Journal of the European Ceramic Society 21 (2001) 1269–1272

www.elsevier.com/locate/jeurceramsoc

Room temperature strength of β-sialon ($z = 0.5$) fabricated using fine grain size alumina powder

Kazushi Kishi *, Seiki Umebayashi, Eiji Tani, Kazuhisa Shobu, Yanping Zhou

Kyushu National Industrial Research Institute, Shuku-machi, Tosu-shi, Saga, 841-0052 Japan

Received 10 May 2000; received in revised form 6 November 2000; accepted 15 November 2000

Abstract

The room temperature strength of β-sialon with $z = 0.5$ fabricated using a mixture of fine grain size Al_2O_3 and Si_3N_4 powders was measured, and the effect of a small, amount of $Al(Oi\text{-}Pr)_3$ addition as a dispersant on its strength was also investigated. This β-sialon showed an average strength of 1.00 GPa. It was much higher than that of β-sia1on from traditionally produced Si_3N_4 and Al_2O_3 powders as a result of decreasing the size of defects responsible for fracture origin. Mixing of Si_3N_4 and Al_2O_3 powders in a $Al(Oi–Pr)_3$ solution brought further increase in strength to 1.25 GPa because of the smaller size and reduced number of defects. However, the strength of β-sialon is still controlled by the defects with the size of less than 10 μm. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Al_2O_3; Defects; Fracture origin; Powder size; Sialons; Strength

---

1. Introduction

β-Sialon ($Si_{6-z}Al_zO_zN_{8-z}$) is of interest for use in ceramic components for engines and other high temperature engineering applications.[1] The authors have reported that the strength of β-sialon fabricated from Si_3N_4 and aluminum iso-propoxide $Al(Oi–Pr)_3$ solution is much improved in comparison with that from Si_3N_4 and Al_2O_3 powders[2–5] due to the decreased size of the defects active as fracture origins. The formation of the defects in the conventional method was assumed to originate from the inhomogeneous dispersion of Al_2O_3. If a sufficiently homogeneous dispersion of Al_2O_3 can be attained by using Al_2O_3 powder, the powder process will be more appropriate from the aspect of cost than that with $Al(Oi–Pr)_3$ as a starting material. The authors have reported that the sedimentation and flocculation pre-treatment of starting Si_3N_4 and Al_2O_3 powders has been effective in decreasing the size of defects which has resulted in increase in the strength of β-sialon.[6] This result means that the use of homogeneously dispersed fine Al_2O_3 powder can bring improved strength of β-sialon. Recently, a fine grain size and high purity Al_2O_3 powder has been commercialized. It is reported that the strength of sintered $A1_2O_3$ using this powder showed very high strength of more than 800 MPa.[7–9] It is considered that the high strength of this Al_2O_3 is due to the starting powder's high purity and uniformity of particle size.[8] The strength of β-sialon is expected to increase by using this Al_2O_3 powder without any pre-treatment. Furthermore, the authors have reported that $Al(Oi–Pr)_3$ adsorbed on the surface of Si_3N_4 powder, accelerated uniform dispersion of the powder into non polar solvents such as n-C_6H_{14}.[10] Since the same effect can occur with Al_2O_3 powder, the uniformity of Si_3N_4 and Al_2O_3 powders mixture can be improved with $Al(Oi–Pr)_3$, resulting in higher strength.

This paper describes the room temperature strength of β-sialon with $z = 0.5$ fabricated using a fine grain size and high purity Al_2O_3 powder and a small amount of $Al(Oi–Pr)_3$ addition as a dispersant.

2. Experimental procedure

The α-Si_3N_4 raw material used was from Tosoh Co. Ltd (Tokyo, Japan, grade TS-7) and the α-Al_2O_3 which was fabricated by pyrolysis of $NH_4AlO(OH)HCO_3$,[11] was from Taimei Chemicals Co. Ltd (Nagano, Japan,

---

* Corresponding author.

