The Effect of Interfacial Chemical Reaction on the Mechanical Properties of Si_3N_4 Whisker/ Al_2O_3 Composite

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Si_3N_4 ウィスカー/ Al_2O_3 複合材料の力学的性質に及ぼす界面化学反応の影響

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Si_3N_4 whisker/ Al_2O_3 composites with various amounts of interfacial β -SiAlON (β' -phase, $\text{Si}_{6-z}\text{Al}_z\text{O}_z\text{N}_{8-z}$) were prepared while controlling the hot-pressing temperature and duration. X-ray diffraction analysis revealed that interfacial chemical reaction produced X-phase and β -SiAlON with a Z-value of approximately 4. A model of the interfacial chemical reaction was proposed to explain the experimental results. The mechanical properties of the composites hot-pressed above 1600°C were markedly different from those of the composites hot-pressed below 1600°C , because X-phase at the whisker/matrix interface was liquefied during the hot-pressing above 1600°C and vitrified X-phase seemed to increase interfacial bonding strength.

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1. Introduction

It is well known that whisker reinforcement is effective for overcoming the brittleness of structural ceramics. Hence, many researchers¹⁾ have studied the fracture resistance of ceramic whisker-reinforced ceramic composites and proposed various toughening mechanisms such as crack bowing,²⁾ crack deflection,^{3),4)} whisker elastic and/or pull-out bridging,^{5),6)} and debonding at the whisker/matrix interface.⁷⁾ We believe that whisker bridging contributes to toughening^{5),6)} much more than any other mechanisms. The physical and chemical bonding at the whisker/matrix interface must be adequately controlled to obtain a composite with high fracture resistance, because the crack extension resistance brought about by whisker bridging is susceptible to interfacial bonding strength. We have already reported that both fracture toughness and the work-of-fracture of SiC whisker/ Al_2O_3 composites vary with the geometrical configuration⁸⁾ and the chemical bonding⁹⁾ of the whisker/matrix interface.

SiC whiskers with superior mechanical properties are usually used as the reinforcing material for ceramic composites. In SiC-whisker reinforced ceramics (e.g., Al_2O_3 matrix and Si_3N_4 matrix), no interfacial chemical reaction is detected as reported by Braue et al.¹⁰⁾ who observed whisker/matrix interfaces by high-resolution transmission electron microscopy (TEM). That is one of the reasons why there is much research dealing with SiC-whisker reinforced ceramic composites. Recently, Si_3N_4 whiskers have also been manufactured industrially as reinforcing materials for structural materials. Several researchers have already conducted studies^{11)–14)} on Si_3N_4 whiskers and Si_3N_4 -whisker reinforced ceramic composites. According to thermodynamic considerations, Si_3N_4 reacts with Al_2O_3 to produce β -phase (β -SiAlON) at high temperatures.¹⁵⁾ Thus, it is expected that the Si_3N_4 whiskers react with the Al_2O_3 matrix at the whisker/matrix interfaces during sintering at high temperatures in Si_3N_4 whisker/ Al_2O_3 composites. Kondo et al.¹³⁾ and Tamari et al.¹⁴⁾ reported the mechanical properties as well as the sinterability, the thermal properties and the oxidation resistance of the Si_3N_4 whisker/ Al_2O_3 composite. Neither group referred to the relationship between the chemical reaction at the interface

and the fracture properties. They maintained that the composite could be fabricated without any chemical reaction at the interface.

In this study, we focused on the quantitative correlation between the interfacial chemical reaction and the fracture resistance of the Si_3N_4 whisker/ Al_2O_3 composite. Si_3N_4 whisker/ Al_2O_3 composites with various amounts of β -SiAlON were fabricated while controlling the maximum temperature and the duration of hot-pressing. Through TEM observation with energy-dispersive spectroscopy (EDS), the effect of the interfacial chemical reaction on the microstructure of the whisker/matrix interface was discussed, and a model of the interface during chemical reaction was proposed. After that, the relationships between the interfacial chemical reaction and the mechanical properties were investigated.

