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Influence of microstructure on strength and fracture toughness of β-Sialon

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β-Sialon ($Si_{6-z}Al_zO_zN_{8-z}$) with $z = 0.5$ was fabricated by hot-pressing of a spray dried mixture of α-Si_3N_4 and aluminium-isopropoxide solution. Phase composition, flexural strength and microstructure of a sintered body were investigated. Phases identified by XRD were β-Sialon and a small amount of O′-Sialon. The flexural strength (three-point bending) was about 1500 MN m^{-2}. This value, about three times higher than that of β-Sialon fabricated from α-Si_3N_4 and α-Al_2O_3 powder, was mainly due to the homogeneous microstructure without large defects such like clusters of large grains. β-Sialon was heat treated at 2000° C for 2 h in 4 MPa N_2 to develop elongated β-Sialon grains with high aspect ratio. Microstructure, flexural strength and fracture toughness (K_{Ic}) of it were investigated. Both strength and K_{Ic} were lower than those of hot-pressed sample, even though an elongated microstructure was achieved. This fact showed that the toughening of β-Sialon with elongated grains could not be achieved without grain boundary phase which resulted in a crack deflection.

1. Introduction

β-Sialon ($Si_{6-z}Al_zO_zN_{8-z}$) is of interest for applications in ceramic components for heat engine and other high temperature engineering [1], because of their excellent properties such like high temperature strength, high oxidation and corrosion resistance.

Commonly, β-Sialon is fabricated from the powders mixture of Si_3N_4, Al_2O_3 and AlN [2, 3] or Si_3N_4, SiO_2 and AlN [4, 5]. However the strength of these β-Sialon [6–8] is still low compared with Si_3N_4 based ceramics such as Si_3N_4 with Al_2O_3 and Y_2O_3 [9] or MgO [10]. The strength of β-Sialon is largely influenced by the existence of unhomogeneous parts such as clusters of large grains or porous parts because these unhomogeneous parts become a fracture origin [6, 7]. The formation of these unhomogeneous parts assumed to originate in the unhomogeneous mixing and dispersing of starting powders. Thus in the parts where much liquid phase formed during sintering and grain growth resulted and in the opposite sense porous parts were formed. It is considered that the strength of β-Sialon must be improved if these unhomogeneous parts as a fracture origin can be removed.

For this purpose, a aluminium alkoxide used as a solution was an effective starting material for β-Sialon to get homogeneous microstructure because of its uniformity and reaction as the surface modified of Si_3N_4 powder [11–13], and the strength of β-Sialon fabricated using aluminium alkoxide instead of Al_2O_3 powder must be greatly improved.

It was believed that the high strength and the high fracture toughness of Si_3N_4 with Y_2O_3 and Al_2O_3 originated in their elongated β-Si_3N_4 grains with high aspect ratio (length/width) [14, 15], and the low fracture toughness of β-Sialon originated in the equiaxed β-Sialon grains from the standpoint of crack deflection. The grain morphology of β-Sialon can be controlled from equiaxed to elongated one by heating it at high temperature under applied N_2 gas atmosphere [16].

To obtain a β-Sialon with a high strength and high fracture toughness, β-Sialon with $z = 0.5$ was fabricated by hot-pressing a spray dried mixture of α-Si_3N_4 and aluminium-isopropoxide solution, and heat treating at at 2000° C for 2 h under 4 MPa N_2 gas atmosphere to develop elongated β-Sialon grains. Some mechanical properties were measured in comparison with those of β-Sialon fabricated from a powder mixture of α-Si_3N_4 and α-Al_2O_3. This paper describes the strength and the fracture toughness of β-Sialon and discusses the influence of microstructure on them.

