

# Fabrication of high strength $\beta$ -sialon by reaction sintering

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The reaction sintering of  $\beta$ -sialon ( $\text{Si}_4\text{Al}_2\text{O}_2\text{N}_6$ ) from a powder mixture of  $\text{Si}_3\text{N}_4$ ,  $\text{Al}_2\text{O}_3$  and AlN was studied to clarify factors affecting the densification. The presence of sufficient SiO vapour on the compact and excess oxide with respect to  $\beta$ -sialon composition was the most important factor. High density  $\beta$ -sialon was fabricated by heating the compact at  $1800^\circ\text{C}$  under 1 atm.  $\text{N}_2$ . The sintering was carried out with sufficient SiO vapour pressure to prevent thermal decomposition of sintered sialon by packing the compact with a powder mixture of  $\text{Si}_3\text{N}_4$  and  $\text{SiO}_2$ . Care was taken to minimize the amount of excess oxide in the starting composition to obtain a high density sialon with a small amount of intergranular X-phase. The maximum density of  $3.04\text{ g cm}^{-3}$  was obtained from the compact with 2 wt % excess  $\text{Al}_2\text{O}_3$  in the composition. The strength of the sintered  $\beta$ -sialon was  $490\text{ MN m}^{-2}$  at room temperature and  $480\text{ MN m}^{-2}$  at  $1200^\circ\text{C}$ . The values are the best among those so far published for sintered  $\beta$ -sialons.

## 1. Introduction

The sialon with  $\beta$ - $\text{Si}_3\text{N}_4$  structure is a solid solution between  $\text{Si}_3\text{N}_4$  and  $\text{AlN}\cdot\text{Al}_2\text{O}_3$  and is represented by a general formula  $\text{Si}_{6-z}\text{Al}_z\text{O}_2\text{N}_{8-z}$  in which  $z = 0$  to 4.2 [1]. Attempts have been made to use  $\beta$ -sialon for high temperature engineering materials because of the ease of fabrication, high strength, high creep resistance, low thermal expansion coefficient and high oxidation resistance of the sialon [1–3].

Some work has been done on the reaction sintering of  $\beta$ -sialon [4–6]. The method enables the formation of high strength sialon into complex shapes. Wills *et al.* [4] did not explain the details of the fabrication conditions, but they reported that the most important factor influencing densification behaviour was milling time. They required  $\geq 72$  h of milling time to obtain a high density  $\beta$ -sialon with  $z = 2$  in the system  $\text{Si}_3\text{N}_4$ – $\text{Al}_2\text{O}_3$ –AlN. Briggs [5] obtained sintered  $\beta$ -sialons with  $z = 1, 2$  and 4 in the system  $\text{Si}_3\text{N}_4$ – $\text{Al}_2\text{O}_3$ –AlN by heating the compact in closely packed  $\text{Si}_3\text{N}_4$  powder. He also referred to the importance of

milling time. Gauckler *et al.* [6, 7] obtained high density  $\beta$ -sialon with  $z = 4$  in the system  $\text{Si}_3\text{N}_4$ – $\text{Al}_2\text{O}_3$ –AlN and  $\text{SiO}_2$ –AlN by heating the compact in a loose powder mixture with the same composition.

Previous workers paid no attention to the effect of a small amount of excess oxide on densification and high temperature strength. The work on high temperature strength of hot pressed  $\beta$ -sialon showed that the presence of a small amount ( $\geq 4$  wt %) of intergranular X-phase greatly degraded the strength, whereas single phase materials retained their strength up to  $1200^\circ\text{C}$  [8]. Lumby and co-workers [9] found that  $\beta$ -sialon is a solid solution of  $\text{Si}_3\text{N}_4$  with  $\text{AlN}\cdot\text{Al}_2\text{O}_3$  and can be represented by  $\text{Si}_{6-z}\text{Al}_z\text{O}_2\text{N}_{8-z}$ . The solid solution does not require vacancies, as originally assumed for solid solubility of  $\text{Al}_2\text{O}_3$  into the  $\text{Si}_3\text{N}_4$  structure [2, 10], to replace Si by Al, and N by O atoms respectively. They also reported that the creep properties of hot pressed sialons were improved by allowing the composition to approach the single phase  $\beta$ -sialon (“balanced”

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composition) from the  $\beta$ -sialon + X-phase region, which is accompanied by a decrease in the intergranular glass or X-phase [9, 11].

