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Processing and mechanical properties of mullite fiber/Fe composite

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The high – speed steel (shorten as HSS) consists of Fe and several kinds of transition metal carbides. The cutting tools or wear – resistant materials made from HSS experience relatively high thermal shock because a coolant such as water or oil is flowed over the surface of heated HSS. The purpose of this research is to increase the hardness, strength, fracture toughness and thermal shock resistance of HSS. A possible strategy is to incorporate a hard ceramic material with high strength in HSS matrix. This paper describes the processing, microstructure and mechanical properties of the oriented unidirectional mullite fiber/HSS composite. The unidirectional mullite fibers of 10 μm diameter were dispersed by the ultrasonic irradiation of 38 kHz in an ethylenglycol suspension containing HSS powder of 11 μm median size. The dried green composites with 4~68 vol % fibers were hot – pressed for 2 h at 1000°C in Ar atmosphere under a pressure of 39 MPa. The higher density was achieved in the composite with a lower content of fibers. The oriented unidirectional fibers were well dispersed in the HSS matrix. The average distance between the center of fibers in the cross section was close to the value calculated from the fiber fraction. No reaction occurred at the interfaces between HSS and mullite fibers in the composites. The composite with 13.6 vol % fibers showed 1000 MPa of four point flexural strength at room temperature. The thermal expansion of composite with heating was influenced by the orientation of mullite fibers.

Key words: High speed steel, Mullite fiber, Composite, Hot – pressing, Microstructure, Strength, Thermal expansion.

Introduction

The high – speed steel (HSS) consisting of Fe and several kinds of transition metal carbides has a high fracture toughness (25~28 MPa · $m^{0.5}$) and high hardness (7.5 GPa in Vickers hardness) [1, 2]. This material is applied to cutting tool or wear – resistant materials. The use of HSS experiences relatively high thermal shock because a coolant such as water or oil is flowed over the surface of heated HSS. To overcome the high thermal shock, a high strength is needed for HSS. The purpose of this research is to increase the hardness, strength, fracture toughness and thermal shock resistance of HSS. A possible strategy is to incorporate a hard ceramic material with high strength in HSS matrix. The strength of the composite depends on the volume fraction and strength of ceramic material incorporated. The fracture toughness ($K_{IC} = (2E\gamma)^{0.5}$) of HSS increases with the addition of ceramic material with a high Young's modulus (E) and by the introduction of fracture energy adsorption mechanisms providing the increase of fracture energy (γ).

The critical temperature difference (ΔTc) to no crack formation for brittle materials is expressed by $\Delta Tc = \sigma(1-\nu)/\alpha E$ where σ is the tensile strength, ν the Poisson ratio, α the thermal expansion coefficient, and E the Young's modulus [3]. It is understood that high σ, low α and low E are the effective factors to increase the thermal shock resistance. A higher Young's modulus and a lower thermal expansion coefficient of ceramic material compared with the properties of HSS provide the opposite influence on the thermal shock resistance of composite. When the product of αE becomes lower in the ceramic/HSS composite, the thermal shock resistance of HSS may be improved under a similar strength. Based on the above scope, the incorporation of mullite long fibers into HSS was planned. This fiber has the high tensile strengths of 1.7~2.2 GPa in addition to the high hardness. The typical values of $\alpha = 5.6 \times 10^{-6}$ K^{-1} and E = 210 GPa for mullite provide the product of $\alpha E = 1.18 \times 10^{-3}$ GPa · K^{-1}. On the other hand, the product of αE for HSS is calculated to be 3.12×10^{-3} GPa · K^{-1} ($\alpha = 13 \times 10^{-6}$ K^{-1}, E = 240 GPa). The above calculation suggests a possibility of increased thermal shock resistance with addition of mullite fiber to HSS. Furthermore, substitution of the typical values for mullite fiber, σ = 1.7~2.2 GPa, ν = 0.27, $\alpha = 5.6 \times 10^{-6}$ K^{-1} and E = 210 GPa into the equation of critical temperature difference leads to ΔTc = 1055~1365 K, supporting the high thermal shock resistance.