2. Experimental techniques

α-Si_3N_4 powder was purchased from Toyo Soda Co. Ltd (TS-7) and aluminium-isopropoxide (Al(Oi-Pr)$_3$, extra pure reagent) from Nakarai Chemicals Co. Ltd. Al(Oi-Pr)$_3$ was dissolved in dry n-C_6H_{14} and the solution was filtered to remove the aluminium hydroxide on the surface of Al(Oi-Pr)$_3$. The precipitant was fired and the amount of Al(Oi-Pr)$_3$ in the filtrate was determined. α-Si_3N_4 powder was added to the filtrate in the ratio of $Al_2O_3 : Si_3N_4 = 9.01 : 90.99$, where the Al/Si ratio was the same as $z = 0.5$ in the formula of $Si_{6-z}Al_zO_zN_{8-z}$. The mixture of Si_3N_4 and Al(Oi-Pr)$_3$/C_6H_{14} was ball milled for 20 h with Si_3N_4 balls and spray dried. The spray dried powder was calcined at 300° C for 2 h in air. The calcined powder was hot-pressed at 1850° C for 1 h under 29 MPa in 1 atm N_2 using a 30 mm × 30 mm carbon mould. A hot-pressed sample was prepared from the powders mixture of

TABLE I Densities and phases of hot-pressed samples

Starting material	Bulk density ($g\,cm^{-3}$)	Phases
Al(Oi-Pr)$_3$	3.14	β + O′(w)
Al_2O_3	3.14	β + α(tr)

β: β-Sialon.
α: α-Si_3N_4.
O′: Si_2N_2O(s.s.).

α-Al_2O_3 (Iwatani Chemicals Co. Ltd RA-40) and α-Si_3N_4 for comparison.

A hot-pressed sample using Al(Oi-Pr)$_3$ was heat treated at 2000° C in 4 MPa N_2 for 2 h.

The density of as hot-pressed and heat treated samples was determined by water immersion method. The phase composition was determined by powder X-ray diffraction analysis (XRD). The polished and fractured surfaces were observed with optical and scanning electron microscopes (SEM).

The specimens (3 mm × 3 mm × 30 mm) for flexural strength measurement were prepared by grinding with No. 600 diamond wheel and chamfering with No. 800 SiC abrasive paper, and then they were fired at 1200° C for 2 h in air to reduce the influence of surface machining flaws. Flexural strength (σ_f) measurement was performed at room temperature using a three-point loading device with a span of 20 mm and a crosshead speed of 0.5 mm min^{-1}. The fracture toughness (K_{Ic}) was measured by Vickers indentation method with load of 10 kg by using the equation given by Niihara [17].

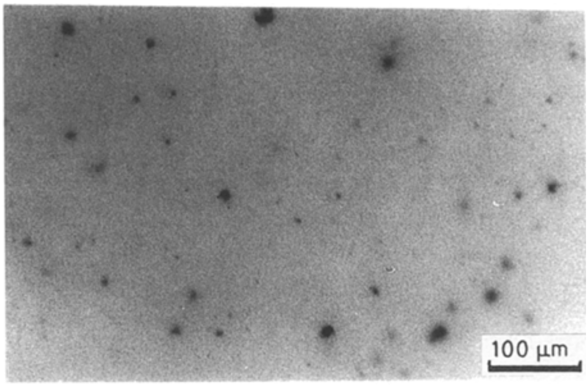


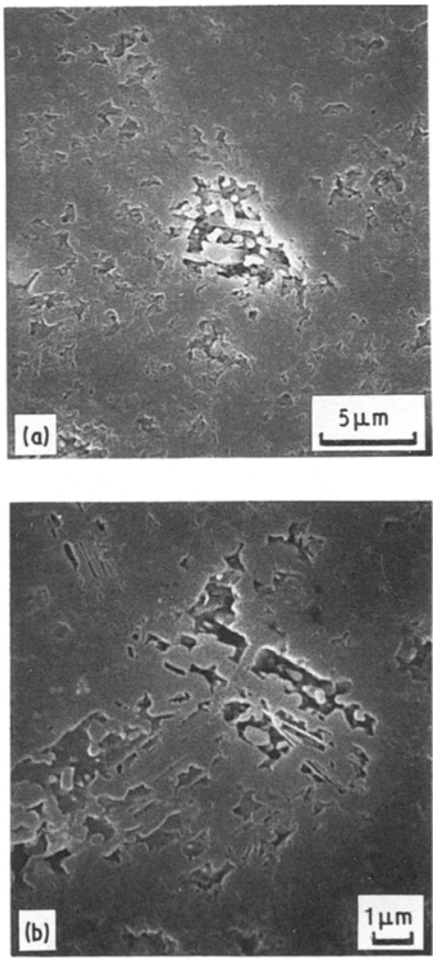
Figure 1 Optical micrograph of sample using α-Al_2O_3.

