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Short communication

Microstructure and mechanical properties of the α-SiAlON/α-SiC composites: Effects of heat treatment

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Abstract

Self-toughening α-SiAlON/6H-SiC composites containing both α-SiAlON and α-SiC elongated grains in the microstructures were previously fabricated by spark plasma sintering (SPS). In this study, the α-SiAlON/6H-SiC composites were heat treated at 1850 °C for 3 h. The effects of the heat treatment on the phase compositions, microstructures, and mechanical properties were investigated. The results showed M′ phases formed from crystallization of the intergranular glassy films. No polytype transformation for the SiC phase was observed. Both α-SiAlON and α-SiC grains grow anisotropic in the heat treatment, rather than converted into the brittle equiaxed morphologies. The flexural strength and fracture toughness of the post heat-treated composites were further increased and the 40 wt% SiC composition reached 907 MPa and 8.5 MPa $m^{1/2}$, respectively.

Keywords: B. Microstructure; C. Mechanical properties; Ceramic composite; Heat treatment

1. Introduction

α-SiAlON and silicon carbide are advanced structural ceramics [1–3] that have a potential for applications at high temperatures up to 1300 °C. Both monolithic ceramics can form a self-toughening microstructure to yield high flexural strength and fracture toughness. In a previous study, α-SiAlON/α-SiC composites with 0–40 wt% 6H-SiC were synthesized by SPS. Similar with the composites fabricated by HP, PLS and GPS [4–6], adding SiC particles to the α-SiAlON ceramics remarkably reduced the sizes of the α-SiAlON grains. Formation of the bimodal microstructures was prevented [4–9]. However, both α-SiAlON and 6H-SiC grains grew anisotropic in the SPS composites, in contrast to the equiaxed grain morphologies in the HP, PLS and GPS materials, hence higher flexural strength and fracture toughness were obtained in the SPS materials, due to the self-reinforcement of the elongated grains.

The anisotropic growth of the 6H-SiC and α-SiAlON grains was partly attributed to the extra liquid introduced by the 4 wt% Y_2O_3 additive [7,10]. The fast heating rate of SPS [11] (100 °C/min) was also suspected of playing a role in forming the self-toughening microstructures, because a higher concentration of transient liquid should be preserved during heating up in SPS than in the conventional sinterings. Therefore, if the formation of the self-toughening microstructures was determined by the different sintering techniques, the survivability of the self-toughening microstructures in a prolonged heat treatment is under suspicion, i.e., an isothermal annealing may convert the self-toughening microstructures, into the brittle equiaxed ones.

In this study, the effects of the annealing at 1850 °C for 3 h on the phase compositions, microstructures and mechanical properties of the SPS-sintered α-SiAlON/α-SiC composites were investigated.

2. Experimental

The investigated materials were the α-SiAlON/α-SiC composites with 0 or 40 wt% 6H-SiC (see Table 1). The fabrication processes, microstructures and mechanical properties of the materials were detailed in an accompany paper [7]. For brief reference, the designed composition of the α-SiAlON phase was $Y_{1/3}Si_{10}Al_2ON_{15}$, i.e., m, $n = 1.0$ in the general formula $RE_{1/3}Si_{12-(m+n)}Al_{m+n}OnN_{16-n}$ for α-SiAlON. 4 wt% Y_2O_3 was used as the sintering additive. Spark plasma sintering was performed at 1800 °C for 5 min under 20 MPa pressure in a

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Table 1
Composition, density and mechanical properties of the compositions.

Material	Starting composition (wt%)			Density (g/cm^3)	Vickers hardness (GPa)	Flexural strength (MPa)	Fracture toughness (MPa m$^{1/2}$)
	$Y_{1/3}Si_{10}Al_2ON_{15}$	Y_2O_3	α-SiC				
0 wt% SiC	96.0	4.0	0.0	3.321	19.8 ± 1.1	742 ± 80	7.4 ± 0.6
40 wt% SiC	96.0	4.0	40.0	3.276	22.2 ± 1.2	907 ± 57	8.5 ± 0.9

0.1 MPa nitrogen atmosphere, produced cylindrical samples with dimensions of 20 mm in diameter and 6 mm in height.

The materials were subsequently heat treated in a gas pressing equipment at 1850 °C for 180 min in a 0.9 MPa nitrogen atmosphere. 50 °C/min were applied for both heating and cooling. After heat treatment, the samples were ground to 4 mm in thickness and then sliced, ground, and polished to 0.5 μm diamond finish.