Recent work on thermal decomposition of sialons [12, 15] showed that two-phase material containing  $\beta$ -sialon and X-phase decomposed thermally to  $\beta$ -sialon single phase, SiO and  $N_2$ . The single phase  $\beta$ -sialon decomposed to 15R-sialon [1], SiO and  $N_2$ . This revealed the importance of maintaining sufficient SiO vapour pressure to prevent thermal decomposition of sintered sialon during heating.

Hot pressing studies on  $\beta$ -sialon formation with a "balanced" powder in the  $Si_3N_4-Al_2O_3-AlN$  system [13] suggested that a sialon with  $z = 2$  was easy to fabricate into a high density, single phase material. The mechanical properties of hot pressed  $\beta$ -sialon were improved with decreasing the solid solubility of  $AlN \cdot Al_2O_3$  in the  $\beta$ -sialon [14].

The present work reports the effect of small amounts of excess oxide and SiO vapour on densification in the system  $Si_3N_4-Al_2O_3-AlN$ . The composition was restricted to  $z = 2$ ,  $Si_4Al_2O_2N_6$ . The strength of the sintered  $\beta$ -sialon was determined at room temperature and  $1200^\circ C$  and discussed in terms of the amount of intergranular X-phase compared with that of hot pressed  $\beta$ -sialon.

## 2. Experimental procedures

The starting materials were  $Si_3N_4$  (Advanced Materials Engineering, England, high purity grade: Si 58.6, N 37.9, Fe 0.25, Al 0.18 and O 1.2 wt %; average particle size  $0.5 \mu m$ ),  $\alpha-Al_2O_3$  (Sumitomo Chemical Ind., Japan, 99.99% pure; average particle size  $0.2 \mu m$ ) and AlN (Tokyo Shibaura Electric Co., Japan: Al 65.0, N 32.3, Si 0.2, Fe 0.1 and O 1.8 wt %; average particle size  $2 \mu m$ ). The powders were weighed and mixed by ball milling with alumina balls for 4 h and put in n-hexane and heated to form  $\beta$ -sialon by the reaction.



The presence of surface silica on  $Si_3N_4$  was compensated for by the addition of a corresponding amount of AlN. The amount of  $Al_2O_3$  in the starting powder mixture was determined to give desired composition, from alumina deficient to rich with respect to  $z = 2$  sialon composition, after ball milling. (All compositions used in present study are shown in Fig. 6 in a part of  $Si_3N_4-SiO_2-Al_2O_3-AlN$  phase diagram).

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About 1.5 g of the powder mixture was pressed in a steel die of diameter 12 mm under a pressure of  $50 MN m^{-2}$ . The compact was then pressed isostatically under  $150 MN m^{-2}$ . The compact was placed in a reaction sintered silicon nitride crucible and covered with a loose powder of an equimolar mixture of  $Si_3N_4$  and  $SiO_2$ , or another powder mixture. The crucible was placed in a graphite container, which was also a susceptor for high frequency induction, and heated to the desired temperature under 1 atm.  $N_2$ .

The change in crystalline composition during heating was investigated by X-ray diffraction. The  $z$  value of sintered  $\beta$ -sialon was obtained by determining unit cell dimensions from the (1 1 1) and (3 0 1) diffraction peaks.

The microstructures of the sintered sialons were observed using a scanning electron microscope. The specimens for strength determination were obtained by heating rectangular bars, about  $5.0 \times 6.5 \times 40 mm$ , at  $1800^\circ C$  for 90 min. Three-point bending strength was measured with a span of 30 mm at a cross-head speed of  $0.5 mm min^{-1}$  in air at room temperature and in  $N_2$  at  $1200^\circ C$ . The specimen was heated at a rate of  $270^\circ C h^{-1}$  and held at  $1200^\circ C$  for 15 min before being fractured.