This paper describes the processing, microstructure and mechanical properties of the mullite long fibers/HSS composite. The reaction between the fibers and matrix is also an important factor in the processing and discussed in text. Usually, HSS powder is heated above 1500°C to melt and solidified to the desirable shape by

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cooling. However, this processing may not be suitable for the preparation of mullite fibers/HSS composite, because grain growth of mullite in the fibers is accelerated at such a high temperature. The strength of oxide fiber decreases with increasing grain growth. In this study, the composite was processed by hot – pressing at a low temperature of 1000°C.

Experimental Procedure

A high-speed steel powder (Mitsubishi Steel Mfg. Co., Ltd., Japan) has a cumulative particle size distribution of 4.43 μm/10 %, 11.23 μm/50 % and 24.93 μm/90% and a following chemical composition: 81.91 mass % Fe, 0.85 mass% C, 4.03 mass% Cr, 1.94 mass % V, 4.88 mass% Mo, and 6.01 mass% W. The true density measured with the pycnometer using kerosine was 7.931 g/cm^3. The unidirectional mullite fiber of an average diameter of 10 μm (Sumitomo Chemical Co., Ltd., Japan) has a chemical composition of 85 mass% Al_2O_3 and 15 mass% SiO_2 (1700 filament/yarn). The true density of the fiber was 3.226 g/cm^3. The tensile strength and tensile modulus are reported by the supplier to be 2.0 GPa and 210 GPa, respectively at room temperature. The unidirectional mullite fibers were dispersed by the ultrasonic irradiation of 38 kHz in an ethylenglycol suspension containing 45 vol% HSS powder in a stainless steel box. The fiber fraction of the composite was controlled in the range of 4.1~68.3 vol %. The dried green composite was heated to 900°C at 10°C/min and pressed by carbon die under a pressure of 39 MPa, and further heated to 1000°C to sinter for 2 h in an Ar atmosphere (FVH-5 type, Fuji Denpa Kogyo Co., Japan). The density of hot – pressed composite was measured in kerosine by the Archimedes method. The microsturecture of the hot-pressed composite was obserbed by scanning electron microscopy (SM-300, Topcon Co., Japan). The distance between the center of the fibers in the cross section was measured by the image analyzer (Luzex-FS, Nireco Co., Japan). The flexural strength of test specimens with sizes of 38 mm length, 4 mm width and 3 mm thickness was measured at room temperature by the four-point flexural method, over spans of 30 mm (lower span) and 10 mm (upper span), at a crosshead speed of 0.5 mm/min (Model UTM-1-5000 BE, Toyo Baldwin Co., Japan). The thermal expansion of the composite was measured by thermal mechanical analyzer in Ar atmosphere (Thermoplus Series 8310, Rigaku Co., Japan).

Results and Discussion

Processing of mullite fiber/HSS composite

Figure 1 shows the crystalline phases produced in the mullite fibers heated for 2 h at 1000°~1300°C. As – recieved fiber with 85 mass% Al_2O_3 and 15 mass% SiO_2 contained γ-alumina phase, which was stable with

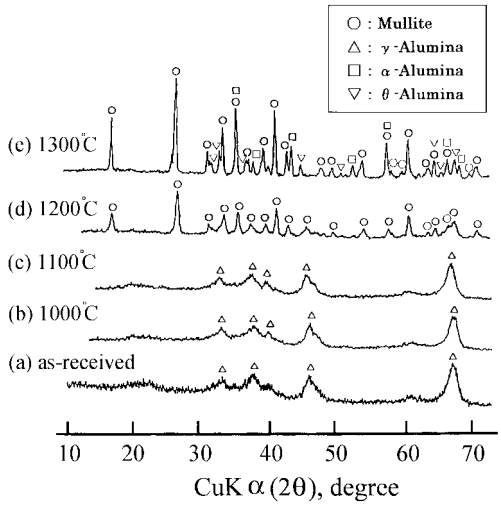

Fig. 1. X-ray diffraction patterns for (a) as – received mullite fiber, and for the fibers heat – treated at (b) 1000°, (c) 1100°, (d) 1200° and (e) 1300°C for 2 h in air.

