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Properties of novel silicon nitride-based materials

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Abstract Our research on the mechanical and thermal properties of magnesium silicon nitride ($MgSiN_2$)-silicon nitride (Si_3N_4) composite specimens has been reviewed in this paper. The specimen was fabricated by hot-pressing the compressed powder at a temperature between 1550°C and 1700°C for 90 min under a pressure of 75 MPa in a nitrogen atmosphere, using 1 mol% ytterbium oxide (Yb_2O_3) addition as a sintering aid. Mechanical and thermal properties of $MgSiN_2$ specimen without Si_3N_4 addition were as follows: Vickers hardness, 18.3 GPa; flexural strength, 371 MPa; fracture toughness, 2.2 MPa·$m^{1/2}$; and thermal conductivity, 22.7 W·m^{-1}·K^{-1}. In order to improve these properties, $MgSiN_2$ composite was fabricated with the addition of 0–89 mol% Si_3N_4. The fracture toughness of $MgSiN_2$ specimen could be enhanced by the addition of Si_3N_4, e.g., 6.6 MPa·$m^{1/2}$ (4 mol% Si_3N_4 addition) and 8.7 MPa·$m^{1/2}$ (49 mol% Si_3N_4 addition). An increase in fracture toughness of $MgSiN_2$-Si_3N_4 specimen was attributed to the elongation of Si_3N_4 grains. Moreover, the thermal conductivity of $MgSiN_2$-Si_3N_4 specimen increased to 32.7 W·m^{-1}·K^{-1} for 29 mol% Si_3N_4 addition.

Key words magnesium silicon nitride • silicon nitride • composite • densification • microstructure • mechanical properties • thermal properties

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Introduction

Many researchers pay much attention to the new ceramics with excellent mechanical, electrical, thermal and optical properties. Some of the non-oxide ceramics have much possibility to fulfill these requirements rather than oxide ceramics, chiefly due to strong covalent bonding. On the basis of such respect, our research has been focused on the development of dense magnesium silicon nitride ($MgSiN_2$) specimen, partly because this compound is regarded as the replacement of $2Al^{3+}$ in the AlN by Mg^{2+}/Si^{4+}, and partly because the magnesium and silicon recourses are abundant in the earth crust.

Slack [10] is a first researcher who predicted the high thermal conductivity of $MgSiN_2$. On the basis of his prediction, Groen *et al.* [3] and Hintzen *et al.* [5] fabricated the dense $MgSiN_2$ ceramics by pressureless-sintering and hot-pressing techniques, respectively; however, the thermal conductivities did not exceed 20 W·m^{-1}·K^{-1} in both cases.

As these cases indicate, the fabrication of dense $MgSiN_2$ ceramic started with the attention to develop a novel inorganic material having high thermal conductivity. Such properties may not be obtained until dense $MgSiN_2$ ceramic with controlled microstructure is fabricated using an advanced sintering technique. On the other hand, Hayashi *et al.* [4] investigated the effect of $MgSiN_2$ addition on the densification of silicon nitride (Si_3N_4) powder, and found that the thermal conductivity of Si_3N_4 ceramic with $MgSiN_2$ addition (sintering aid; Yb_2O_3) is as high as 142 W·m^{-1}·K^{-1}. This

fact suggests that the addition of Si_3N_4 to the $MgSiN_2$ ceramic must also contribute to enhancing the thermal conductivity as well as the fracture toughness of $MgSiN_2$, because the monolithic Si_3N_4 ceramic possesses excellent mechanical and thermal properties. On the basis of such background, this paper reviews our research on the fabrication of $MgSiN_2$-Si_3N_4 composite specimens with controlled microstructure, and mechanical/thermal properties of the resulting specimens.

Experimental procedure

The starting $MgSiN_2$ powder was prepared by the nitridation of magnesium silicide (Mg_2Si; Mg/Si = 2.0) powder at 1350°C for 10 min in a nitrogen atmosphere. The resulting $MgSiN_2$ powder was mixed with 0 to 89 mol% Si_3N_4 (SN-E10; Ube Industries, Ube; α/β ratio > 95%, oxygen content < 2.0%, carbon content < 0.2%) and 1 mol% Yb_2O_3 (99.99% purity, Wako Pure Chemical, Osaka) in the presence of *n*-hexane. After drying, approximately 1.5 g of the mixed powder was uniaxially pressed at 30 MPa to fabricate a compact with a diameter of 20 mm and a thickness of 2 mm. Each compact was hot-pressed at a temperature between 1550°C and 1700°C for 90 min in a nitrogen atmosphere under a pressure of 75 MPa.