## 3. Results and discussion

### 3.1. Sintering under SiO vapour

Recent work on the thermal decomposition of  $\beta$ -sialon [12, 15] has shown that the single phase  $\beta$ -sialon decomposed thermally to 15R-sialon, SiO and  $N_2$  when the SiO pressure on the  $\beta$ -sialon is less than  $3.9 \times 10^{-1}$  atm. at  $1800^\circ C$ . When the SiO partial pressure exceeds  $6.0 \times 10^{-1}$  atm. on the other hand, the condensation of gaseous materials as X-phase sialon in sintered  $\beta$ -sialon is detected. The stable region of single phase  $\beta$ -sialon in terms of SiO partial pressure was estimated as,  $6.0 \times 10^{-1} > p_{SiO} > 3.9 \times 10^{-1}$  atm. at  $1800^\circ C$  assuming  $p_{N_2} + p_{SiO} = 1$  atm. [15]. The weight loss due to thermal decomposition of sialon took place throughout the compact and resulted in the shift in composition to the AlN corner in  $Si_3N_4-SiO_2-Al_2O_3-AlN$  phase diagram [1]. When the composition became AlN-rich with respect to the sialon composition, the compact sintered no further, resulting in incomplete shrinkage. This fact shows the necessity of maintaining SiO vapour to prevent thermal decomposition of the compact during heating.

TABLE I The effect of various powder mixtures on the weight loss and density of sintered sialon

Powder mixture (wt %)	Weight loss (wt %)	Density of sintered sialon	
		Bulk density ( $\text{g cm}^{-3}$ )	Porosity (%)
$\text{Si}_3\text{N}_4$	14.2	1.72	44.7
$\text{Si}_3\text{N}_4 + \text{Al}_2\text{O}_3 + \text{AlN}^*$ (66; 24; 10)	7.3	2.11	32.2
$\text{Si}_3\text{N}_4 + \text{Al}_2\text{O}_3$ (60; 40)	4.1	2.58	17.1
$\text{Si}_3\text{N}_4 + \text{SiO}_2$ (70; 30)	0.4	2.96	4.9

\* Corresponds to  $\beta$ -sialon with  $z = 2$ .

Various kinds of powder mixtures, which produce SiO at high temperatures, were tested for a buffer to keep the SiO vapour around the compact. The powder mixture of  $\text{Si}_3\text{N}_4$ ,  $\text{Al}_2\text{O}_3$  and AlN containing 2 wt% excess alumina over the  $z = 2$  composition was used for the present study because of the lack of sinterability of the "balanced" composition. The compact was covered with various kinds of powder mixtures and heated for 60 min at  $1800^\circ\text{C}$  under 1 atm.  $\text{N}_2$ . The results are listed in Table I. The results after heating in  $\text{Si}_3\text{N}_4$  powder are also listed in the table for comparison.

The reaction sintering of a sialon with  $z = 2$  in a loose powder of  $\text{Si}_3\text{N}_4$  or similar composition was not successful. The sintering in powder mixtures of  $\text{Si}_3\text{N}_4$  and  $\text{SiO}_2$  was the most successful.

The difficulty of sintering silicon nitride compared to other covalent solids is due to the higher decomposition pressure of  $\text{Si}_3\text{N}_4$  at sintering temperature. The equilibrium partial pressure of  $\text{N}_2$  on  $\text{Si}_3\text{N}_4$  is calculated as  $1.4 \times 10^{-1}$  atm. at  $1700^\circ\text{C}$ . The weight loss due to thermal decomposition of  $\text{Si}_3\text{N}_4$  during heating inhibits the densification because of pore formation on the surface of the compact. The addition of a densification aid, which enhances liquid phase sintering at low temperatures, might result in the formation of low strength material at high temperatures. The successful sintering of  $\text{Si}_3\text{N}_4$  under high nitrogen pressures [16, 17] was emphasized from the thermodynamic point of view. The method enabled the sintering of  $\text{Si}_3\text{N}_4$  at higher temperatures than  $1900^\circ\text{C}$  at which the diffusion of materials should be accelerated.

Thermal decomposition of sialon was due to the volatilization of SiO and  $\text{N}_2$  as already shown. Under  $\text{N}_2$  atmosphere, the weight loss was mainly controlled by the partial pressure of SiO on  $\beta$ -sialon.

A slight densification of the compact when heated in loose powder of  $\text{Si}_3\text{N}_4$  (Table I) was

explained by the thermal decomposition of sialon due to the absence of SiO vapour around the compact. The heating in loose powder of the same composition also resulted in incomplete densification. The weight loss during heating was much larger than that during heating in the mixture of  $\text{Si}_3\text{N}_4$  and  $\text{SiO}_2$ .