heating to 1100°C. In the heating at 1200°C, γ-alumina phase changed to mullite. The heating at 1300°C produced mullite, θ- and α-alumina. The formation of mullite and α-alumina was in accordance with the phase diagram of the SiO_2-Al_2O_3 system [4]. The phase change in the fiber with heating is accompanied by the volume change due to the different density. In addition, the heating at a high temperature accelerates the grain growth of the crystalline phases in the fiber. The above two phenomena with heating result in the decreased strength of fiber because of the increase of the flaw size. Therefore, the processing of the mullite fiber/HSS composite may be designed below 1100°C.

Figure 2 shows the relation between mullite fiber content and relative density of the composite hot – pressed at 1000°C under a pressure of 39 MPa. The higher density was achieved in the composite with a lower content of fiber, indicating that the fiber plays as an obstacle for the mass transport of HSS during

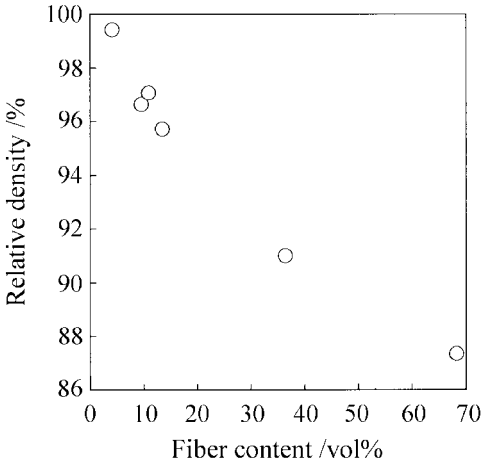

Fig. 2. Relative density of the composites hot – pressed at 1000°C and mullite fiber content.

heating. The density of composite may be influenced by the degree of plastic deformation of HSS, dispersibility of the fibers and the pressure applied during the hot – pressing. Increase of the plastic deformation of HSS with increasing hot – pressing temperature may be an essential condition for the penetration into the spaces between fibers. However, the hot – pressing temperature is restricted below 1100°C owing to the phase change and grain growth in the fiber (Fig. 1). Loading of a certain pressure to the heated HSS enables its plastic deformation to densify the composite. Increase of the dispersibility of fibers is also an effective factor to the transport of HSS. At this moment, an increase of pressure in the hot-pressing at 1000°~1100°C may be the improved processing for the HSS penetration in the composite.

Figure 3 shows the cross section of the composites with (a) 4.6, (b) 13.6 and (c) 36.6 vol% mullite fibers. The oriented unidirectional mullite fibers were well dispersed in the HSS matrix. The increased fiber fraction caused the contact between fibers. The dispersibility of mullite fibers was quantitatively evaluated. Figure 4 shows the configuration model of fibers in the HSS matrix. The area (S_1 μm^2) of triangle ABC is expressed by Eq. (1) as a function of distance (R) between center,

$$S_2 = R \cdot \frac{\sqrt{3}R}{2} \cdot \frac{1}{2} = \frac{\sqrt{3}R^2}{4} \tag{1}$$

The area (S_2 μm^2) of the mullite fibers with a radius r, included in the triangle ABC is expressed by Eq. (2),

$$S_2 = \frac{\pi r^2}{2} \tag{2}$$

For the volume fraction V of fiber in the composite, Eq. (3) is derived from Eqs. (1) and (2),

$$S_1 V = S_2 \tag{3}$$

Figure 4 represents the calculated relation for the fiber fraction (V) and distance (R) between the center of fibers with 10 μm diameter. R increases in proportion to $V^{-1/2}$.

Figure 5 shows the distribution of distance between the center of fibers in the cross section of the composite with 13.6 vol% fibers. The sharp distribution indicates a uniform dispersion of fibers. The peak of the distribution was measured at around 24~26 μm of the distance between the center. The distance calculated by Eqs. (1)~(3) resulted in 25.8 μm. Both the values were very close, indicating the high dispersibility of mullite fibers in the HSS matrix.