The relative density of hot-pressed specimen was calculated using the bulk and true densities; the bulk density was measured using the Archimedes method, while the true density was determined picnometrically at 25.0°C, after pulverizing the hot-pressed specimen. Crystalline phases in the hot-pressed specimen were examined using an X-ray diffractometer (XRD) (Model RINT2000, Rigaku, Tokyo) with monochromatic CuK_α radiation at 40 kV and 40 mA. The magnesium and silicon contents in the hot-pressed specimen were determined using an energy dispersive X-ray spectroscope (EDX; Model EMAX5770, Horiba, Kyoto), whereas the oxygen and nitrogen contents were examined using an N/O determinator (Model TC-436, Leco, St. Joseph, MI, USA).

The microstructure of hot-pressed specimen was investigated using a field-emission scanning electron microscope (FE-SEM; Model S-4500, Hitachi, Tokyo). The Vickers hardness (H_V) was measured using an indentation load of 9.81 N for 15 s (Model MVK-E, Akashi, Tokyo). Moreover, the fracture toughness (K_{IC}) of a specimen with sizes of 15 × 2.5 × 3 mm^3 was measured using a single-edge notched beam technique; the specimen was fabricated by cutting the sintered specimen. The thermal diffusivity was measured at room temperature, using a laser-flash technique (Model TC-7000, Shinku-Riko, Tokyo). On the basis of thermal diffusivity data, the thermal conductivity was calculated using specific heats of $MgSiN_2$ (61.71 $J \cdot mol^{-1} \cdot K^{-1}$) [1, 2] and β-$Si_3N_4$ (90.68 $J \cdot mol^{-1} \cdot K^{-1}$) [1].

Results and discussion

We first examined the properties of high-purity $MgSiN_2$ powder prepared by the reaction of Mg_2Si with nitrogen. Typical density and specific surface area of the $MgSiN_2$ powder were 3.102 $g \cdot cm^{-3}$ and 16.8 $m^2 \cdot g^{-1}$, respectively. The primary particle size, calculated on the basis of these data, was 0.12 μm, while the crystallite size calculated on the basis of the broadening of XRD reflection was 0.042 μm (42 nm). The chemical composition of this $MgSiN_2$ powder was examined [14], together with the data on carbothermal reduction of magnesium metasilicate ($MgSiO_3$) [15] and solid-state reaction of Mg_3N_2 with Si_3N_4 [3]. The magnesium and silicon contents were in accordance with those of theoretical contents, independent of the preparation technique. On the other hand, the oxygen contents in the $MgSiN_2$ powders were varied, according to the preparation technique: 0.61% (present technique; direct nitridation) < 2.54% (carbothermal reduction [15]) < 3.7% (solid-state reaction [3]). As the above data indicate, the amount of oxygen in the present powder is comparatively low. The contamination of oxygen seems to be minimized by the nitridation of $MgSiN_2$ from Mg_2Si, except for the case that the small amount of oxygen is inevitably included in the starting magnesium and silicon powders used for the preparation of Mg_2Si.

By making use of this $MgSiN_2$ powder, we examined the fabrication conditions of dense $MgSiN_2$ ceramic, using Yb_2O_3 as a sintering aid. This Yb_2O_3 was selected for the sintering aid, because the thermal conductivity of $MgSiN_2$ ceramic with 1 mass% of Yb_2O_3 addition showed a maximum (26.6 $W \cdot m^{-1} \cdot K^{-1}$) among the rare-earth oxides (Y_2O_3, La_2O_3, Nd_2O_3, Sm_2O_3, Gd_2O_3, Er_2O_3 and Yb_2O_3) examined previously [12].

The relative density of $MgSiN_2$ specimen hot-pressed at 1550°C for 90 min attained 98.0%. Mechanical properties of this ceramic were as follows: Vickers hardness, 18.3 GPa; flexural strength, 371 MPa; and fracture toughness, 2.2 $MPa \cdot m^{1/2}$.