The necessity of maintaining sufficient SiO vapour pressure on the surface of the compact during sintering to fabricate high density sialon, as will be shown later, required the presence of  $\text{Si}_2\text{N}_2\text{O}$  or X-phase around the compact. The powder mixture with the same composition did not provide sufficient SiO vapour to prevent the thermal decomposition of  $\beta$ -sialon.

Wills *et al.* [4] could fabricate high density sialon with  $z = 2$  in the  $\text{Si}_3\text{N}_4\text{-Al}_2\text{O}_3\text{-AlN}$  system. They pointed out that the most significant variable was the milling time. They obtained a sialon with maximum density of  $3.04 \text{ g cm}^{-3}$  by sintering a powder mixture milled for 72 h. The second most important variable in their work was the type of  $\text{Si}_3\text{N}_4$  powder. Briggs [5] has also referred to the importance of milling time in the fabrication of high density sialon by heating the compact in closely packed  $\text{Si}_3\text{N}_4$  powder. Gaukler *et al.* [18] and Boskovic *et al.* [6] succeeded in the fabrication of sialon with  $z = 4$  in the system  $\text{Si}_3\text{N}_4\text{-Al}_2\text{O}_3\text{-AlN}$  and  $\text{SiO}_2\text{-AlN}$  by heating the compact in a loose powder of the same composition. They reported the fact that powder mixture milled for 1 h in alcohol containing a small amount of water densified easily [18]. They concluded that oxide formation on AlN was essential to produce high density sialon.

The present authors [13] and Benn *et al.* [19] have pointed out the effect of overall composition on densification behaviour during hot pressing. A small shift in composition from  $\beta$ -sialon to the oxide corner in the  $\text{Si}_3\text{N}_4\text{-SiO}_2\text{-Al}_2\text{O}_3\text{-AlN}$  phase diagram [1] greatly accelerated the densification. The presence of a small amount of excess

oxide at a grain boundary in hot pressed sialon, which is generally the case, degraded the high temperature strength [8]. During the reaction sintering of  $\beta$ -sialon, the acceleration of densification was also observed when the composition was shifted from the  $\beta$ -sialon towards the oxide corner, as will be shown later.

The composition of the compact used in the present investigation had 2 wt% excess  $\text{Al}_2\text{O}_3$  over the  $z = 2$  sialon composition. When the composition had a high oxide content, the compact densified easily without the addition of sufficient  $\text{SiO}$  pressure on the compact. The successful sintering of sialon in closely packed  $\text{Si}_3\text{N}_4$  [5] might be attributed to the presence of such excess oxide. The sintering of oxidized powder mixture [18] is also related to the shift in composition to the oxide corner. When the deviation in composition was less than 4 wt%, the compact did not densify without the control of  $\text{SiO}$  pressure. The reported successful fabrication of sialons might be attributed to the presence of  $>4\%$  excess oxide, which should degrade the high temperature properties of sintered materials.

The shift in composition to the oxide corner resulted in the formation of two-phase material which consisted of  $\beta$ -sialon and X-phase (or glass phase) [20]. The equilibrium  $\text{SiO}$  partial pressure of X-phase was higher than that of  $\beta$ -sialon at the same temperature [15]. The shrinkage of sintered sialon in the powder mixture of  $\text{Si}_3\text{N}_4$  and  $\text{Al}_2\text{O}_3$  (Table I) was explained by the formation of X-phase in the mixture. The fabrication of high density sialon without appreciable weight loss by heating the compact in powder mixture of  $\text{Si}_3\text{N}_4$  and  $\text{SiO}_2$  was attributed to the formation of  $\text{Si}_2\text{N}_2\text{O}$  in the mixture which had higher  $\text{SiO}$  partial pressure than  $\beta$ -sialon.

All evidence shown here suggests that the compact must be sintered under sufficient  $\text{SiO}$  vapour pressure to fabricate high strength  $\beta$ -sialon at high temperature.

