1400°C in flowing N_2 gas for 20hrs.

XRD (CuKα, D/MAX-250, Rigaku, Japan) technique was employed to identify the phases of the samples.The microstructure, pore size, pore distribution and the morphology of the Si2N_2O fibers of porous nitrided Si_2N_2O-Si_3N_4 bodies were investigated by FE-SEM (JSM-635F, JEOL, Japan) technique.

Fig. 1 shows low magnification images (a, b) of the extruded body. The EDS profiles (c) and (d) were taken from P and Q regions in Fig. 1(a), respectively. The core part (Q: dark contrast) was comprised of carbon/polymer and the frame part (P: comparatively bright contrast) showed a mixture of powder/polymer which appeared with an alternate layer. The diameter of the pore-forming agent was about 260μm. After the burn-out process, the polymer and pore-forming agent were almostly removed and then a continuously porous sample was obtained. The porous green body was nitrided at 1400ºC in a N_2 atmosphere to produce a continuously porous Si_2N_2O-Si_3N_4 body.

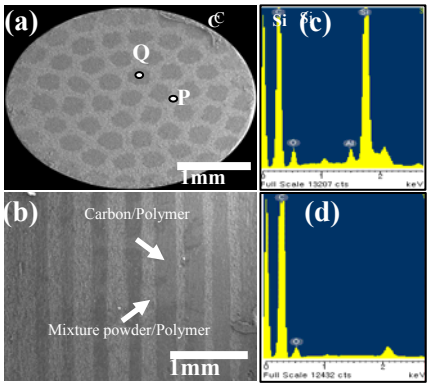

Fig. 1. SEM transverse (a), longitudinal (b) images and EDS profiles (c, d) of the extruded bodies.

Fig. 2 shows the SEM fracture surfaces of continuously porous 2nd passed nitrided Si_2N_2O-Si_3N_4 bodies. The diameter of the continuous pores was about 240μm. This value was slightly decreased compared with the pore-forming agent in the original extruded body as shown in Fig. 1(a), due to the large amount of fibers in the continuous pore surface regions. In the case of no carbon addition in the mixture powder, the continuous pores of the nitrided bodies were fully covered with network type fibers, with high aspect ratio, with diameters of 150-200nm as shown in the enlarged image that was taken from the continuous pore region in Fig. 2(a). The formation of the fibers may be due to the vapor phase reaction among SiO and N_2 during the nitidation process [5]. The droplets on the fiber surfaces may be formed because of the Al component in the liquidus phase. However, as shown in the enlarged image of Fig. 2(b), in the case of 9wt% carbon addition sample, the number of fibers was decreased without the formation of a network compared with the former case. A few amounts of straight fibers with an increasing diameter were also observed in the continuous pores.

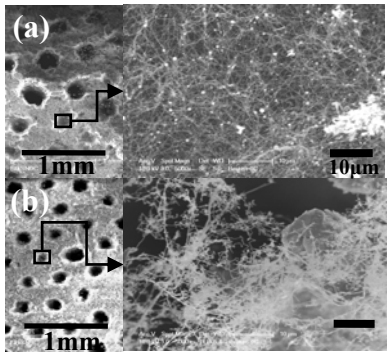

Fig. 2. SEM fracture surfaces of porous nitrided Si_2N_2O-Si_3N_4 bodies; (a) 0wt% C and (d) 9wt% C addition.

3. Summary

Microstructures of the porous in-situ synthesized Si_2N_2O-Si_3N_4 ceramics, fabricated by the multi-pass extrusion process, were investigated depending on the addition of carbon in the mixture powder. The Si_2N_2O nanofibers observed in the continuous pores, as well as in the pore-frame regions, can provide a beneficial effect on filtration efficiency when used as filter materials. The diameter of the fibers with 0wt% and 9wt% carbon addition was about 150-200nm and 200-250nm, respectively.

4. Acknoledgement

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5. References

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