2. Experimental procedure

2.1 Raw materials

The authors used Al_2O_3 powder supplied by Taimai Chemicals (Taimicron TM-100; γ - $\text{Al}_2\text{O}_3 > 99.99\%$, average primary particle diameter: $0.01\ \mu\text{m}$, BET specific surface area: $140\ \text{m}^2/\text{g}$; according to catalog values) as the raw material of the matrix. Si_3N_4 whiskers supplied by Ube Industries (UBE-SN-WB; β - $\text{Si}_3\text{N}_4 > 98\%$, diameter: 0.5 – $1.5\ \mu\text{m}$, length: 10 – $30\ \mu\text{m}$; according to catalog values) were used in this study as the reinforcing material of the composites. Observations by scanning electron microscopy (SEM) revealed that the Si_3N_4 whiskers were almost straight in their longitudinal direction with smooth surfaces.

2.2 Fabrication of Si_3N_4 whisker/ Al_2O_3 composites

Agglomerated Si_3N_4 whiskers were dispersed by ultrasonic wave vibration in *n*-butyl alcohol for 10 min. After that, they were additionally dispersed in *n*-butyl alcohol with a tumbling mixer for 4h using a Nylon pot and Nylon-coated steel balls. The deagglomerated Si_3N_4 whiskers were mixed with Al_2O_3 powder in *n*-butyl alcohol with the tumbling mixer for 20h. The volume fraction of Si_3N_4 whiskers in the composite was adjusted to be 10%. The mixed slurry was quickly dried with a rotary evaporator to prevent the separation of the whiskers from the powder owing to the differ-

ence of sedimentation speed. In addition, the mixed powder was fully dried at 100°C for 24h in a drying oven. The composites were fabricated by hot-pressing of the mixed powder, which was carried out at $1550\text{--}1700^\circ\text{C}$ under 33 MPa of pressure for 1–3h in an N_2 gas atmosphere.

2.3 Procedure for evaluation

The apparent density of specimens was measured by the Archimedes method with *n*-butyl alcohol. The four-point bending strength of specimens was evaluated on the basis of JIS R 1601 (inner span : 10 mm and outer span : 30 mm). It has been reported that whiskers lie vertically to the hot-pressing direction at two-dimensional random.¹⁶⁾ The plane on the tensile side of each specimen was prepared perpendicular to the hot-pressing direction and was finished by longitudinal grinding with a #600 diamond wheel. Hence, the main cracks in each specimen should propagate perpendicularly to the plane in which the whiskers were preferentially oriented. The elastic modulus of specimens was evaluated with a strain gauge (Showa Sokki Co., Ltd., Type N22-FA-1-120-11-VS1, gauge length : 1 mm) during a four-point bending test based on JIS R1602. Fracture toughness was evaluated with a chevron notched specimen in the three-point bending test (thickness : 3 mm, width : 4 mm, span : 30 mm) under a cross-head speed of 0.005 mm/min.¹⁷⁾ The tip angle of the chevron was designed to be 90° . The notches of all specimens were machined parallel to the hot-pressing direction. Thereby, the main crack should propagate perpendicularly to the plane with whisker orientation. The detailed explanation of the fracture toughness evaluation was given in Ref. 17). At the same time, the work-of-fracture Γ was also evaluated with a chevron notched specimen, and was derived from the work (U) done by a testing machine and the projected area (A) of the fractured surface using the following equation.¹⁸⁾