### 3.2. Reaction process during sintering

The sintering process of the compact during heating in a loose powder mixture of  $\text{Si}_3\text{N}_4$  and  $\text{SiO}_2$  was investigated at 1600 to 1850°C. The compact was heated to the desired temperature at a constant rate of  $10^\circ\text{C min}^{-1}$  and then quenched immediately the temperature was reached. The crystalline composition both on and in the compact was determined by X-ray diffraction. Fig. 1 shows the

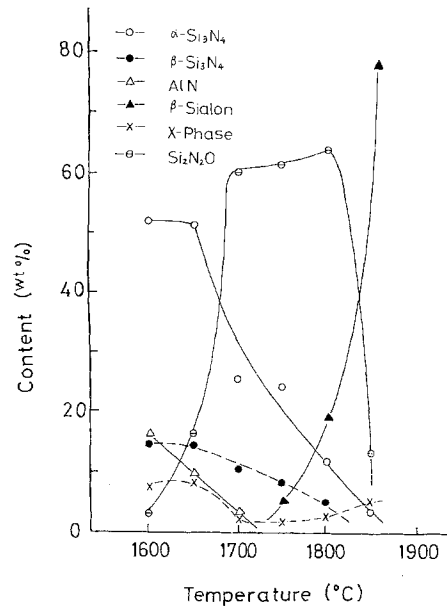


Figure 1 The change in composition on the surface of the compact with temperature.

change in crystalline composition on the surface of the compact with temperature. The formation of  $\text{Si}_2\text{N}_2\text{O}$  should be noted here. There was no  $\text{Si}_2\text{N}_2\text{O}$  in the compact as seen in Fig. 2. Fig. 3 shows the change in weight and density of the compact during heating. The weight gain observed at 1650 to 1800°C might be related to the reaction of the surface with surrounding powder mixture to form  $\text{Si}_2\text{N}_2\text{O}$ .

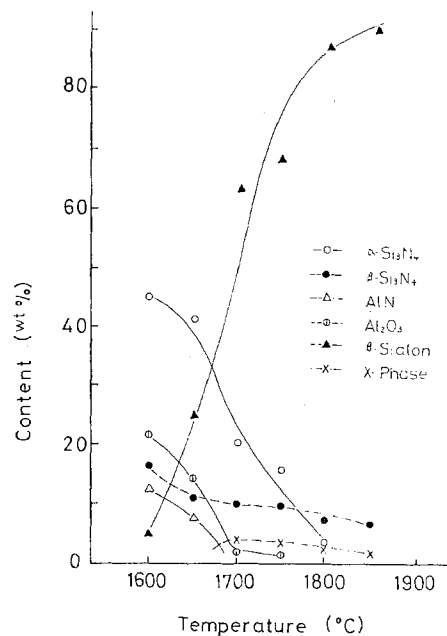


Figure 2 The change in composition in the compact with temperature.

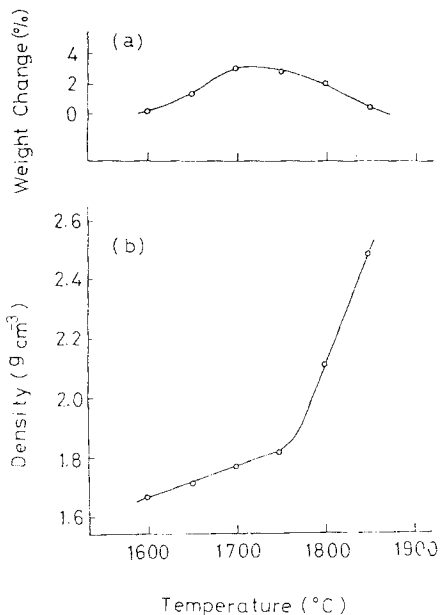
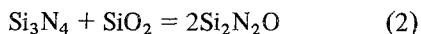


Figure 3 The change in (a) weight and (b) density of the compact during heating.

In the present system, shrinkage was observed at 1750°C, which was about 150°C higher than that in hot pressing of  $\beta$ -sialon with same composition. The reactions observed during reaction sintering of sialon were basically similar to those in hot pressing [13]: (i) the formation of  $\beta$ -sialon with  $z \sim 3$  and the X-phase at temperatures lower than 1700°C; and (ii) the reaction of  $\beta$ -sialon with  $\text{Si}_3\text{N}_4$  to form  $z = 2$  sialon at temperatures higher than 1700°C.

The  $z$ -values of the sintered sialon were 3 at 1600°C and 2.7 at 1700°C. The value decreased gradually from 2.3 to 2.0 in 30 min at 1800°C.