E-mail address: kishi@kniri.go.jp (K. Kishi).

PII: S0955-2219(00)00331-9

grade TM-DR). Some properties of the Al_2O_3 powder are shown in Table 1.[12] Si_3N_4 and Al_2O_3 powders with the gravimetric mixing ratio of 90.99:9.01 where the Si/Al ratio is set to $z = 0.5$ in the formula of $Si_{6-z}Al_zO_zN_{8-z}$, were mixed in ethanol using a plastic bottle and Si_3N_4 balls for 20 h. The mixed slurry was spread in a stainless steel tray and dried before hot-pressing. Another mixture in which 10% of Al_2O_3 powder was displaced by $Al(Oi–Pr)_3$ which was dissolved in dry n-C_6H_{14} as a ca. 2% solution, was prepared by the same mixing and drying route. Hot-pressing was carried out at 1800°C for 1 h under a pressure of 29 MPa in N_2 atmosphere using a 30×30 mm carbon die. Hereinafter these hot-pressed samples are indicated as sample-A and sample-B respectively. Six specimens with a size of 3×3×30 mm for bending test were prepared by previously reported machining from each hot-pressed plate.[4] A half of these specimens were heat treated at 1200°C for 2 h in air to decrease the influence of surface grinding flaws. Bending test was carried out using a 3-point loading device with a span of 20 mm and a cross-head speed of 0.5 mm/min at room temperature. Fracture origins were observed with an optical microscope.

3. Results and discussion

The room temperature strength of the samples is shown in Table 2. Typical fracture origins of sample-A and -B are shown in Figs. 1 and 2 respectively. The average strength of as ground specimens of sample-A was 910 MPa and all specimens were fractured from surface flaws such as that in Fig. 1(a). The heat treated specimens showed relatively high average strength of 1.00 GPa. However, all of them were fractured from an internal defect with the size of less than 20 μm such as that in Fig.1(b). In the case of as ground specimens, although the material also included internal defects, one of which could be a fracture origin, the fracture was initiated from a surface flaw with less stress than from an internal defect because of the sharp crack tip of the as ground machining flaw. So, the dulling of the crack tips of the surface flaws by heat treatment caused fracture to be initiated from internal defects with larger stress after heat treatment. β-Sialon prepared using previous Al_2O_3 powder showed the strength of about 500 MPa and its fracture origins were internal defects with the size of 30–60 μm even in the case without any heat treatment,[2] so the increase in strength of sample-A was due to decreasing the size of fracture origins by using this fine particle size Al_2O_3 powder.

The average strength of as ground specimens of sample-B increased to 1.07 GPa. One of them was fractured from a surface flaw as shown in Fig. 2(a) and the rest were from an internal defect as shown in Fig. 2(b).

Table 1
Properties of starting Al_2O_3 powder

Crystaline type	α
BET specific surface area (m^2/g)	14
Average particle size (μm)	0.2
Primary particle size (μm)	0.1

Impurity (ppm)

K	Na	Fe	Ca	Mg	Si	Cu	Mn	Cr
3	8	8	3	2	15	1	< 1	< 1

Table 2
Strength of samples

	Surface finishing	Strength (MPa)	Fracture origin
Sample-A	As ground	870	Surface flaw
	As ground	957	Surface flaw
	As ground	904	Surface flaw
	Heat treated	1013	Internal defect
	Heat treated	855	Internal defect
	Heat treated	1135	Internal defect
Sample-B	As ground	1112	Surface flaw
	As ground	1062	Internal defect
	As ground	1049	Internal defect
	Heat treated	1225	Internal defect
	Heat treated	1338	Surface flaw
	Heat treated	1174	Internal defect