Densities were measured by the Archimedes' principle. Phase compositions were investigated by X-ray diffraction (XRD) using Cu K_α irradiation. Vickers hardness was tested using 49 N load and dwell time of 15 s. Flexure strength and fracture toughness were measured by three-point bending and single-edge-notched beam method, respectively. The dimensions of the three-point bending samples were 4.0 mm × 1.5 mm × 18.0 mm, with a 15.0 mm span and 1.5 mm height. The fracture direction was normal to the SPS pressing direction. The dimensions of the single-edge-notched beams were 4.0 mm × 1.5 mm × 18.0 mm, using a 16.0 mm span and 4.0 mm height. A 2.0 mm-deep notch was cut by a 0.2 mm thick diamond wafer. The fracture direction was parallel to the SPS pressing direction. Three specimens were tested for each condition. Microstructures and fracture surfaces were observed by scanning electron microscopy (SEM) with energy dispersive X-ray spectroscopy (EDXS).

3. Results

The densities of the composites after heat treatment are shown in Table 1. A very slight weight loss up to 0.3% was noticed, presumably due to evaporation of some ingredients of the intergranular phases.

X-ray diffraction patterns of the heat-treated materials are shown in Fig. 1. α-SiAlON and trace M′ ($Y_2Si_{3-x}Al_xO_{3+x}N_{4-x}$) were detected in the 0 wt% SiC composition and α-SiC was also evident in the 40 wt% SiC material. The M′ phase should be formed by devitrification of the grain boundary phases, similar with the previous report [10].

The α-SiC phase was the same 6H SiC in the as-sintered materials. The identical X-ray diffraction peak positions indicated neither SiC solid solutions nor polytype transformation [12]. The α-SiAlON lattices were slightly enlarged to give a higher calculated composition of $m = 1.17$ than the $m = 1.09$ in the as-sintered materials, suggesting more Y^{3+} incorporation into the α-SiAlON lattice during the heat treatment [10,13]. The SiC contents in the composites had no significant effects on the α-SiAlON compositions [5,9], as the α-SiAlON phase in both materials had similar m values.

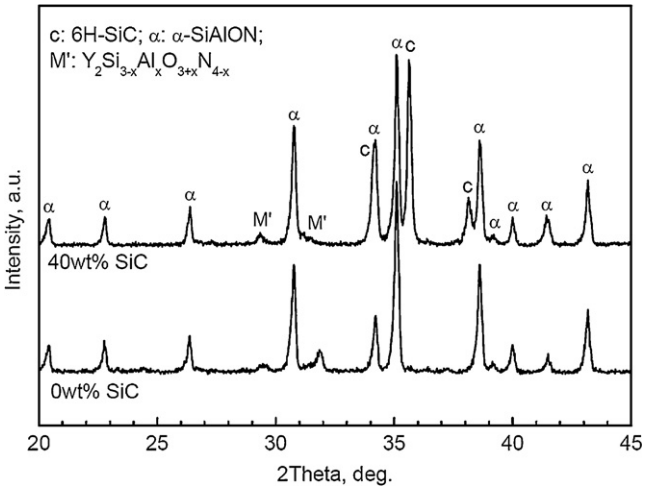

Fig. 1. XRD patterns of the post heat-treated α-SiAlON/α-SiC composites.

The SEM micrographs of the microstructures are shown in Fig. 2. No porosity was observed, indicating full densities of the α-SiAlON/α-SiC composites, despite the slight weight losses. The 6H-SiC, α-SiAlON and grain boundary phases, having dark, gray and bright contrast, respectively, were homogeneously dispersed. No large segregations of any phases were observed. The M′ phase should locate at the multi-grain conjunctions due to devitrification of the grain boundary liquid [10]. Considering its thermodynamic persistence, thin glassy films along the grain boundaries could be expected [14]. The glassy films shared the same bright contrast with the M′ phase.

The typical bimodal microstructure of the 0 wt% SiC material, which composed of a few large grains having an average diameter of ∼3.5 μm and aspect ratio of ∼10, dispersed homogeneously in the fine grained matrix with an average diameter of ∼1.2 μm, was in consistency with the previous observations [7,10,15–17], and thus was of trivial scientific value, because it has been well established that excess Y_2O_3 facilitated anisotropy of the α-SiAlON grains, and heat treatment at high temperatures enlarged the grain growth [10,15].

In the 40 wt% SiC material, the large bimodal α-SiAlON grains were not formed, yielding a normal fine microstructure [4–9]. The self-toughening microstructures of the as-sintered composites survived the 1800 °C 3 h annealing. The α-SiAlON and 6H-SiC grains had an average grain diameter of ∼0.8 μm and 1.1 μm, respectively, both kept the high aspect ratio up to 8.0 as that of the as-SPS-sintered materials.

