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Effect of Alumina Coating on Mechanical Properties of SiC Whisker Reinforced Silicon Nitrate Ceramic Composite

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Alumina coated SiC whiskers were prepared by homogeneous precipitation of aluminum sulfate. The Si_3N_4 composites reinforced with coated SiC whiskers were fabricated by hot-pressing at 1800 ℃ for 2 h under an N_2 atmosphere of 0.1 MPa to examine the effects of coated whiskers on the mechanical properties of SiC whisker reinforced Si_3N_4 composite.

By the addition of alumina coated SiC whiskers instead of as received ones, the fracture toughness of composite was about 6.7 $MPam^{1/2}$ which was slightly lower than as received SiC whisker reinforced composite. This result seems to be caused by the fact that the crack deflection and whisker pull-out were decreased. Thus, alumina coated SiC whiskers were considered to form relatively strong interface bond with Si_3N_4 matrix.

Keywords *: SiC whisker, Si_3N_4 composite, hot-pressing, mechanical property*

1. Introduction

Si_3N_4 is the most promising material for structural applications at high temperature due to its excellent mechanical properties. However, the fracture toughness value of monolithic Si_3N_4 is low. Many attempts[1)-2)] have been made to enhance the fracture toughness by the incorporation of SiC whisker(SiC(w)). The mechanical properties of SiC(w)/Si_3N_4 composite depend on following structural factors; (1) matrix, (2) whisker and (3) interface between matrix and whiskers. For a given SiC(w)/Si_3N_4 composite, interface between matrix and whisker is an important factor, because it occupies a very large area in composite. It is well known that the toughening mechanisms such as crack deflection, whisker pull-out and bridging become active when interfacial strength between matrix and whiskers is relatively weak, contributing fracture toughness improvement. Much attention has been paid on methods for controlling interfacial strength[3)-6)] and its mechanical properties of coated whisker reinforced ceramic composite. However, previous studies did not evaluate the degree of crack deflection quantitatively, and did not also investigate the microstructural factors such as grain size and orientation nature of β-Si_3N_4 acicular grains in matrix and its effect on mechanical properties.

Based on these things, in the present study, alumina coated SiC(w) was prepared, and coated SiC(w)/Si_3N_4 composite was fabricated by hot-pressing. The monolithic Si_3N_4 and as-received SiC(w)/Si_3N_4 composite were also fabricated under the same condition for comparison. The effects of coated whiskers on the mechanical properties of SiC(w)/Si_3N_4 composite were carefully investigated by considering the degree of crack deflection and above mentioned microstructural factors.

2. Experimental procedures

As starting powders, Si_3N_4 (Ube Kosan Co.; α -crystal phase>95%; average particle size, 0.17㎛), Y_2O_3 (0.25㎛) and Al_2O_3 (0.39㎛) were used. Alumina coated layers on the surface of SiC whiskers(Tokai Carbon Co. LTD.; TWS-400 type; β phase; ϕ1.0-1.4㎛×L20-30㎛) were prepared by homogeneous precipitation of aluminum sulfate ($Al_2(SO_4)_3$). The details for coating treatments are reported in our previous study.[7)] The thickness of alumina coated layers were about 0.075~0.1 μm. Table 1 shows the composition of sintering aids and the kinds of SiC(w) in composites.

Si_3N_4, Y_2O_3 and Al_2O_3 were mixed by wet ball milling using MC jar, Si_3N_4 balls and ethyl alcohol. After ball milling for 70 h, as-received SiC(w), and alumina coated SiC(w) were added to mixed powders, respectively. To disperse whiskers homogeneously, ultrasonic dispersion and stirrer were used at the same time for 20 minutes.

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Table 1. Symbol of specimens and compositions of matrices.

Symbol of specimen	Compositions of matrices	Amount of SiC (w)	Kinds of SiC (w)
S-4Y-3A	Si_3N_4-4mol%Y_2O_3-3mol%Al_2O_3 (S-4Y-3A)	-	-
S-4Y-3A/Was		20vol%	as-received (Was)
S-4Y-3A/W_A		20vol%	Al_2O_3 coated (W_A)
S-8Y-6A	Si_3N_4-8mol%Y_2O_3-6mol%Al_2O_3 (S-8Y-6A)	-	-
S-8Y-6A/Was		20vol%	as-received (Was)
S-8Y-6A/W_A		20vol%	Al_2O_3 coated (W_A)

Subsequently, mixed powders were dispersed by ball milling for 2 h, and then, dried for 3 h at 120 ℃ in dry oven. Mixed powders without SiC(w) were also prepared under the same ball-milling condition for comparison. Hot-pressed compacts of 4×28×35 mm^3 were fabricated by hot-pressing under 20 MPa at 1800 ℃ for 2 h under an N_2 atmosphere of 0.1 MPa. Specimens of 3×4×40 mm^3 were cut out from the hot-pressed compact. The surfaces of specimens were grinded and/or polished with a 200 or 1200 mesh diamond wheel.