These mechanical properties are similar to those reported by Groen *et al*. [3], e.g., (i) Vickers hardness of 14–16 GPa, (ii) flexural strength of 230–280 MPa, and (iii) fracture toughness of 3.1–4.4 $MPa \cdot m^{1/2}$. Relating to the mechanical properties of Si_3N_4 ceramic or a typical material for engine components, Vickers hardness is 17.7–20 GPa, whereas the flexural strength and fracture toughness are 980 MPa and 7–8 $MPa \cdot m^{1/2}$, respectively [13]. Thus Vickers hardness is almost comparable to that of the Si_3N_4 ceramic, but both flexural strength and fracture toughness are somewhat lower than the case of Si_3N_4 ceramic.

On the basis of research by Hayashi *et al*. [4], who succeeded in the enhancement of thermal conductivity of Si_3N_4 ceramic to 142 $W \cdot m^{-1} \cdot K^{-1}$ owing to the addition of $MgSiN_2$, we examined the fabrication conditions of dense $MgSiN_2$ specimens with Si_3N_4 addition.

As a typical case, the effect of hot-pressing temperature on the relative density of $MgSiN_2$ specimen with 4 mol% Si_3N_4 addition is shown in Fig. 1 [11]. Although the relative density of this specimen was 94.3% at 1550°C, it increased to 98.6% at 1600°C; on further increase in hot-pressing temperature, however, the relative density gradually decreased and became 96.4% at 1700°C.

Typical FE-SEM micrographs of the fracture surfaces of hot-pressed $MgSiN_2$ specimens with 4 mol%

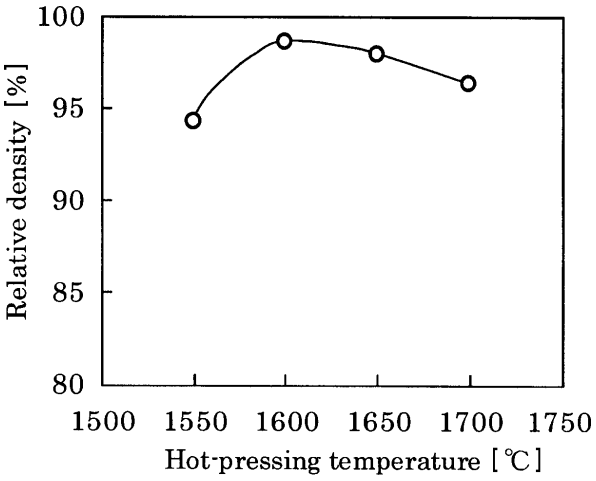

Fig. 1. Effect of hot-pressing temperature on the relative density of $MgSiN_2$ specimen with 4 mol% Si_3N_4 and 1 mol% Yb_2O_3 addition. Note that the hot-pressing time was 90 min.

Si_3N_4 addition are shown in Fig. 2 [11]. The elongated grains were randomly present in the $MgSiN_2$ matrix at 1600°C (Fig. 2a), whereas the elongated grains stuck together to the $MgSiN_2$ matrix at 1700°C (Fig. 2b).

The above elongation and sticking phenomena may be attributed to the accelerated mass transfer in the presence of liquid phase during the hot pressing, i.e., the anisotropic crystal growth (elongation of grains) and chemical reaction at the interfaces (sticking of grains to the matrix). The elongation of grains must be related to the reaction process among $MgSiN_2$, Si_3N_4 and Yb_2O_3 during the hot pressing. Then the crystalline phases of these $MgSiN_2$ specimens were examined using XRD. The crystalline phases at 1600°C were $MgSiN_2$ (JCPDS Card No. 25-530), β-Si_3N_4 (JCPDS Card No. 33-1160), $Yb_2Si_3O_3N_4$ (JCPDS Card No. 32-1423) and $Yb_2Si_3O_5N_2$ (JCPDS Card No. 31-1454). The reaction of $MgSiN_2$ with Yb_2O_3 may, therefore, occur as follows:

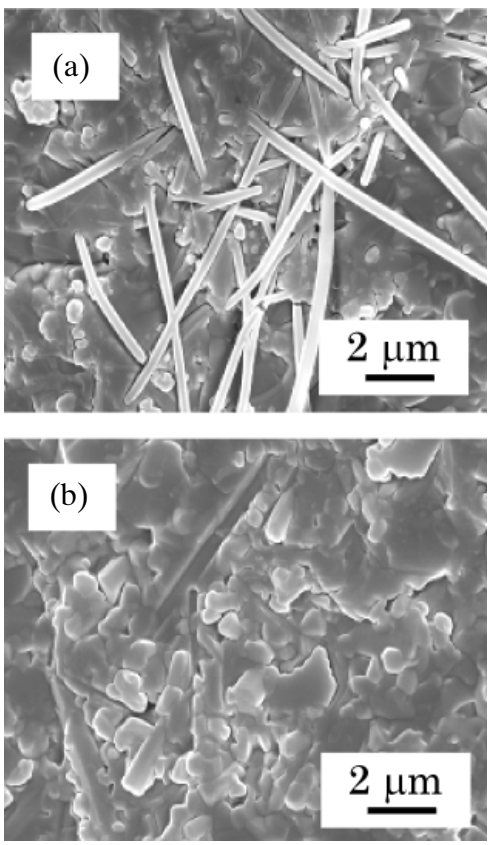

Fig. 2. Typical FE-SEM micrograph of the $MgSiN_2$ specimen with 4 mol% Si_3N_4 and 1 mol% Yb_2O_3 addition hot-pressed at (a) 1600°C and (b) 1700°C for 90 min.

(1) $3MgSiN_2 + Yb_2O_3 \rightarrow Yb_2Si_3O_3N_4 + Mg_3N_2$

(2) $3MgSiN_2 + 2Yb_2O_3 \rightarrow Yb_2Si_3O_5N_2 + 2YbN + Mg_3N_2 + ½O_2$

Although Mg_3N_2 was not detected by XRD, it seems to partly be decomposed and/or evaporated during the hot pressing. In addition to these solid-state reactions, Si_3N_4 may react with Yb_2O_3 to form $Yb_2Si_3O_3N_4$ and $Yb_2Si_3O_5N_2$:

(3) $Si_3N_4 + Yb_2O_3 \rightarrow Yb_2Si_3O_3N_4$

(4) $3Si_3N_4 + 5Yb_2O_3 \rightarrow 3Yb_2Si_3O_5N_2 + 4YbN + N_2$

The densification of $MgSiN_2$ specimen with Si_3N_4 and Yb_2O_3 addition seems to occur along with the formation of liquid phase. With respect to the liquid composition, Inomata *et al.* [6] pointed out that an eutectic liquid in the Si_3N_4-$MgSiN_2$ system forms at approximately 1520°C. Such liquid phase helps not only the elongation of grains due to transformation of the α- to β-phase of Si_3N_4 [8, 9] but also the densification due to rearrangement of the grains. The relative density of $MgSiN_2$ specimen with 4 mol% Si_3N_4 addition decreases with hot-pressing temperature above 1600°C, which suggests that the thermal decomposition may proceed from surfaces to the inside of $MgSiN_2$ specimen during the hot pressing.

Vickers hardness of the $MgSiN_2$ specimen with 4 mol% Si_3N_4 addition was examined, as a function of the hot-pressing temperature. Although the Vickers hardness of the $MgSiN_2$ specimen with 4 mol% Si_3N_4 addition was 18.2 GPa at 1550°C, no appreciable changes in Vickers hardness were observed, regardless of an increase in hot-pressing temperature from 1650°C up to 1700°C.

Effect of the Si_3N_4 addition on the fracture toughness of $MgSiN_2$ specimen is shown in Fig. 3, as a function of the hot-pressing temperature [11]. On the other hand, the fracture toughness of $MgSiN_2$ specimen with 4 mol% Si_3N_4 addition increased to 6.6 $MPa \cdot m^{1/2}$ with hot-pressing temperature up to 1600°C. On further increases in hot-pressing temperature, however, the fracture toughness was slightly reduced down to 5.1–5.5 $MPa \cdot m^{1/2}$.