The goodness of maintaining the self-toughening microstructures was evident by the mechanical properties listed in

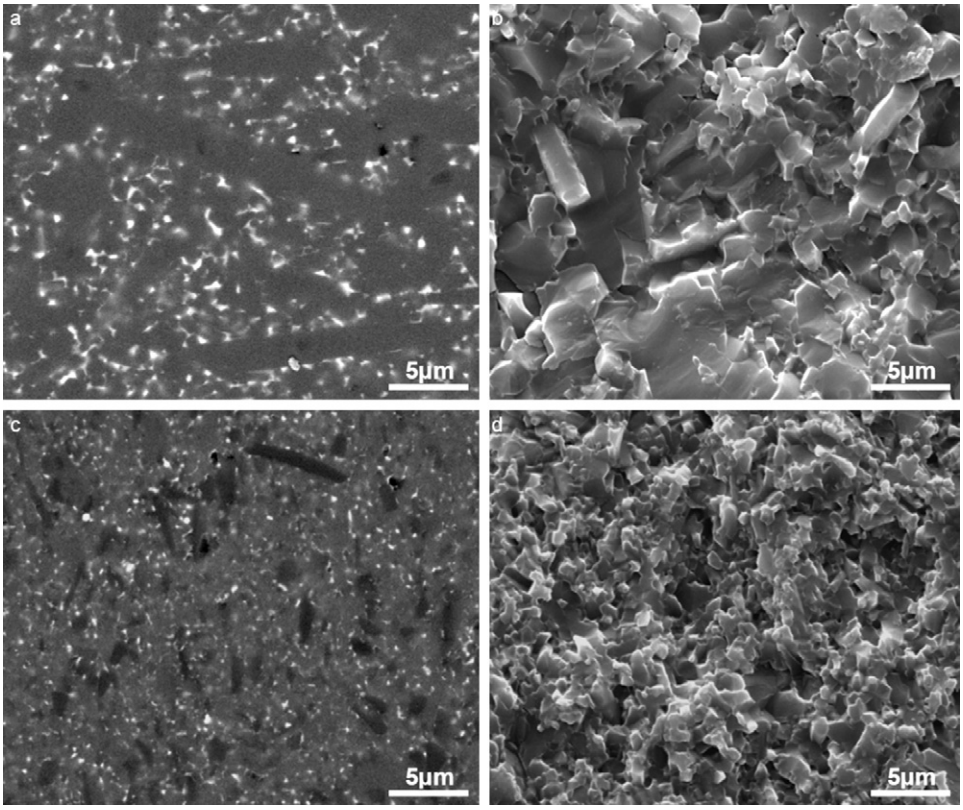

Fig. 2. SEM images of the microstructures and fractures of the post heat-treated α-SiAlON/α-SiC composites. (a and b) 0 wt% SiC, and (c and d) 40 wt% SiC composition.

Table 1. Flexural strength and fracture toughness were further increased after heat treatment, compared to the as-sintered counterparts [7,10,15–17]. A remarkable combination of flexural strength and fracture toughness of 907 MPa and 8.5 MPa $m^{1/2}$ were obtained in the 40 wt% SiC composite. The 40 wt% SiC material also had higher hardness because the hardness of α-SiC is generally 10% higher than α-SiAlON.

4. Discussion

4.1. Microstructure development

In the early studies of the α-SiAlON/SiC composites, Santos et al. [4,5] and Wang et al. [6] prepared α-SiAlON/SiC with 0–20 wt% and 0–40 wt% β-SiC by gas pressing at 1950 °C and hot pressing at 1400–1800 °C, respectively. β-SiAlON/SiC composites with 70–80 vol% α-SiC were also fabricated by HIP at 1850 °C under 200 MPa in an argon atmosphere [18] Very fine homogeneous microstructures were obtained in those materials [4–6,18]. But no elongation of either SiC or SiAlON (α or β) grains were observed.

In comparison, we synthesized α-SiAlON/α-SiC composites with 0–40 wt% 6H-SiC by SPS. The α-SiAlON and α-SiC grains were both elongated [7]. Although the few bimodal α-SiAlON grains were not presented, similar with references [4–7,18], self-toughening microstructures were formed. Microstructure observations in Fig. 2 indicated that the heat treatment grew the α-SiAlON and α-SiC grains by 37% in size in the 40 wt% SiC composite, however, the large aspect ratio of 10 and 8.0 was not changed (Fig. 2c).

Therefore it seemed clear that the elongated growth of the α-SiAlON grains in the α-SiAlON/α-SiC composites was not necessarily affected by the coexistent α-SiC particles. Once formed, the self-toughening microstructures could survive high-temperature annealing and would not convert to the brittle equiaxed microstructures.