3. Results and discussion

3.1. As hot-pressed sample

Table I showed the bulk density and phase composition of hot-pressed samples fabricated from Si_3N_4–Al(Oi-Pr)$_3$ and Si_3N_4–Al_2O_3. The density of both samples was 3.14 $g\,cm^{-3}$ (> 99% theoretical). Sample using Al(Oi-Pr)$_3$ consisted of β-Sialon and a small amount of O′-Sialon. This phase composition corresponded to the phase diagram of Si–Al–O–N system given by Jack [1] or Gauckler *et al.* [18]. On the other hand, α-Si_3N_4 remained and no O′-Sialon was observed in sample using α-Al_2O_3. This could be explained by the unhomogeneous dispersion of aluminium and oxygen, in the sample using α-Al_2O_3 powder. That is Al_2O_3-rich parts were formed where more glassy phase was formed and Al_2O_3 lacking parts were α-Si_3N_4 remained.

Fig. 1 showed an optical micrograph of sample using α-Al_2O_3. There could be found a lot of unhomogeneous parts observed as black spots. These black spots included large amounts of glassy phase and were easily chemically etched with HF. No such large black spots could be found in the sample using Al(Oi-Pr)$_3$. Fig. 2 shows SEM photographs of the samples. In the sample using α-Al_2O_3, many pores were formed by chemical etching (Fig. 2a) and clusters of large grains (Fig. 2b) were observed. Sample using Al(Oi-Pr)$_3$ showed a homogeneous microstructure consisting of



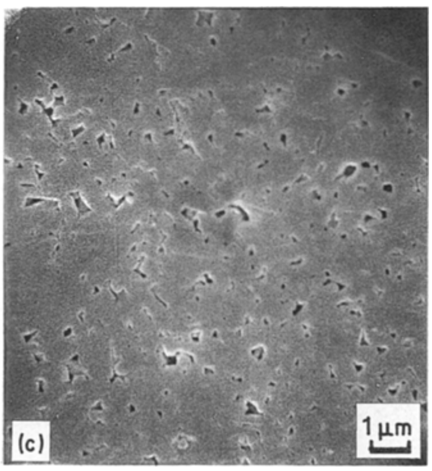


Figure 2 SEM photographs of samples (polished and etched with HF and HNO_3): (a), (b) sample using α-Al_2O_3, (c) sample using Al(Oi-Pr)$_3$.

TABLE II Mechanical properties of hot-pressed samples

Starting material	σ_f ($MN\,m^{-2}$)	Hv ($GN\,m^{-2}$)	K_{Ic} ($MN\,m^{-3/2}$)
$Al(Oi\text{-}Pr)_3$	1480	16.0	3.3
Al_2O_3	484	15.7	3.2

TABLE III Mechanical properties of heat-treated sample

σ_f ($MN\,m^{-2}$)	Hv ($GN\,m^{-2}$)	K_{Ic} ($MN\,m^{-3/2}$)
956	13.9	2.8

0.5–1 μm β-Sialon grains and a small amount of grain boundary phase removed by chemical etching (Fig. 2c). The distribution of the grain boundary phase was more homogeneous compared with the sample using α-Al_2O_3.

These results suggested that the use of $Al(Oi\text{-}Pr)_3$ solution was more desirable to get β-Sialon with homogeneous microstructure.

Table II showed the mechanical properties of hot-pressed samples. Vickers hardness and K_{Ic} of both β-Sialons showed approximately the same value of $16\,GN\,m^{-2}$ and $3.3\,MN\,m^{-3/2}$, respectively. However, strength of the sample using $Al(Oi\text{-}Pr)_3$ was about $1500\,MN\,m^{-2}$, about three times higher than that of the sample using α-Al_2O_3 and conventional β-Sialon [6–8].