The microstructures and fracture surfaces of specimens were investigated by optical microscopy, SEM (Hitachi, X-4200) and an X-ray diffractometer (XRD; Cu Kα, Philips, PW3719). The relative density(Ds), flexural strength (FS; 3-point bending test with a span of 30mm), and fracture toughness{K_{IC}; indentation fracture (IF) method and controlled surface flaw (CSF) method[8]}were measured. From the XRD analysis, the degree of orientation anisotropy of acicular β-Si_3N_4 grains was semiquantitatively evaluated from the intensity ratio of $\{I_{(200)}/(I_{(200)}+I_{(101)})\}$.[9] Where, the (200) plane is a prismatic plane parallel to the c-axis of β-Si_3N_4, and the (101) plane is a pyramidal plane about 66° from the c-axis. This and $I_{(200)} \fallingdotseq I_{(101)}$ imply that the c-axes (major axes) are oriented isotropically if the intensity ratio is 0.5, turn more vertical to hot-press direction as the ratio approaches unity, and turn more parallel with the c-axis as it approaches zero.

3. Results and discussion

Observation of polished surface for monolithic Si_3N_4 and SiC(w)/Si_3N_4 composites revealed that major axes of SiC(w) oriented to vertical direction by hot-pressing in vertical planes to the hot-press direction and were distributed randomly in parallel planes to the hot-press direction. There was no difference in aspect ratio of whisker between coated one and as-received one. Thus, it implies no whisker damage during coating treatment.

Fig. 1 shows the properties for three kinds of specimens.

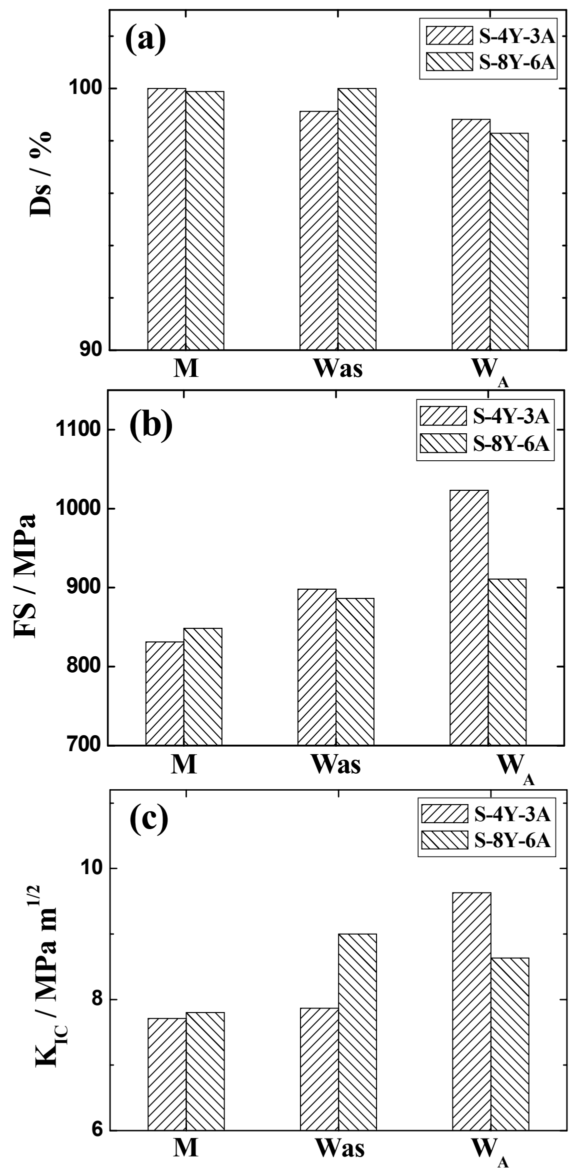


Fig. 1. Relative density (a), flexural strength (b) and fracture toughness (c; IF method) of monolith (M) and two kinds of SiC(w)/Si_3N_4 composites(W_{as}, and W_A).