$$\Gamma = U/2A \quad (1)$$

These estimations were executed only for the specimens whose main cracks propagated quasi-statically until the separation of the specimens into two pieces. The fractured surfaces of the chevron notched specimens were observed by scanning electron microscope (SEM). X-ray diffraction (XRD) analysis of the crushed powder of the composites was carried out to identify the interfacial reaction products and quantify the β -SiAlON content in Si_3N_4 whiskers (called the volume fraction of β -SiAlON in Si_3N_4 whiskers : R_{SiAlON}). The lattice parameters of β -SiAlON are larger than those of β - Si_3N_4 owing to solid solution.¹⁹⁾ Thus, the X-ray peaks due to β -SiAlON appear as β - Si_3N_4 peaks shifted to the low angle side. Curve fitting was carried out for three X-ray (Cu K α) peaks diffracted at $2\theta=35.1^\circ$ for (104) of α - Al_2O_3 and at $2\theta=36.1^\circ$ for (210) and $2\theta=33.7^\circ$ for (101) of β - Si_3N_4 using the Pearson Type VII function²⁰⁾ to identify the β -SiAlON peaks. Then, the value of the R_{SiAlON} was obtained using the following equation:

$$R_{\text{SiAlON}} = A_{\text{SiAlON}} / \{A_{\text{SiAlON}} + A_{(210)} + A_{(101)}\} \quad (2)$$

where A_{SiAlON} is the total area of the X-ray peaks due to β -SiAlON within the range of $32.0^\circ < 2\theta < 36.5^\circ$. $A_{(210)}$ and $A_{(101)}$ are the areas of the X-ray peaks for (210) and (101) of β - Si_3N_4 , respectively. The microstructure of the whisker/matrix interface in Si_3N_4 whisker/ Al_2O_3 composites was observed by TEM. At the same time, quantitative elemental analysis of the interface was also carried out by EDS.

3. Results and discussion

3.1 Chemical reaction at Si_3N_4 whisker/ Al_2O_3 interface

The XRD profiles within the range of $32.0^\circ < 2\theta < 36.5^\circ$ are shown in Fig. 1. An interfacial chemical reaction was recognized by identifying the XRD profiles of β -SiAlON

Fig. 1. X-ray diffraction profiles of Si_3N_4 whisker/ Al_2O_3 composites under various hot-pressing conditions.

Fig. 2. Phase relation in the system Si_3N_4 - SiO_2 - AlN - Al_2O_3 at 1760°C .¹⁹⁾

($\text{Si}_{6-z}\text{Al}_z\text{O}_2\text{N}_{8-z}$) and X-phase ($\text{Si}_6\text{Al}_6\text{O}_9\text{N}_8$).^{15),19)} Phase relation reported by Jack¹⁹⁾ in the system Si_3N_4 - SiO_2 - AlN - Al_2O_3 at 1760°C is shown in Fig. 2. The coexistence of β -SiAlON with X-phase was reasonably explained by the phase relation, since the chemical composition at the whisker/matrix interfaces was on the Si_3N_4 - Al_2O_3 diagonal line of the map and the line passed across the area where β -SiAlON could stably coexist with X-phase. Moreover, the experimental result obtained in this study coincided with the results reported by Koyama and Kamigaito,¹⁵⁾ Jack,¹⁹⁾ Mitomo et al.²¹⁾ and Dew and Lewis,²²⁾ which revealed that β -SiAlON coexisted with X-phase as a reaction product after sintering of Si_3N_4 - Al_2O_3 mixed powder. The R_{SiAlON} values of each composite, which corresponded to the degree of the reaction, were obtained and are shown in Table 1. The intensity of the β -SiAlON peaks increased with increasing maximum temperature and duration of hot-pressing owing to the progress in the chemical reaction. Mitomo et al.²¹⁾ reported the correlation between lattice parameters and Z -values of β -SiAlON. Then, the X-ray diffracted angles due to β -SiAlON crystal could be calculated as a function of the Z -value and are indicated by arrows in Fig. 1. Comparing

Table 1. The Relationship between Hot-Pressing Conditions and the Volume Fraction of β -SiAlON in Si_3N_4 Whiskers