Fig. 3 showed the fracture origins of the samples. The fracture origins in the sample using α-Al_2O_3 (Fig. 3a) were large internal defects originated in unhomogeneous parts shown in Figs 1 and 2. The fracture origins of sample using $Al(Oi\text{-}Pr)_3$ were internal defects (Fig. 3b) and machining flaws (Fig. 3c). However, the size of the internal defects were much smaller than those in the sample using α-Al_2O_3. Fig. 4 showed SEM photographs of a typical internal defect as a fracture origin in the sample formed from $Al(Oi\text{-}Pr)_3$. These defects consisted of a slightly larger size of grains compared with the matrix. And the shape of defects was flat and lying perpendicular to hot-pressing direction. These internal defects resulted from the Al_2O_3 concentrated parts and the size of these Al_2O_3 concentrated parts (about $10\,\mu$m) was assumed to have been limited by the size of the particles formed by spray drying.

Thus, the higher strength of the sample using $Al(Oi\text{-}Pr)_3$ was mainly due to the homogeneous microstructure without large defects. And it was expected that if the influence of surface machining flaws could be removed completely, the strength must be higher as β-Sialon was very sensitive to microcracks for its low K_{Ic}, and the lowest strength of it would be controlled by the maximum size of spray dried particles.

Fig. 5 shows a fractured specimen after the bending test. The cracks formed symmetrical fragments from a fracture origin and the shape of these fragments were almost the same as the distribution of stress before fracture [19]. This result suggests that only one defect where a crack began to move became a fracture origin and propagation of this crack released the stress of loading instantaneously.

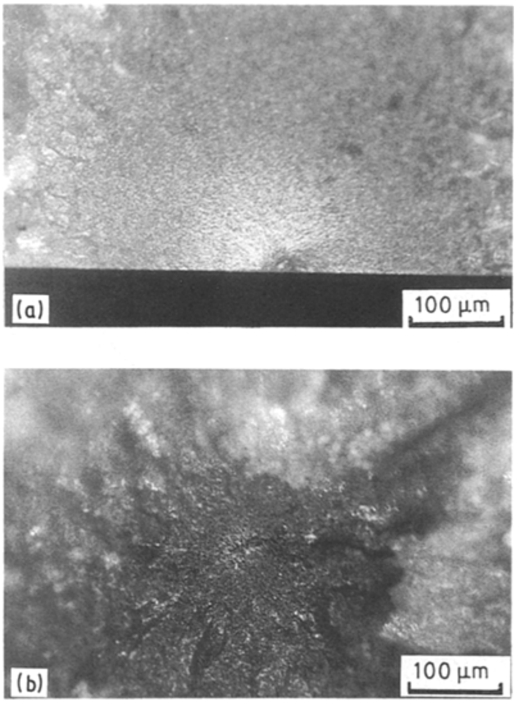
3.2. Heat-treated sample

The suppressed thermal decomposition of β-Sialon and the weight loss and the change of density after heat treatment at 2000° C for 2 h in 4 MPa N_2 atmosphere were not observed. The crystalline phase identified by XRD in the heat treated sample was β-Sialon.

Table III shows the mechanical properties of the heat treated sample. Both hardness and K_{Ic} decreased to the value of $13.9\,GN\,m^{-2}$ and $2.8\,MN\,m^{-3/2}$, respectively. Room temperature strength also decreased to about $950\,MN\,m^{-2}$.

The microstructure of heat treated sample (Fig. 6) consisted of grown and elongated β-Sialon grains, and a small amount of grain boundary phase. This microstructure was almost the same as that of the Si_3N_4 sintered body with high fracture toughness [20].