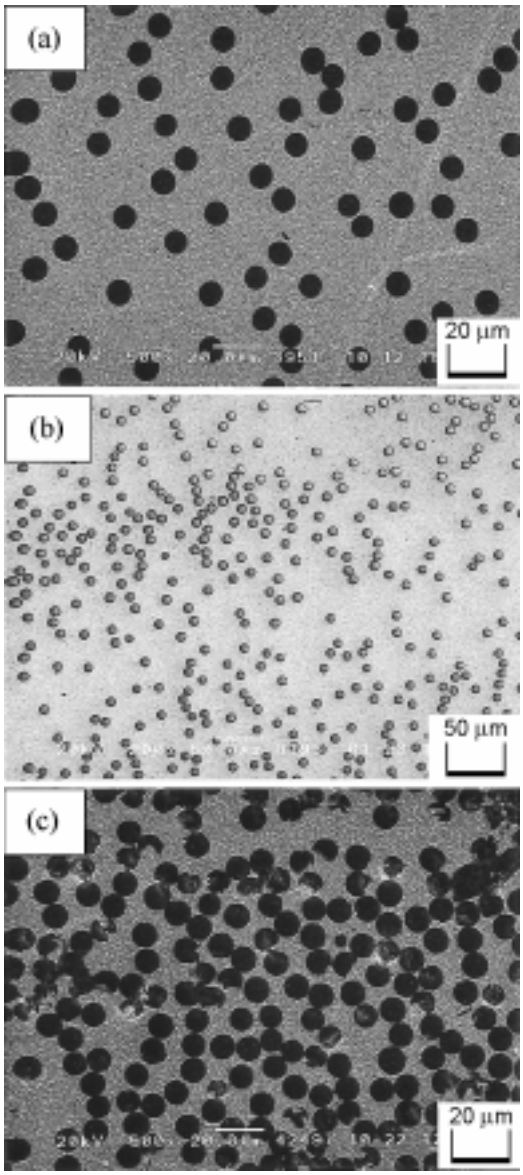

Fig. 3. Cross section of the composites with (a) 4.6, (b) 13.6 and (c) 36.6 vol% mullite fibers.

Chemical reaction, strength and thermal expansion

Figure 6 shows the standard free energy change for the reaction of the mullite fibers – Fe system [5]. The calculation resulted in plus in the temperature range

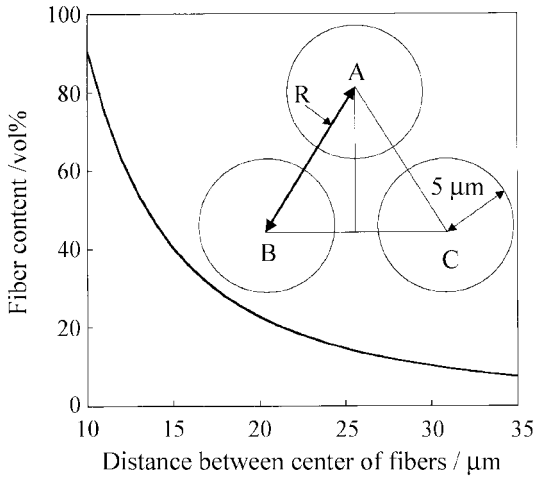

Fig. 4. Calculated relation for the fiber fraction and distance between the center of fibers with 10 μm diameter.

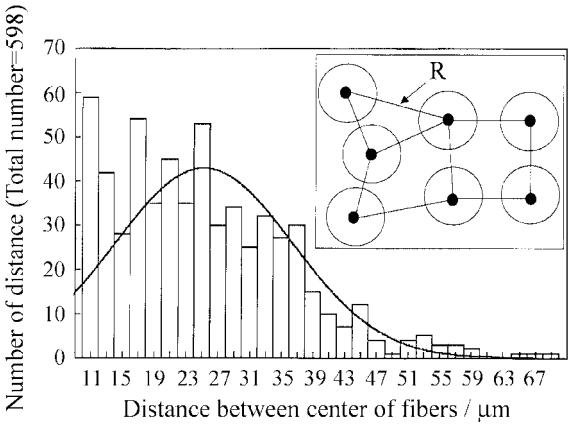

Fig. 5. Distribution of distance between the center of fibers in the cross section of the composite with 13.6 vol% fibers.