In the present method, the densification is closely related to the formation of  $\text{Si}_2\text{N}_2\text{O}$  on the surface of the compact and in the loose powder around the compact. The  $\text{Si}_2\text{N}_2\text{O}$  layer on the surface was formed by the reaction of the  $\text{Si}_3\text{N}_4$  on the compact and the surrounding  $\text{SiO}_2$  resulting in a weight gain as shown in Fig. 3.



The  $\text{Si}_2\text{N}_2\text{O}$  layer was less than 100  $\mu\text{m}$  in thickness and did not contribute to the shift in composition of sintered sialon. It might be reasonable to assume that  $\text{Si}_2\text{N}_2\text{O}$  on the compact and in the surrounding powder prevented the thermal decomposition of  $\beta$ -sialon during sintering by producing SiO by a reaction,

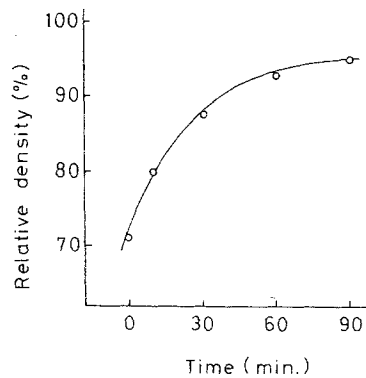
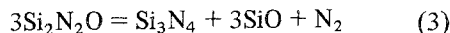


Figure 4 The densification curve at 1800°C.



The equilibrium partial pressure of SiO on  $\text{Si}_2\text{N}_2\text{O}$  was calculated as  $4.8 \times 10^{-1}$  atm. at 1800°C, assuming  $p_{\text{SiO}} + p_{\text{N}_2} = 1$  atm. The value is in the stable region of single phase sialon. The  $\text{Si}_2\text{N}_2\text{O}$  phase on the compact and in the surrounding powder disappeared completely after heating at 1800°C for 1 h or longer. The solid phase remaining around the compact was  $\beta$ - $\text{Si}_3\text{N}_4$ , which coincided with that expected from Reaction 3.

Fig. 4 shows the densification curve at 1800°C. The maximum density was obtained by heating for 90 min. The average density was  $2.97 \text{ g cm}^{-3}$  (95.4% of theoretical density  $3.112 \text{ g cm}^{-3}$  [13]). The maximum density obtained was  $3.04 \text{ g cm}^{-3}$  (97.7% of theoretical). The compact shrank uniformly with a linear shrinkage of about 16%. The weight loss after heating for 90 min was less than 1 wt%. Longer heating resulted in a greater weight loss and lower density.

One of the most important factors to be controlled in fabricating high density sialon, besides the SiO pressure and the composition of the compact, is the sintering temperature. It must be maintained at  $1800 \pm 30^\circ \text{C}$ . The chemical reactions and the volatilization of SiO in the surrounding powder sometimes made it difficult to control the temperature in a restricted range. At lower temperatures, the shrinkage was too little to obtain high density material. At higher temperatures, thermal decomposition of sintered sialon was observed and metallic Si was sometimes detected on the surface.

The fractured surface of sintered sialon with a density of  $3.02 \text{ g cm}^{-3}$  was etched with a diluted solution of HF and  $\text{HNO}_3$  at 70°C for 5 min. The

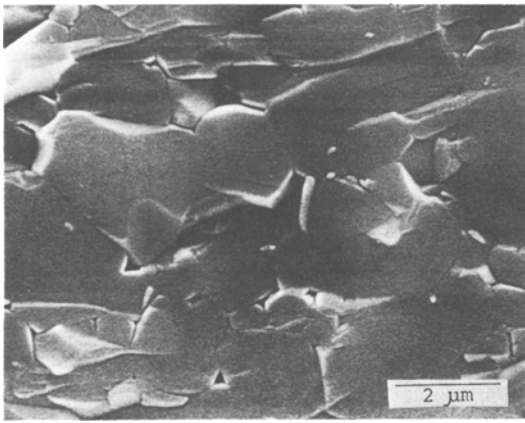


Figure 5 SEM photograph of the fracture surface of sintered  $\beta$ -sialon

surface is shown in Fig. 5. The closed pores were less than  $5\ \mu\text{m}$  in size and distributed randomly. The grain size of the sialon was about  $2\ \mu\text{m}$ . X-ray diffraction studies revealed that a small amount of X-phase ( $\sim 2\ \text{wt}\%$ ) and  $\beta$ - $\text{Si}_3\text{N}_4$  ( $\sim 3\ \text{wt}\%$ ) remained in the material.