In general, it was believed that the lower fracture toughness of pure β-Sialon was due to its microstructure with equiaxed grains where crack deflection was difficult. However, according to this study, this is not correct at least in the case of pure β-Sialon. In the case of MgO-doped [21] or Y_2O_3-doped β-Sialon [22], the



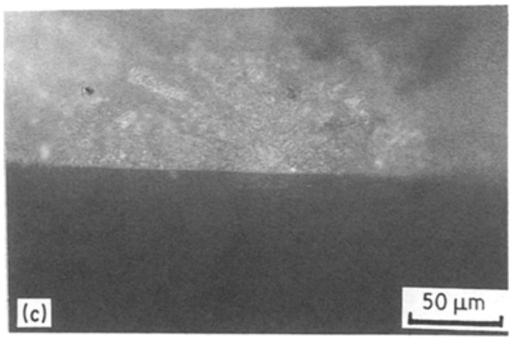


Figure 3 Optical micrographs of fracture origins: (a) defect in sample using α-Al_2O_3 ($\sigma_f = 447\,MN\,m^{-2}$), (b) defect in sample using $Al(Oi\text{-}Pr)_3$, ($\sigma_f = 1379\,MN\,m^{-2}$), (c) surface machining flaw in sample using $Al(Oi\text{-}Pr)_3$ ($\sigma_f = 1555\,MN\,m^{-2}$).

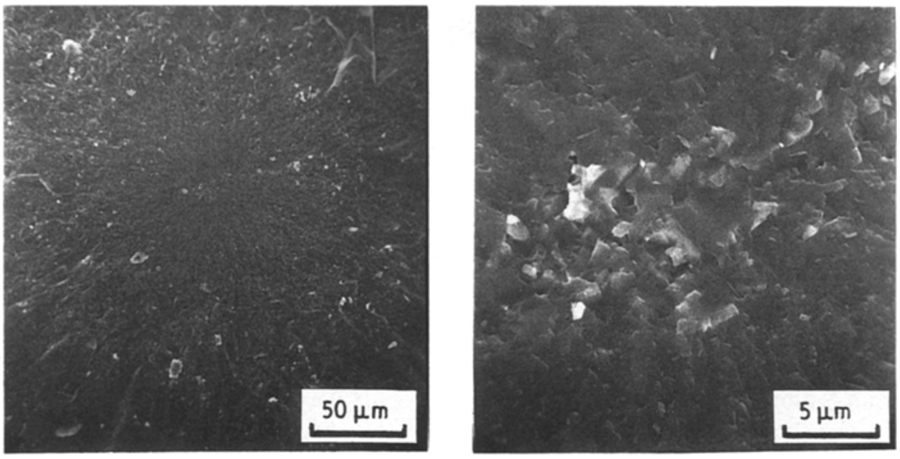


Figure 4 SEM photographs of a defect as a fracture origin (same specimen as Fig. 3b).

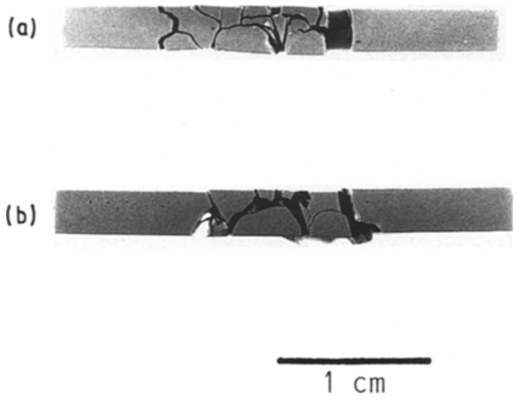


Figure 5 Fractured specimen of sample using Al(Oi-Pr)$_3$ after bending test. (a) $\sigma_f = 1402\,\mathrm{MN\,m^{-2}}$; (b) $\sigma_f = 1399\,\mathrm{MN\,m^{-2}}$.

concentration of Mg and O or Y and O in the grain boundary phase and the formation of a low melting glassy phase promoted the development of elongated grains. This grain boundary phase was weak compared with β-Sialon grains. Therefore, crack deflection took place along the elongated β-Sialon grains and resulted in the improvement of fracture toughness. In the case of pure β-Sialon, it was considered that according to the presence of a small amount of grain boundary phase and a small difference in the physical properties between the grain boundary phase and β-Sialon grains, no crack deflection took place along the elongated grains. In addition to the increase in defect size by grain growth, the presence of elongated β-Sialon grains might be one reason for the decrease in the strength and K_{Ic} of β-Sialon.