The Ds of these specimens were above 98%, showing that the densification of the specimens was nearly complete. The FS of SiC(w)/ Si_3N_4 composite was higher than that of the monolithic Si_3N_4. The K_{IC}(measured by IF method) of SiC(w)/Si_3N_4 composites were also higher than that of monolithic Si_3N_4, irrespective of the kinds of SiC(w). By the addition of alumina coated SiC(w) instead of as-received ones, the K_{IC} of SiC(w)/Si_3N_4 composite with 4 mol%Y_2O_3+3 mol%Al_2O_3 was slightly increased, but the K_{IC} of composite with 8 mol% Y_2O_3+6 mol% Al_2O_3 remained unchanged. Thus, the K_{IC} was evaluated again by CSF method which was more precise than IF method. The results are shown in Fig. 2. The K_{IC} of SiC(w)/Si_3N_4 composites were higher than that of monolithic Si_3N_4. The K_{IC} of alumina coated SiC(w)/Si_3N_4 composite was about 6.7 MPam$^{1/2}$ which was slight lower than that of as-received SiC(w)/Si_3N_4 composite.

Fig. 3 shows the SEM micrographs of etched surfaces. In the micrographs of SiC(w)/Si_3N_4 composites, the SiC (w) could not be distinguished from Si_3N_4 grains, owing to their similar acicular shape and size. However, by considering that the amount of SiC(w) is much smaller than that of acicular Si_3N_4 grains, the Si_3N_4 grain size in the as-received SiC(w)/Si_3N_4 composite was estimated to be nearly equal to that in the corresponding monolith. On the other hand, the grain size of Si_3N_4 in alumina coated SiC(w)/Si_3N_4 composite decreased, although the phase transformation of Si_3N_4 was completed. The decrease in grain size may be due to the change in the composition of sintering aids by additional alumina. XRD analysis for monolithic Si_3N_4 and SiC(w)/Si_3N_4 composites were conducted to identify the grain boundary phase. For S-4Y-3A, no XRD peaks corresponding to grain boundary phase were detected.

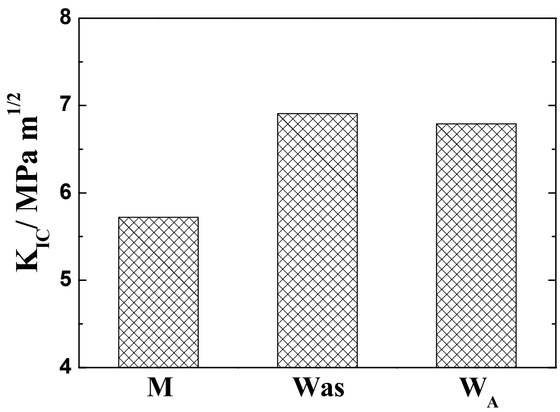


Fig. 2. Fracture toughness (K_{IC}; CSF method) of monolith (S-8Y-6A) and two kinds of SiC(w)/Si_3N_4 composites(W_{as}, and W_A).