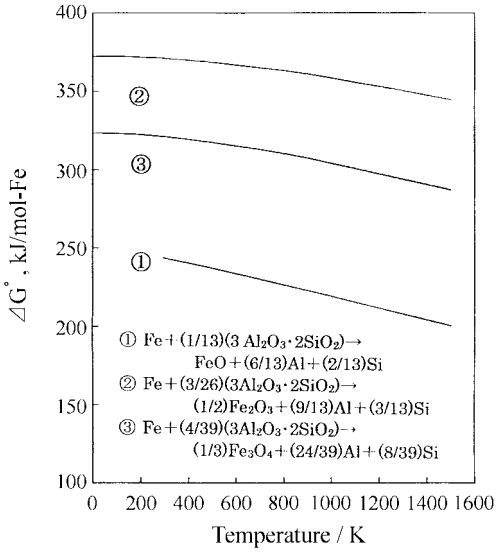

Fig. 6. Standard free energy change for the reaction of the mullite – Fe system.

0~1500 K for the reaction proposed in Fig. 6, suggesting no reaction between mullite fibers and Fe matrix. Figure 7 shows the concentration profile of Al, Fe and Si elements by electron probe micro analyzer (JSM-820, Jeol Co. Ltd., Japan) for the composite hot – pressed at 1000°C. The result shown in Fig. 7 supports no chemical interaction between the mullite fibers and HSS matrix.

Figure 8 shows the stress ñ strain curves of mullite fiber (13.6 vol%)/HSS composite hot – pressed at 1000°C. The elastic deformation curve for mullite fiber was calculated from the tensile strength and Young's modulus reported by the manufacturer. The three specimens showed the similar ductility with fracture strengths of 700~1000 MPa. The difference of apparent Young's module among the specimen was small. As seen in Fig. 8, the flexural strengths of the composites were 1/2~1/3 of the tensile strength of mullite fiber. The detailed analysis of the fracture behavior of the composites will be studied with many samples in a next paper.

Figure 9 shows the thermal expansions of monolithic HSS and the mullite fiber (13.6 vol%)/HSS composite upon heating to 1000°C in Ar atmosphere. Both the