The fracture surface of heat treated sample was mostly occupied by transgranular fracture except for the long grain boundary lying parallel to a crack propagation (Fig. 7). This is assumed to indicate that the existence of this kind of long grain boundary makes the crack propagation easy and the crack deflection that was observed in Si_3N_4 [23] does not occur. In the case of this sample, this was the reason for the degradation of the strength and K_{Ic}.

From these results, it can be concluded that the toughening of β-Sialon with elongated microstructure could not be achieved without the existence of a weak grain boundary phase where a crack could easily propagate. If there was no weak grain boundary phase for crack deflection, the existence of elongated grains results in a decrease in both toughness and strength.

4. Conclusion

β-Sialon with $z = 0.5$ was fabricated by hot-pressing of spray dried mixture of α-Si_3N_4 and Al(Oi-Pr)$_3$ solution. Phase composition, flexural strength and microstructure of a sintered body were investigated.

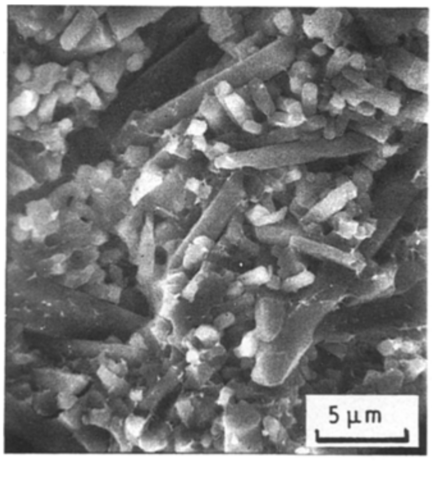


Figure 6 Microstructure of heat-treated sample.

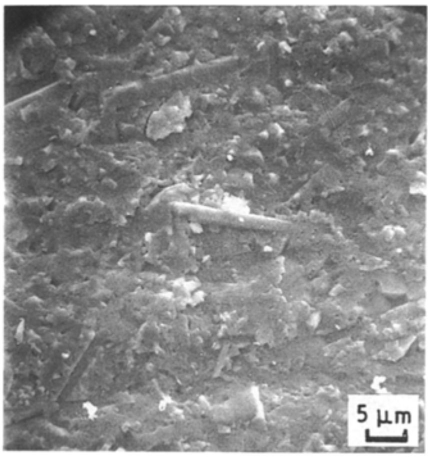


Figure 7 Fracture surface of heat-treated sample.

Phases identified by XRD were β-Sialon and a small amount of O′-Sialon, but no α-Si_3N_4 that remained in the hot-pressed sample from the powder mixture of α-Si_3N_4 and α-Al_2O_3 was observed. This assumed to show that starting powder using $Al(Oi\text{-}Pr)_3$ was more homogeneous than that using Al_2O_3 powder. The flexural strength (three-point bending) was about 1500 MN m^{-2}. This value was remarkably higher than that of the sample using Al_2O_3 powder mainly due to the homogeneous microstructure without large internal defects such as clusters of large grains which were observed in sample using Al_2O_3 powder.

The hot-pressed β-Sialon using $Al(Oi\text{-}Pr)_3$ was heat treated at 2000° C for 2 h in 4 MPa N_2 to form an elongated microstructure with high aspect ratio β-Sialon grains. Flexural strength, fracture toughness (K_{Ic}) and microstructure were investigated.

Both the strength and K_{Ic} were decreased compared with the as hot-pressed sample even though the elongated microstructure with high aspect ratio β-Sialon grains was achieved. And a crack deflection which was observed in Si_3N_4- and Y_2O_3-doped β-Sialon was not observed. These results are assumed to be due to the existence of elongated β-Sialon grains without a weak grain boundary phase to result crack deflections.

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