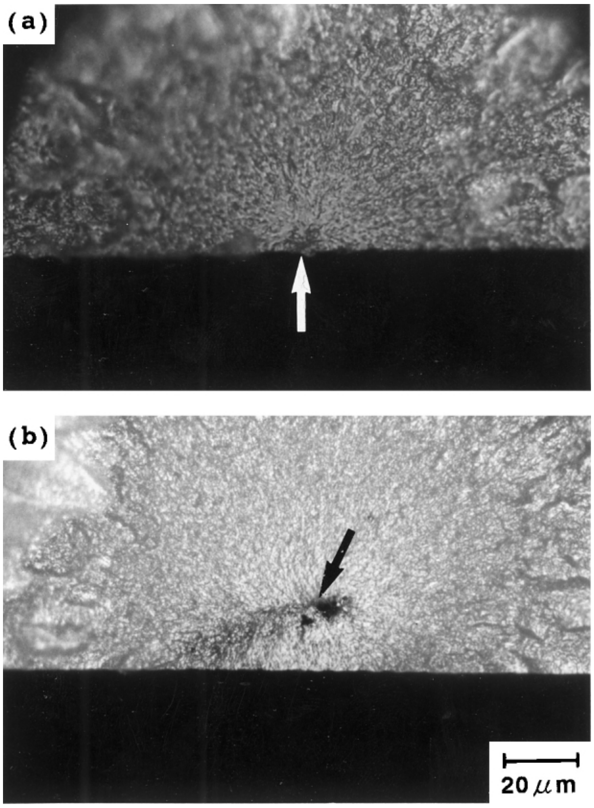

Fig. 1. Optical microgragh s of typical fracture origins of sample-A (an arrows indicates a fracture origin); (a): a surface flaw (as ground, σ_f = 904 MPa); (b): an internal defect (heat treated, σ_f = 855 MPa).