### 3.3. The effect of excess alumina

The great effect of composition on densification and high temperature strength of sialon has already been pointed out in Section 3.1. The effect of the deviation in composition from  $z = 2$  sialon on densification was studied. The amount of surface silica on  $\text{Si}_3\text{N}_4$  powder was compensated by the addition of AlN. The amount of  $\text{Al}_2\text{O}_3$  in the starting powder mixture was determined to give restricted content after ball milling for 4 h by alumina balls and pot. The compositions studied here were plotted in a part of the  $\text{Si}_3\text{N}_4$ - $\text{SiO}_2$ -

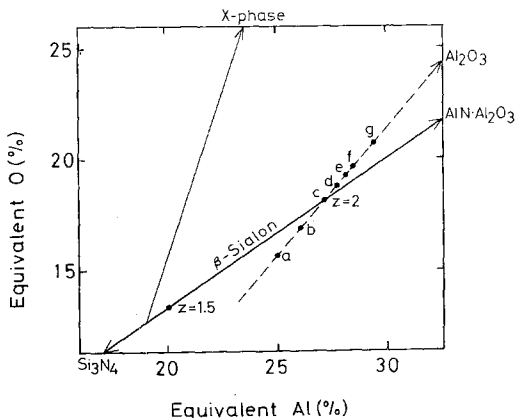


Figure 6 All compositions investigated in present work shown as a part of the  $\text{Si}_3\text{N}_4$ - $\text{SiO}_2$ - $\text{Al}_2\text{O}_3$ -AlN phase diagram [1].

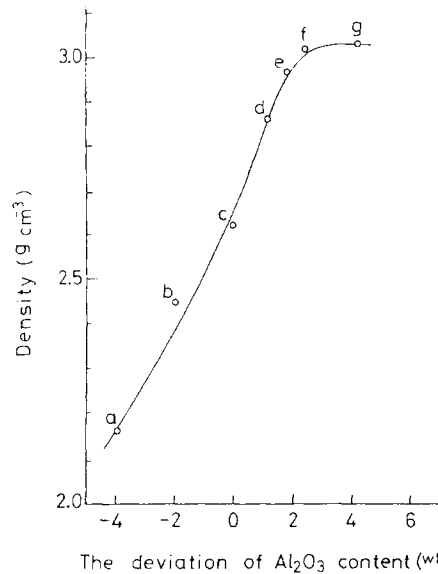


Figure 7 The effect of the degree of deviation of  $\text{Al}_2\text{O}_3$  content from  $z = 2$  sialon composition on densification.

$\text{Al}_2\text{O}_3$ -AlN phase diagram [1] (Fig. 6). The compact was heated in the powder mixture of  $\text{Si}_3\text{N}_4$  and  $\text{SiO}_2$  at  $1800^\circ\text{C}$  for 90 min under 1 atm.  $\text{N}_2$ . The result is shown in Fig. 7. The balanced composition, which is shown as the origin of the horizontal axis, is the composition for single phase  $\beta$ -sialon. The figure shows that the density of sintered sialon increased with the increase in  $\text{Al}_2\text{O}_3$  content. The composition for single phase sialon was difficult to sinter into high density material.

The single phase and fully dense  $\beta$ -sialon could be obtained by hot pressing a balanced powder mixture [13]. The compact densified with the presence of liquid phase at  $1600$  to  $1700^\circ\text{C}$ . At higher temperatures, the liquid disappeared by reaction with  $\text{Si}_3\text{N}_4$ , 15R-sialon and  $\text{Al}_2\text{O}_3$  to form  $\beta$ -sialon. The process was referred to as "transient" liquid phase sintering [21]. In reaction sintering, the densification curve shifted to about  $150^\circ\text{C}$  higher than that in hot pressing. The processes of densifying by "transient" liquid phase sintering and eliminating the liquid phase by chemical reaction overlapped in the present case, resulting in incomplete densification.

To fabricate high density sialon, with density greater than  $2.90\ \text{g cm}^{-3}$  (93% of theoretical), the presence of more than 1.5 wt% excess  $\text{Al}_2\text{O}_3$  was necessary. The important conclusion deduced from Fig. 7 is that the sinterability of the compact was only the function of the composition if the same starting powders were used. When two

compacts with the same composition obtained by different milling time were heated under the same conditions, they gave about the same density.