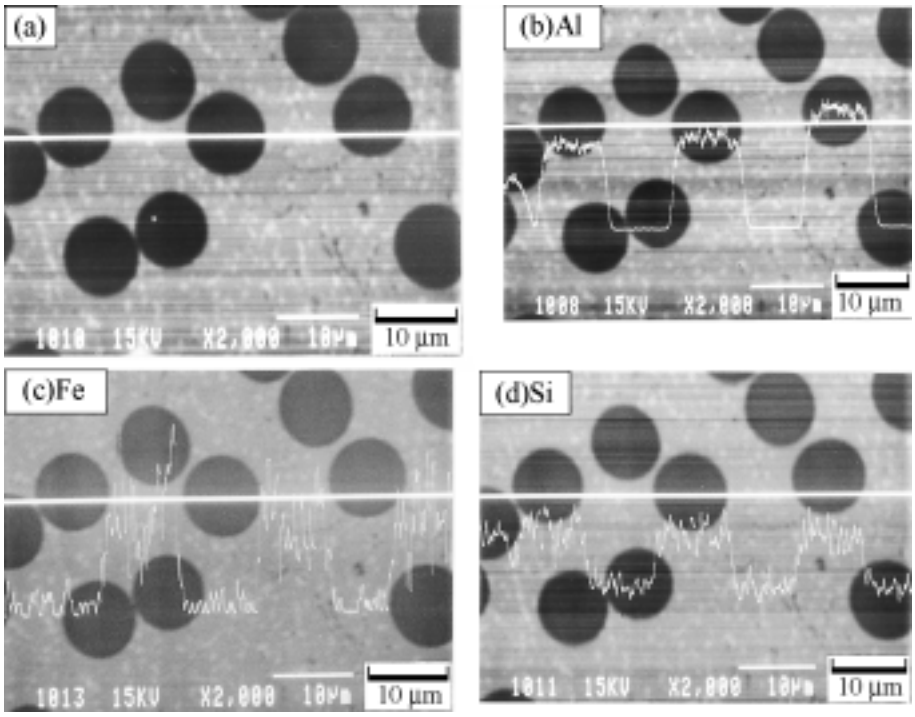

Fig. 7. Cross section of composite (a) and concentration profile Al (b), Fe (c) and Si (d) elements by electron probe micro analyzer for the composite with 13.6 vol% mullite fibers.

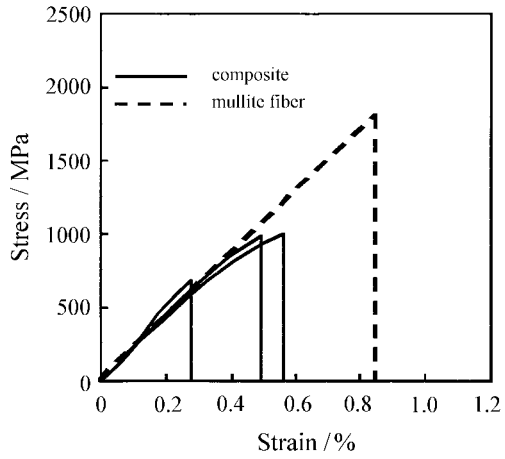

Fig. 8. Stress – strain curves of the composites with 13.6 vol% mullite fibers. The curve for mullite fiber was calculated from the tensile strength and Young's modulus.

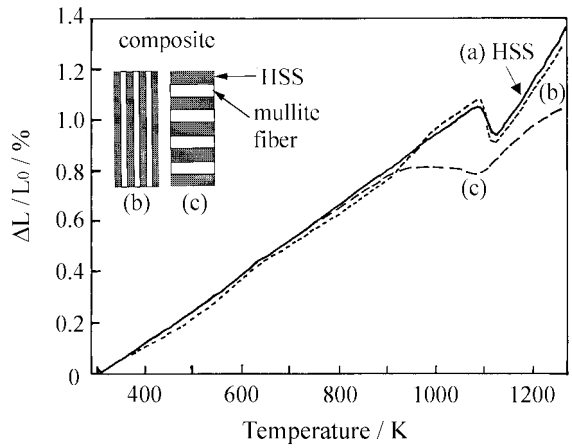

Fig. 9. Thermal expansion behavior of monolithic HSS and the composite with 13.6 vol% fiber in Ar atmosphere.

samples were hot – pressed at 1000°C under a pressure of 39 MPa. The relative density was 99.9 and 95.7 % for the hot – pressed HSS and composite, respectively. The thermal expansion behavior of the composite parallel to the oriented unidirectional mullite fibers was similar to that of monolithic HSS. The thermal expansion coefficients in the temperature range of 300~1073 K were 12.5×10^{-6}/K. The change of the thermal expansion at around 1073 K is associated with the phase transformation of ferrite $\rightleftarrows$ austenite in the HSS matrix [6]. The thermal expansion of the composite perpendicular to the mullite fibers became lower at temperatures above 900 K. That is, the thermal expansion of the composite is influenced by the orientation of the mullite fibers.

Conclusions

The composite of oriented unidirectional mullite fiber (4~68 vol%)/HSS matrix was formed in an ethylenglycol solution under the ultrasonic irradiation at 38 kHz. The density of composites hot – pressed at 1000 °C under a pressure of 39 MPa became higher for lower content of fibers. The unidirectional fibers were well dispersed in the HSS matrix. The average distance between the center of fibers in the cross section was close to the value calculated from fiber fraction. No reaction occurred at the interfaces between HSS and mullite fibers. The composite with 13.6 vol% fiber showed 1000 MPa of four point flexural strength at room temperature. The thermal expansion of composite was influenced by the orientation of fibers included.

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