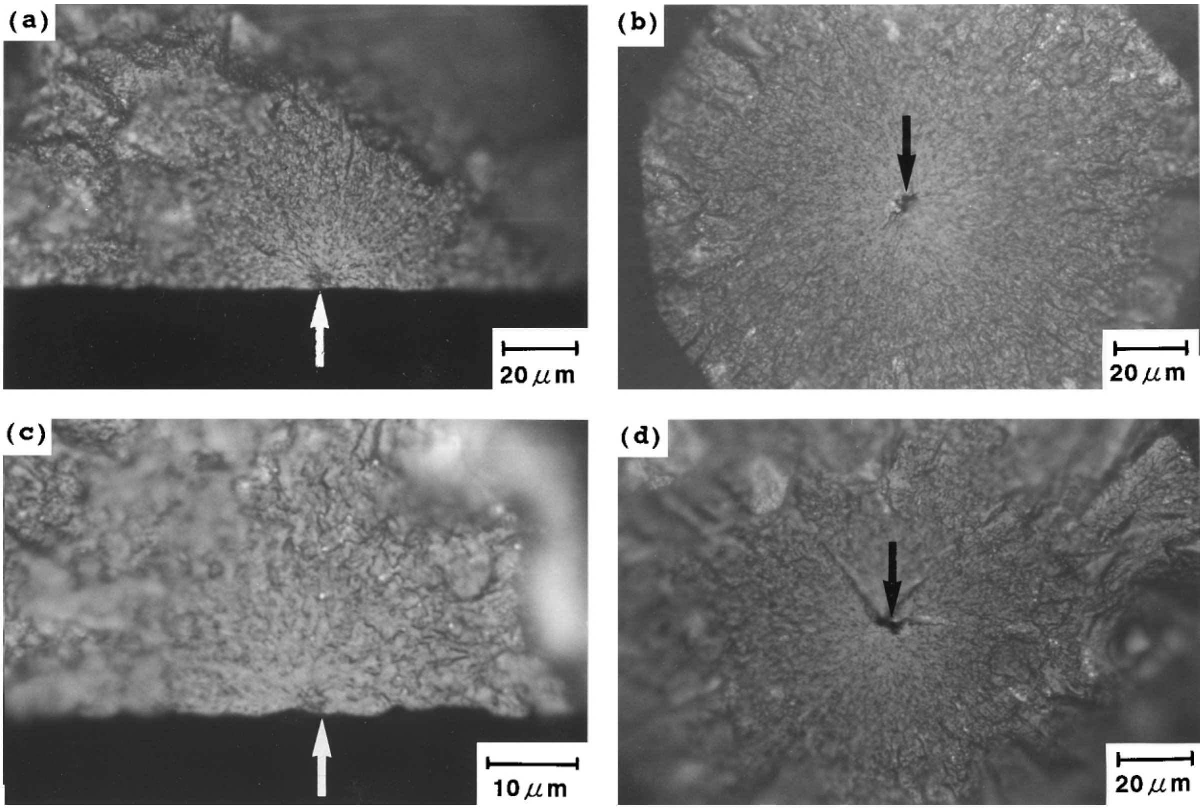

Fig. 2. Optical microgra ghs of typical fracture origins of sample-B. (an arrows indicates a fracture origin); (a): a surface flaw (as ground, $\sigma_f = 1112$ MPa); (b): an internal defect (as ground, $\sigma_f = 1062$ MPa); (c): a surface flaw (heat treated, $\sigma_f = 1338$ MPa); (d): an internal defect (heat treated, $\sigma_f = 1225$ MPa).

This change of fracture origin compared with sample-A suggested better ground surfaces than in the case of sample-A. The average strength of heat treated specimens of sample-B increased to 1.25 GPa. One of them was fractured from a surface flaw as shown in Fig. 2(c) and two were from an internal defect as shown in Fig. 2(d) with the size of less than 10 μm. Since all defects as fracture origin in sample-B were approximately the same size and smaller than those in sample-A, the higher strength of sample-B was assumed to result from the more homogeneous dispersion and mixing of Si_3N_4 and Al_2O_3 powders caused by the addition of Al(Oi–Pr)$_3$. Thus Al(Oi–Pr)$_3$ reacted with -OH on the surface of both Si_3N_4 and Al_2O_3 particles and improved their dispersability into a solvent as previously reported.[10] Even after heat treatment, the specimen fractured from a surface flaw showed the maximum strength of 1.34 GPa. It was considered that the more moderate surface finishing may bring further increase in strength for the specimens fractured from a surface flaw.

Even though hot-pressed β-sialons fabricated from Si_3N_4 and Al_2O_3 system showed varying strength, they have almost the same microstructure except for the particular defects acting as fracture origins, and the same mechanical properties other than strength.[2,3,6,13] In general it is accepted that the following relationship holds between K_{IC} and strength (σ_f):[14]

$$K_{IC} = \sigma_f Y c^{1/2} \tag{1}$$

where c is length of a fracture origin and Y is a dimensionless constant. The original size of surface flaw as a fracture origin could not be measured exactly because the original crack end incident to a surface machining flaw is indistinguishable on the fractured surface. However, in the case of the specimens fractured from internal defects, it is considered that this relationship between σ_f and c could be estimated. It is known that the stress applied at the internal fracture origin (σ) hold the following relationship with width of a specimen (a), thickness of a specimen (h), applied load (P), a span (L) and distance of a fracture origin from tensile surface (y).[15]

$$\sigma = \frac{3PL}{ah^3}\cdot(h/2 - y) = \sigma_f(1 - 2y/h) \tag{2}$$

Since y was about 18, 100 and 122 μm for the specimens shown in Figs. 1(b), 2(b) and (d) respectively, estimated σ by using Eq. (2) was 0.84, 1.00 and 1.13 GPa respectively. On the other hand, the length of fracture origins showing in Figs. 1(b), 2(b) and (c) was about 15, 10 and 8 μm respectively. The estimated strength by using Eq. (1) and the size of fracture origins was about 0.82 and 1.13 GPa for the specimens shown in Figs. 1(b) and 2(d) respectively, on the basis of the strength of the specimen shown in Fig. 2(b). These values corresponded to the stress applied at a fracture origin above mentioned approximately. Ultimately it is said to be that the strength of this β-sialons is controlled by the size of a defect acting as a fracture origin.