Present work showed the difficulty in fabricating fully dense and single phase  $\beta$ -sialon by reaction sintering. The amount of intergranular phase may affect the high temperature strength of sintered sialon as in hot pressed  $\text{Si}_3\text{N}_4$  with  $\text{MgO}$  [22]. The improvement of properties should be possible by minimizing the amount of excess oxide.

### 3.4. Strength of the sintered sialon

The powder mixture with the composition containing 2.4 wt% excess alumina sintered to high density was used to measure the strength of sintered sialon, as shown in Fig. 7. The powder mixture was pressed uniaxially followed by isostatic pressing to form a rectangular shape. The compact was heated in a mixture of  $\text{Si}_3\text{N}_4$  and  $\text{SiO}_2$  at  $1800^\circ\text{C}$  for 90 min under 1 atm.  $\text{N}_2$ . The linear shrinkage was about 15%. The bulk density of sintered sialon was about  $2.99\text{ g cm}^{-3}$  (96.1% of theoretical).

After polishing the surface, a specimen about  $4.5 \times 6.0 \times 40\text{ mm}$  in size was tested for three-point bending strength over a span 30 mm at room temperature, and also at  $1200^\circ\text{C}$  in  $\text{N}_2$  atmosphere.

The strength of four specimens at room temperature was  $490 \pm 40\text{ MN m}^{-2}$ ; Wills *et al.* [4] measured the strength of a sialon with  $z = 2$  over a span 9.53 mm. The average strength was  $350\text{ MN m}^{-2}$ . Briggs [5] obtained the strength of sialon with  $z = 4$  over a span 10 mm. The values were in the range 390 to  $430\text{ MN m}^{-2}$ . The value measured with a smaller specimen is generally higher than that with a larger one. The difference in strengths between the present work and that by Wills *et al.* and Briggs would be greater if the measurements were carried out under the same conditions.

The room temperature strength of  $490\text{ MN m}^{-2}$  obtained in the present work is the largest value published so far. It is comparable with those of hot pressed single phase sialon. The strength of hot pressed sialon containing about 4 wt% by present authors [8] and  $530\text{ MN m}^{-2}$  obtained by Gauckler *et al.* [14].

The bending strength of four specimens at  $1200^\circ\text{C}$  was determined as  $480 \pm 40\text{ MN m}^{-2}$ . The strength did not degrade at  $1200^\circ\text{C}$  as in the case

of hot pressed sialon containing about 4 wt% X-phase as an intergranular phase was  $650\text{ MN m}^{-2}$  at room temperature and  $490\text{ MN m}^{-2}$  at  $1200^\circ\text{C}$ , which was about 75% of room temperature strength. The amount of X-phase in reaction sintered sialon studied here was about 2 wt%. The difference in mechanical properties between two sialons at high temperature could presumably be related to the difference in X-phase content. The difference in distribution of X-phase might also be responsible. In hot pressed sialon, the X-phase was distributed randomly, whereas there was no X-phase on the surface of reaction sintered sialon.

Further work on measurements of high temperature strength is now going on and will be published separately.

## 4. Conclusions

(1) High strength  $\beta$ -sialon with  $z = 2$  in  $\text{Si}_{6-z}\text{Al}_z\text{O}_z\text{N}_{8-z}$  was obtained by heating the compact in a loose powder mixture of  $\text{Si}_3\text{N}_4$  and  $\text{SiO}_2$  to maintain sufficient  $\text{SiO}$  vapour pressure on the compact to prevent the thermal decomposition of the sintered sialon.

(2) The important variable which affected the densification behaviour was the composition of the compact. If the composition was rich in oxide, the compact sintered easily. The minimum amount of excess  $\text{Al}_2\text{O}_3$  to get high density  $\beta$ -sialon,  $> 2.90\text{ g cm}^{-3}$  (93% of theoretical), was 1.5 wt%.

(3) The three-point bending strength of sintered sialon containing 2.4 wt% excess  $\text{Al}_2\text{O}_3$  was  $490\text{ MN m}^{-2}$  at room temperature and  $480\text{ MN m}^{-2}$  at  $1200^\circ\text{C}$ . The strength was the greatest value for reaction sintered sialon published until now.

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