Fracture toughness of the $MgSiN_2$ specimens with 4 mol% Si_3N_4 addition was the highest at the hot-pressing temperature of 1600°C, which may be related to the elongation of Si_3N_4 grains due to α- to β-phase transformation. The formation of elongated grains contributes to de-bonding during the crack propagation, thereby enhancing the fracture toughness [9]. Decreases

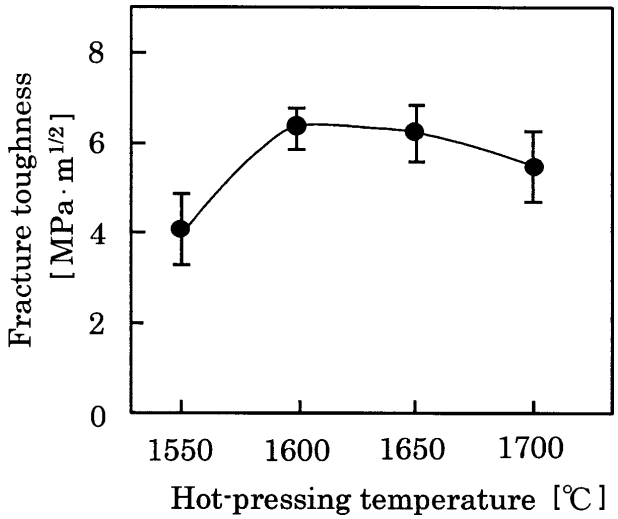

Fig. 3. Changes in fracture toughness of the $MgSiN_2$ specimen containing 4 mol% Si_3N_4 and 1 mol% Yb_2O_3 addition with increasing hot-pressing temperature.

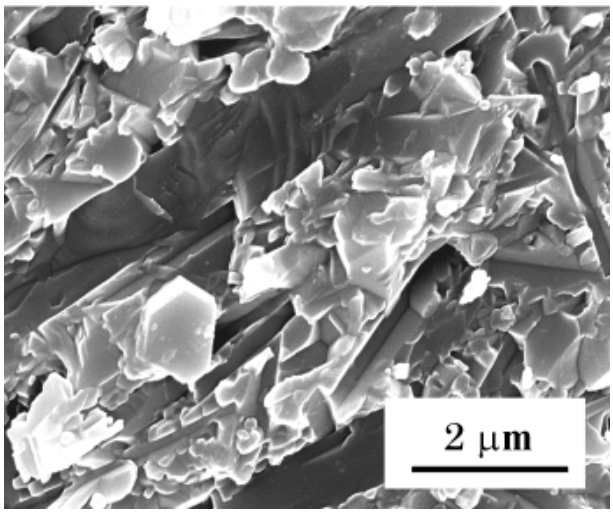

Fig. 5. Typical FE-SEM micrograph of the $MgSiN_2$ specimen with 49 mol% Si_3N_4 and 1 mol% Yb_2O_3 addition hot-pressed at 1600°C for 90 min.

in fracture toughness with a further increase in hot-pressing temperature may be attributed to the changes of elongated shapes into plate-like shapes and to the enhancement of bonding between $MgSiN_2$ matrix and elongated Si_3N_4 grains.

In order to make clear the effect of Si_3N_4 addition, we furthermore investigated the mechanical and thermal properties of $MgSiN_2$ specimens with Si_3N_4 addition to 89 mol%. Figure 4 shows the changes in relative density of the hot-pressed $MgSiN_2$ specimen with increasing amount of Si_3N_4 [7]. Although the relative density of $MgSiN_2$ specimen without Si_3N_4 addition was 98.0%, it was reduced down to 94% for the case of 49 mol% Si_3N_4. Nearly full density was, however, achieved with a further increase in amount of Si_3N_4 to 69 mol% or more. Although the relative density showed a minimum for the case of 49 mol% Si_3N_4, it should be noted that the relative densities always exceed 94%.

Figure 5 shows a typical FE-SEM micrograph of the hot-pressed $MgSiN_2$ specimen with 49 mol% Si_3N_4 addition [7]. The hot-pressed $MgSiN_2$ specimen was composed of the polyhedral grains with sizes of approximately 1 μm and elongated grains.

The hot-pressed $MgSiN_2$ specimen without Si_3N_4 addition was composed of the grains with sizes of 1 μm. The elongated grains formed by the incorporation of Si_3N_4 are assumed to be Si_3N_4, because they did not exist until the $MgSiN_2$ specimen was hot-pressed by the incorporation of Si_3N_4. These elongated grains seem to be formed in the presence of liquid phase during the hot pressing.