Maximum temperature -Duration	β -SiAlON ratio in Si_3N_4 whiskers
1550°C-1hr.	0.20
1575°C-1hr.	0.28
1600°C-1hr.	0.30
1650°C-1hr.	0.33
1650°C-3hr.	0.49
1675°C-1hr.	0.69
1675°C-3hr.	0.82
1700°C-1hr.	0.85

the measured values with the calculated ones, it was found that β -SiAlON with a Z -value of approximately 4 was produced during the interfacial chemical reaction. To explain the interfacial reaction in detail, a model of Si_3N_4 whisker/ Al_2O_3 interface during hot-pressing was proposed as shown in Fig. 3. β -SiAlON coexisted with O' -phase at the first stage of the interfacial chemical reaction, since the degree of substitution of Al^{3+} and O^{2-} to Si^{4+} and N^{3-} sites in β - Si_3N_4 was small ($Z < 1$). Thus, the microstructure of the interface was illustrated schematically in Fig. 3(a), in the case of Z -value lower than unity. At this stage, O' -phase

Fig. 3. Schematic illustrations of a structural model of the interface.

(a) The first stage of interfacial chemical reaction.
(b) The final stage of interfacial chemical reaction.

played the role of the medium for the substitution²³⁾ of Al^{3+} and O^{2-} to Si^{4+} and N^{3-} . When the Z -value became high, β -SiAlON and X-phase should coexist at the interface according to Fig. 2. At the final stage of the reaction, the Z -value was approximately 4.2 which was the maximum Z -value, and O' -phase changed to X-phase by the substitutional solid solution of Al^{3+} and O^{2-} . Then, X-phase played the role of the medium for ion substitution. The interfacial phases are illustrated in Fig. 3(b) when the Z -value ranged from 3 to 4. The interfacial structure shown in Fig. 3(a) disappeared immediately and changed to the one shown in Fig. 3(b), when the solid solution of Al^{3+} and O^{2-} into β - Si_3N_4 through O' -phase and into O' -phase occurred much faster than that into β - Si_3N_4 through β -SiAlON. Then, we could explain the reason why β -SiAlON with a Z -value of approximately 4 coexists with X-phase using the model.

3.2 Si_3N_4 whisker/ Al_2O_3 interfacial structure observed by TEM

Si_3N_4 whiskers in the composites hot-pressed at 1550 and 1700°C are shown in Fig. 4 together with the concentrations of Si and Al around the whisker as analyzed by EDS. The intensity of Si $K\alpha$ rose steeply and that of Al $K\alpha$ fell sharply at the interfaces in the composites hot-pressed below 1600°C. In contrast, the intensity of Si $K\alpha$ increased gradually and that of Al $K\alpha$ decreased at the interface and/or interfacial layer in the composite hot-pressed above 1600°C. Hence, the interfacial layer must be constituted of β -SiAlON and/or X-phase. The interfacial reaction model suggested the coexistence of β -SiAlON and X-phase at the interface as illustrated in Fig. 3(b); however, no double-layer-structure could be found at the interface. As reported by Mitomo et al.²¹⁾ and Dew and Lewis,²²⁾ X-phase liquefies above 1600°C. Thus, we consider the possibility that the melt of X-phase penetrated the Al_2O_3 grain boundaries as well as the whisker/matrix interface. Then, the interfacial layer seemed to be chiefly composed of β -SiAlON with a small amount of X-phase.

3.3 Interfacial chemical reaction and densification

The relationship between R_{SiAlON} and the apparent density of the composites is shown in Fig. 5. Apparent density

Fig. 4. TEM observation and the EDS analysis around a whisker in Si_3N_4 whisker/ Al_2O_3 .

Fig. 5. Apparent density of Si_3N_4 whisker/ Al_2O_3 composites as a function of the volume fraction of β -SiAlON in Si_3N_4 whiskers. Solid lines are obtained from theoretical calculation.