As reported in the previous sections, a factor benefitting the elongated growth of the α-SiAlON grains [4–6] was the chosen SiAlON composition. The investigated α-SiAlON with Y-dopants, m, $n = 1.0$ composition, and 4 wt% Y_2O_3 as the sintering additive, yielded adequate transient liquids to promote elongation of the α-SiAlON grains [10,19]. The few percent SiO_2 on the SiC particle surfaces which was not compensated when calculating the starting compositions, should have increased the liquid concentration. In comparison Wang et al. [6] chose the Ca-α-SiAlON with $m = 2$, $n = 1.0$ composition and obtained an equiaxed microstructure [17] formed by some large grains homogenously dispersed among the finer grained matrix.

In Souza et al. [4,5], although the hot pressed monolithic α-SiAlON ceramics produced the self-toughening microstructures, the elongation of the α-SiAlON grains was gradually diminished with the SiC concentration increase from 5 to 20 wt%. Wang at el and Souza et al. used β-SiC (cubic or 3C-SiC) rather than α-SiC (6H-SiC) [4–7]. Thus, whether the different SiC polytypes (6H vs. 3C SiC) had played a role in the

elongation growth of the coexistent α-SiAlON grains is under debate, but is not clear.

β-SiC could not develop elongated morphology due to its cubic crystal symmetry. In SiC ceramic production practice, the β-cubic to α-hexagonal SiC phase transformation is usually exploited to introduce elongated (4H polymorph) or plate-like grains (usually 6H) to the microstructures [20–23]. In the references on the SiAlON/SiC materials, such a phase transformation was not presented, explaining the equiaxed SiC grains in the microstructures.

However, when 6H-SiC and SPS were applied instead of 3C-SiC and conventional HP, HIP, and GPS [4] to synthesize the α-SiAlON/α-SiC materials, elongation of the SiC grains was realized without the polytype transformation. During heat treatment, the further anisotropic growth of the 6H-SiC grains was presented as shown in Fig. 2c, by Ostwald ripening. Smaller grains should have dissolved in the surrounding liquid and further grew the selected grains to lower surface energy by anisotropic growth.

4.2. Mechanical properties

The benefits of the heat treatment were demonstrated by the mechanical properties as listed in Table 1. The flexural strength and fracture toughness of the 0 wt% SiC composition reached that for a self-reinforced Ca-α-SiAlON, produced by seeding the starting powders [17]. Higher flexural strength and fracture toughness up to 907 MPa and 8.5 MPa $m^{1/2}$ were obtained in the 40 wt% SiC composition, compared to 812 MPa and 8.2 MPa $m^{1/2}$ for the as-sintered counterpart.

The mechanical property improvements were partly due to the enhanced growth of the elongated grains. Larger elongated grains were more sufficient than smaller ones in deflecting and bridging cracks, promising higher fracture toughness. Flexural strength increment was also expectable from the higher load transfer and bearing abilities by the moderately enlarged grains [24], but extremely large grains may decrease strength for possible crack initiations.

Absence of the bimodal α-SiAlON grains in the 40 wt% SiC composition avoided early crack initiation, hence high flexure strength, with a lower data deviation (Table 1).

Table 1 shows the fracture toughness of the 0 wt% SiC material increased from 5.6 MPa $m^{1/2}$ for the as-sintered material to 7.4 MPa $m^{1/2}$ after heat treatment [7]. The fracture toughness improvement should also be correlated with an interfacial bond weakening effect, as evidenced by Fig. 2b where a much coarser fracture than the as sintered material [7] was shown. During fracturing, the large α-SiAlON grains were intersected by the advancing cracks in the as-sintered 0 wt% SiC material, due to lack of the interfacial debonding, resulting in a rather plane fracture surface [7,10]. However, local residual stresses from the thermal expansion coefficient differences between the 6H-SiC and the α-SiAlON favored interfacial debonding to impart a better toughening to the 40 wt% SiC composition.

The formation of the M′ phase should also contribute to the interfacial bond weakening effects. Crystallization of the M′ phase should have altered the Y:Al and O:N ratios in the intergranular glassy films during the annealing. And increases in the Y:Al and O:N ratios generally promoted interfacial debonding, similar with the β-Si_3N_4 ceramics [25].

5. Conclusions

α-SiAlON and α-SiC grains in the α-SiAlON/6H-SiC composites were grown anisotropically by heat treatment at 1850 °C, further optimized the self-toughening microstructures of the composites. The α-SiAlON/6H-SiC with 40 wt% SiC had a fine self-toughening microstructure to exhibit high flexure strength and fracture toughness up to 907 MPa and 8.5 MPa $m^{1/2}$, respectively. Crystallization of M′ from the intergranular glassy phases in combination with the local residual thermal stresses induced by the thermal expansion coefficient difference between the α-SiAlON and the 6H-SiC grains, weakened interfacial bonding and thus enhanced the toughening mechanisms.

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