Figure 6 shows the changes in flexural strength of the $MgSiN_2$ specimen with increasing amount of Si_3N_4 [7]. The flexural strength of $MgSiN_2$ specimen without Si_3N_4 addition was 371 MPa. The flexural strength of $MgSiN_2$ specimen increased with increasing amount

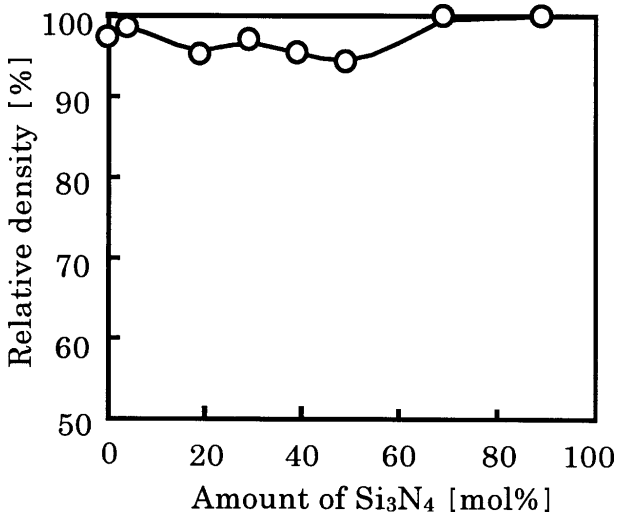

Fig. 4. Changes in relative density of the $MgSiN_2$ specimen with increasing amount of Si_3N_4 addition (the amount of Yb_2O_3: 1 mol%). Note that the specimens were hot-pressed at 1600°C for 90 min.

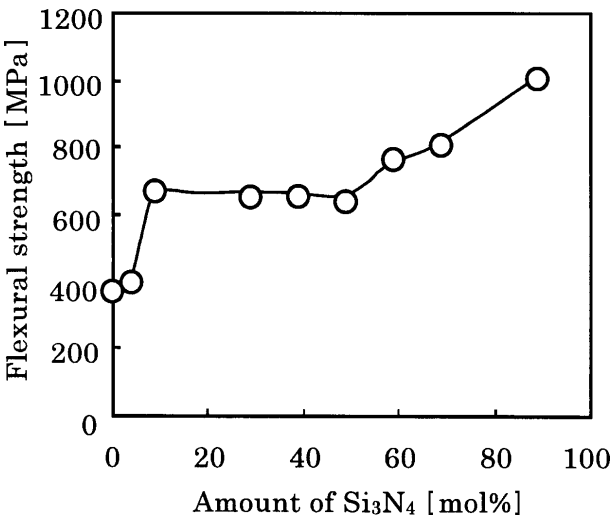

Fig. 6. Changes in flexural strength of the $MgSiN_2$ specimen with increasing amount of Si_3N_4 (the amount of Yb_2O_3: 1 mol%). Note that the specimens were hot-pressed at 1600°C for 90 min.

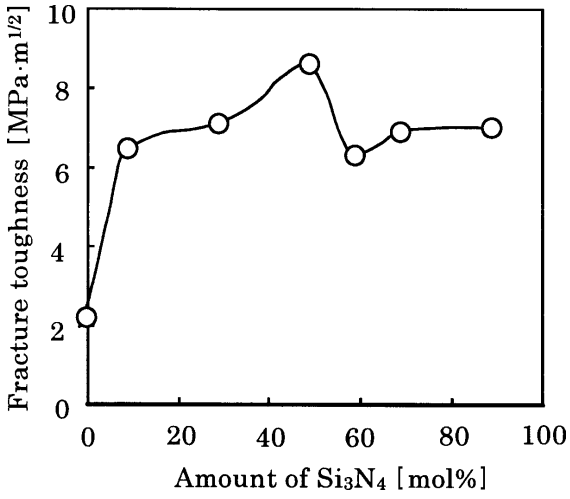

Fig. 7. Changes in fracture toughness of the $MgSiN_2$ specimen with increasing amount of Si_3N_4 (the amount of Yb_2O_3: 1 mol%). Note that the specimens were hot-pressed at 1600°C for 90 min.

of Si_3N_4 and attained 1000 MPa (1 GPa) for the case of 89 mol% Si_3N_4.