decreased with increasing R_{SiAlON} despite an increase in hot-pressing temperature. The change in the theoretical density

$$d_{\text{comp}} = \frac{\{3.98(1 - V_f) + 3.188 V_f\} \left\{ 3.188(1 - x) + \frac{280 - 42z}{280 + z} d_z x \right\}}{3.188 \{1 - (1 - V_f)x\} + \frac{280 - 42z}{280 + z} (1 - V_f) d_z x - \frac{3.188}{3.98} \frac{43z}{280 + z} V_f d_z x} \quad (3)$$

where x and z represent R_{SiAlON} and Z -value, respectively. V_f indicates the volume fraction of whiskers, and should be substituted for 0.1 in this case. The values 3.98 and 3.188 mean the theoretical densities of Al_2O_3 and Si_3N_4 , respectively. The value of $(280 + z)$ corresponds to the molecular weight of β -SiAlON ($\text{Si}_{6-z}\text{Al}_2\text{O}_z\text{N}_{8-z}$), then $(280 - 42z / 280 + z)$ means the percentage weight of Si and N in SiAlON. $(43z / 280 + z)$ means the percentage weight of Al and O in β -SiAlON. d_z is the theoretical density of β -SiAlON as a function of Z -value. Mitomo et al.²¹⁾ reported the theoretical density of β -SiAlON as a function of Z -value as follows,

$d_0 = 3.188$, $d_1 = 3.151$, $d_2 = 3.112$, $d_3 = 3.070$, $d_4 = 3.045$ (units: g/cm^3).

The theoretical densities of the composite derived by Eq. (3) appear as solid lines in Fig. 5 together with the experimental densities. The higher Z -value was, the lower the density was. Moreover, the density decreased with increasing R_{SiAlON} even in fully densified composites. Comparing the experimental density with the calculated ones, the composites hot-pressed above 1600°C , in which the Z -value was nearly equal to four, were sufficiently densified in spite of low apparent density. In contrast, the composites hot-pressed below 1600°C , whose R_{SiAlON} was below 0.3, was not enough for densification. On the other hand, hot-pressing at higher temperature (above 1650°C) should be carried out to obtain dense bodies in SiC whisker/ Al_2O_3 composite in which the volume fraction and size of SiC whiskers were the same as those of the Si_3N_4 whiskers. Tamari et al.¹⁴⁾ also reported that sufficient densification of an Si_3N_4 whisker/ Al_2O_3 composite required lower hot-pressing temperature than that of an SiC whisker/ Al_2O_3 composite. We considered that this improvement in the sinterability was caused by the melting of X-phase above 1600°C ,^{21), 22)} which promoted matrix grain rearrangement and mass transportation.

3.4 Interfacial reaction and mechanical properties

The fracture toughness and the work-of-fracture of Si_3N_4 whisker/ Al_2O_3 composites are shown in Figs. 6 and 7 as a

of the composite with the amount of β -SiAlON must be considered in the evaluation of the degree of densification. Theoretical density of β -SiAlON is smaller than that of β - Si_3N_4 owing to lattice expansion.¹⁹⁾ In addition, X-phase also affects the change in the apparent density of the composite. However, it was difficult to take account of the effect of X-phase on the density, because X-phase exists not only as a crystal phase but also as a glass phase. Thus, we set up the following three premises to simplify the calculation of the theoretical density.

[Premise 1] Although X-phase coexists with β -SiAlON as a reaction product, the amount of X-phase is not taken into account. Then, the amount of β -SiAlON is the only factor considered in calculating the theoretical density.

[Premise 2] The value of R_{SiAlON} , which indicates the progress of the interfacial chemical reaction, corresponds to the volume fraction of β -SiAlON in Si_3N_4 whiskers.

[Premise 3] No decomposition occurs during the interfacial chemical reaction in the composite. Hence, the mass of the composite remains the same before and after sintering.

We derived an equation for evaluating the theoretical density of the composite (d_{comp}) as follows,

Fig. 6. Fracture toughness of Si_3N_4 whisker/ Al_2O_3 composites as a function of the volume fraction of β -SiAlON in Si_3N_4 whiskers.

Fig. 7. Work-of-fracture of Si_3N_4 whisker/ Al_2O_3 composites as a function of the volume fraction of β -SiAlON in Si_3N_4 whiskers.