Takahasi et al.[8] reported that the fracture origins of HIPed Al_2O_3 material using the same Al_2O_3 powder were defects with the size of about 10 μm which consisted of inhomogeneously aggregated finer Al_2O_3 grains than matrix. This result suggested that the formation of these defects is due to the original property of this Al_2O_3 powder. It can be considered that a similar matter may occur in the case when β-sialon is prepared using this powder and the strength of it is still controlled by these defects, since the size of fracture origins in sample-A were approximately the same as this size.

4. Conclusion

It can be concluded that using fine grain size and high purity Al_2O_3 powder was effective to get high strength of β-sialon by decreasing the size of defects responsible for fracture origin comparing with previously used Al_2O_3 powder. Mixing of Si_3N_4 and Al_2O_3 powders in a $Al(Oi–Pr)_3$ solution brought further increase in strength because of decreasing size and number of defects. However, the strength of this β-sialon was still controlled by the defects with the size of less than 10 μm resulting from the original property of this Al_2O_3 powder.

References

1. Jack, K. H., Sialons and related nitrogen ceramics. *J. Mater Sci.*, 1976, **11**, 1135–1158.
2. Kishi, K., Umebayashi, S., Tani, E. and Kobayashi, K., Synthesis of β-sialon from Si_3N_4 and aluminum-iso-propoxide. *Yogyo-Kyokai-Shi*, 1985, **93**, 629–635.
3. Idem, Room temperature strength of β-sialon fabricated from aluminum-iso-propoxide and Si_3N_4. ibid., 1986, **94**, 179–182.
4. Idem, Effect of heat treatment on strength of β-Sialon. ibid., 1987, **95**, 630-637.
5. Kishi, K., Umebayashi, S. and Tani, E., Influence of microstructure on strength and fracture toughness of β-sialon. *J. Mater. Sci.*, 1990, **25**, 2780–2784.
6. Kishi, K., Umebayashi, S., Tani, E., Shobu, K., Pompe, R. and Kristoffersson, A., Effect of sedimentation and flocculation pretreatment of starting Si_3N_4 and $A1_2O_3$ powders on room temperature strength of β-sialon ($z=1$). *J. Eur. Ceram. Soc.*, 1998, **18**, 2015–2018.
7. Mizuta, M., Shibasaki, Y., Sakai, S., Katagiri, M. and Fujimoto, H., *J. Jpn. Soc. Powder and Powder Metall.*, 1988, **35**, 619–624.
8. Takahashi, T., Fujimura, Y. and Suzuki, H., Some properties of alumina ceramics having extremely fine grains. *J. Jpn. Soc. Powder and Powder Metall.*, 1993, **40**, 805–808.
9. Tashima, S., Hashimoto, S. and Kuroki, H., High speed centrifugal compaction and low temperature sintering of submicron alumina powder. *J. Jpn. Soc. Powder and Powder Metall.*, 1994, **41**, 180–183.
10. Kishi, K., Umebayashi, S., Pompe, R. and Persson, M., Interaction of aluminum-iso-propoxide solution and Si_3N_4 powder. *J. Ceram. Sci. Jpn.*, 1988, **96**, 698–701.
11. Kato, S., Iga, T., Hatano, S. and Isawa, Y., Synthesis of $NH_4AlO(OH)HCO_3$. *Yogyo-Kyokai-Shi*, 1976, **84**, 215–219.
12. Catalogue of Taimei Chemicals Co., Ltd.
13. Kishi, K., Umebayashi, S. and Kobayashi, K., Room temperature strength of β-sia1on fabricated using α-Si_3N_4 powder and aluminum hydroxide solution. *J. Ceram. Sci. Jpn.*, 1988, **96**, 546–550.
14. Yogyo-Kyokai ed. Seramikkusu no Kikaiteki Seisitsu, 1982. Yogyo-Kyokai, Tokyo, pp. 96.
15. Idem., pp. 6.