An increase in flexural strength of the $MgSiN_2$ specimen with increasing amount of Si_3N_4 to 49 mol% may be related to the elongation of Si_3N_4 grains on the basis of the α- to β-phase transformation. Moreover, a further increase in flexural strength due to the incorporation of 49 mol% or more Si_3N_4 addition seems to be ascribed to the increase in relative density.

Figure 7 shows the changes in fracture toughness of $MgSiN_2$ specimen with increasing amount of Si_3N_4 [7]. The fracture toughness of $MgSiN_2$ specimen without Si_3N_4 addition was 2.2 $MPa{\cdot}m^{1/2}$. The fracture toughness of the $MgSiN_2$ specimen increased with increasing amount of Si_3N_4 and attained 8.7 $MPa{\cdot}m^{1/2}$ for 49 mol% Si_3N_4 addition. On further increase in amount of Si_3N_4, however, the fracture toughness was slightly reduced.

The highest fracture toughness of $MgSiN_2$ specimen with 49 mol% Si_3N_4 addition also seems to be ascribed to the elongation of Si_3N_4 grains. As mentioned before, the formation of elongated grains contributes to debonding during the crack propagation [9], thereby enhancing the fracture toughness. Decreases in fracture toughness with a further increase in amount of Si_3N_4 may be explained in terms of the shortening of elongated grains, due to the reaction of Si_3N_4 with $MgSiN_2$ matrix.

The thermal conductivity of $MgSiN_2$ specimen increased with increasing amount of Si_3N_4 addition and attained 32.7 $W{\cdot}m^{-1}{\cdot}K^{-1}$ for 29 mol% Si_3N_4 addition. On further increase in amount of Si_3N_4, however, the thermal conductivity was slightly reduced.

The thermal conductivity is affected not only by the relative density and grain size but also by the oxygen content. With reference to the oxygen content, the oxygen content of the $MgSiN_2$ specimen with 4 mol% Si_3N_4 addition hot-pressed at 1600°C for 90 min was only 0.4 mass%. Thus the changes in thermal conductivity of the $MgSiN_2$ specimen with Si_3N_4 addition must be chiefly related to the changes in microstructure. Further examination is, however, needed in order to make clear this phenomenon.

Conclusion

Our research on the mechanical and thermal properties of magnesium silicon nitride ($MgSiN_2$)-silicon nitride (Si_3N_4) composite specimens has been reviewed in this paper. $MgSiN_2$ compacts with 1–89 mol% of Si_3N_4 and 1 mol% ytterbium oxide (Yb_2O_3; sintering aid) addition were hot-pressed at a temperature between 1550°C and 1700°C for 90 min in a nitrogen (N_2) atmosphere under the pressure of 75 MPa. The results obtained were summarized as follows:

1. Mechanical and thermal properties of $MgSiN_2$ specimen without Si_3N_4 addition but with 1 mol% Yb_2O_3 addition (sintering aid) hot-pressed at 1550°C for 90 min were as follows: Vickers hardness, 18.3 GPa; flexural strength, 371 MPa; fracture toughness, 2.2 $MPa{\cdot}m^{1/2}$; and thermal conductivity, 22.7 $W{\cdot}m^{-1}{\cdot}K^{-1}$.
2. The fracture toughness of $MgSiN_2$ specimen with 4 mol% of Si_3N_4 and 1 mol% Yb_2O_3 addition hot-pressed at 1600°C for 90 min was 6.6 $MPa{\cdot}m^{1/2}$, which was three times higher than the value of the hot-pressed $MgSiN_2$ specimen without Si_3N_4 addition (2.2 $MPa{\cdot}m^{1/2}$). The maximum fracture toughness of $MgSiN_2$ specimen (8.7 $MPa{\cdot}m^{1/2}$) was obtained by the addition of 49 mol% Si_3N_4. The enhancement of fracture toughness seemed to be attributed to the elongation of Si_3N_4 grains. The thermal conductivity of $MgSiN_2$ specimen increased with increasing amount of Si_3N_4 addition and attained 32.7 $W{\cdot}m^{-1}{\cdot}K^{-1}$ for 29 mol% Si_3N_4 addition.

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