Fig. 8. SEM images of the fractured surfaces of Si_3N_4 whisker/ Al_2O_3 composites with various volume fractions of β -SiAlON in Si_3N_4 whiskers.

function of R_{SiAlON} , respectively. In Fig. 6, the fracture toughness increased with increasing R_{SiAlON} up to 0.3, and then decreased slightly when R_{SiAlON} exceeded 0.3. The work-of-fracture showed the same behavior against R_{SiAlON} as shown in Fig. 7. The marked fracture toughness and work-of-fracture increment within the range of $0 < R_{\text{SiAlON}} < 0.3$ was caused by the progress of densification in the composite. The inflection point of fracture toughness and work-of-fracture at R_{SiAlON} of 0.3 corresponded to the melting of X-phase during hot-pressing. The composites obtained when R_{SiAlON} exceeded 0.3 were hot-pressed above 1600°C . The fractured surfaces of the chevron notched specimens were observed by SEM and are shown in Fig. 8. Pulled-out whiskers from the fractured surface are hardly observed regardless of the value of R_{SiAlON} . Thus, whisker bridging seemed not to contribute so much to the toughening of the Si_3N_4 whisker/ Al_2O_3 composite compared with the case of SiC whisker/ Al_2O_3 composite.⁶⁾ This may be caused by strong bonding of the whisker/matrix interface owing to the interfacial chemical reaction. Then, the inflection of fracture toughness and work-of-fracture at R_{SiAlON} of 0.3 and the slight decrease in both when R_{SiAlON} exceeded 0.3 were considered to be brought about by the vitrification of X-phase, which increased interfacial bonding strength. Moreover, the degradation of whisker strength due to chemical reaction should be considered as another reason for the decrease of fracture toughness and work-of-fracture.

In Fig. 9, the elastic modulus of the composite increased with increasing R_{SiAlON} up to 0.3, and then decreased gradually when R_{SiAlON} exceeded 0.3. Thus, the marked increase in elastic modulus within the range of $0 < R_{\text{SiAlON}} < 0.3$ was due to the densification. The elastic modulus was small in the range of $0.3 < R_{\text{SiAlON}}$, since the amount of β -SiAlON whose elastic modulus was smaller than that of β - Si_3N_4 ²⁴⁾ increased owing to the progress of the interfacial chemical reaction.

Fig. 9. Elastic modulus of Si_3N_4 whisker/ Al_2O_3 composites as a function of the volume fraction of β -SiAlON in Si_3N_4 whiskers.

Fig. 10. Four-point bending strength of Si_3N_4 whisker/ Al_2O_3 composites as a function of the volume fraction of β -SiAlON in Si_3N_4 whiskers.

As shown in Fig. 10, the bending strength of the composite decreased with increasing R_{SiAlON} up to 0.3, in comparison with the case of monolithic Al_2O_3 . The bending strength became constant when R_{SiAlON} exceeded 0.3. The decrease of the bending strength seemed to be due to the degradation of whisker strength and the increment of vitrified X-phase. These were gradually brought about by the progress of the interfacial chemical reaction. Then, the constant value of the bending strength was considered to be due to the strength of the matrix in which X-phase diffused into Al_2O_3 grain boundaries.

4. Conclusions

The correlation between the interfacial chemical reaction and the mechanical properties of Si_3N_4 whisker/ Al_2O_3 composites was investigated. As a result, the following conclusions were obtained.

(1) It was found that both X-phase and β -SiAlON with Z-value of approximately 4 were produced by the interfacial chemical reaction. A model of reacted whisker/matrix interface was proposed which explained well the results obtained in this study.

(2) The densification of the composite was promoted by the liquefaction of X-phase during hot-pressing above 1600°C .

(3) There was a large difference between the composites hot-pressed below 1600°C and those hot-pressed above 1600°C in terms of fracture toughness and work-of-fracture. The decrease of fracture toughness and work-of-fracture was brought about by the degradation of whisker strength and the vitrification of X-phase, which increased interfacial bonding strength.

(4) The decrease of the bending strength was due to the degradation of whisker strength and the increment of vitrified X-phase. The bending strength became constant in

the composite hot-pressed above 1600°C, which corresponded to the strength of the matrix in which vitrified X-phase diffused into grain boundaries.

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