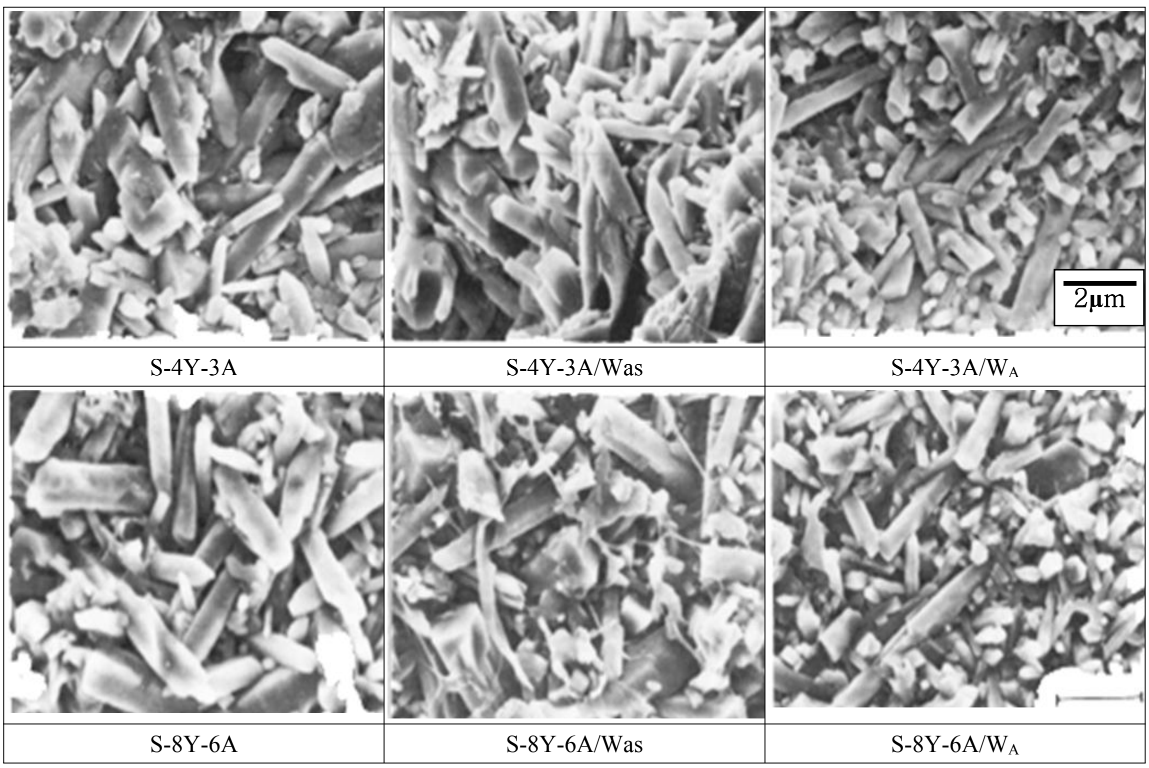


Fig. 3. SEM photographs of etched surface for monolithic Si_3N_4 and SiC(w)/Si_3N_4 composites.

This may be attributed to the small amount of crystalline phase. Grain boundary phases in S-8Y-6A/Was were Y_2SiO_5 and $Y_2Si_2O_7$, but, no XRD peaks in S-8Y-6A/Wa were observed. Thus, grain boundary phase is considered to exist in amorphous form. The degree of orientation of acicular β-Si_3N_4 grains were investigated on the basis of XRD analysis on vertical and parallel surface to hot-press direction.

Fig. 4 shows XRD intensity ratio of $\{I_{(200)}/(I_{(200)}+I_{(101)})\}$. The ratios of monolith and as-received SiC(w)/Si_3N_4 composites were about 0.7~0.8 in surfaces vertical to hot-press direction, but about 0.4 in surfaces parallel to hot-press direction. This means that the major axes of β-Si_3N_4 acicular grains oriented vertical to hot-press direction. Whereas the intensity ratio of alumina coated SiC(w)/Si_3N_4 composite were about 0.6~0.65 in surface vertical to hot-press direction, and about 0.5 in surface parallel to hot-press direction. It means that the β-Si_3N_4 grains in composite are oriented nearly random.

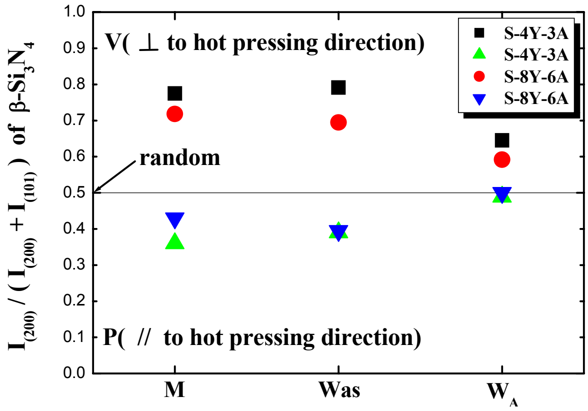


Fig. 4. X-ray intensity ratio of $I_{(200)}/(I_{(200)}+I_{(101)})$ of β-Si_3N_4 grains in surfaces vertical(V) and parallel(P) to hot-press direction for each specimen.

Fig. 5 shows SEM micrographs of cracks formed at the corners of Vickers indentation in each specimen. The degree of crack deflection in S-4Y-3A was observed to be larger than that in S-8Y-6A. It also become larger by the addition of as-received SiC(w).

Fig. 6 shows cumulative frequency and length of deflection angle of crack segment to main crack direction for monolithic Si_3N_4 and SiC(w)/Si_3N_4 composites. These were investigated for cracks of a total length of 300 ㎛ on SEM micrographs with a magnification of 2,000. The lowering of the location of the curve means that the crack deflects at the interface of SiC(w) or Si_3N_4 grains up to a higher angle

From Fig. 6, it appears that the degree of crack deflection of as-received SiC(w)/Si_3N_4 composite was higher than that of monolithic Si_3N_4. But, alumina coated SiC(w)/Si_3N_4 composite showed lower degree of crack deflection

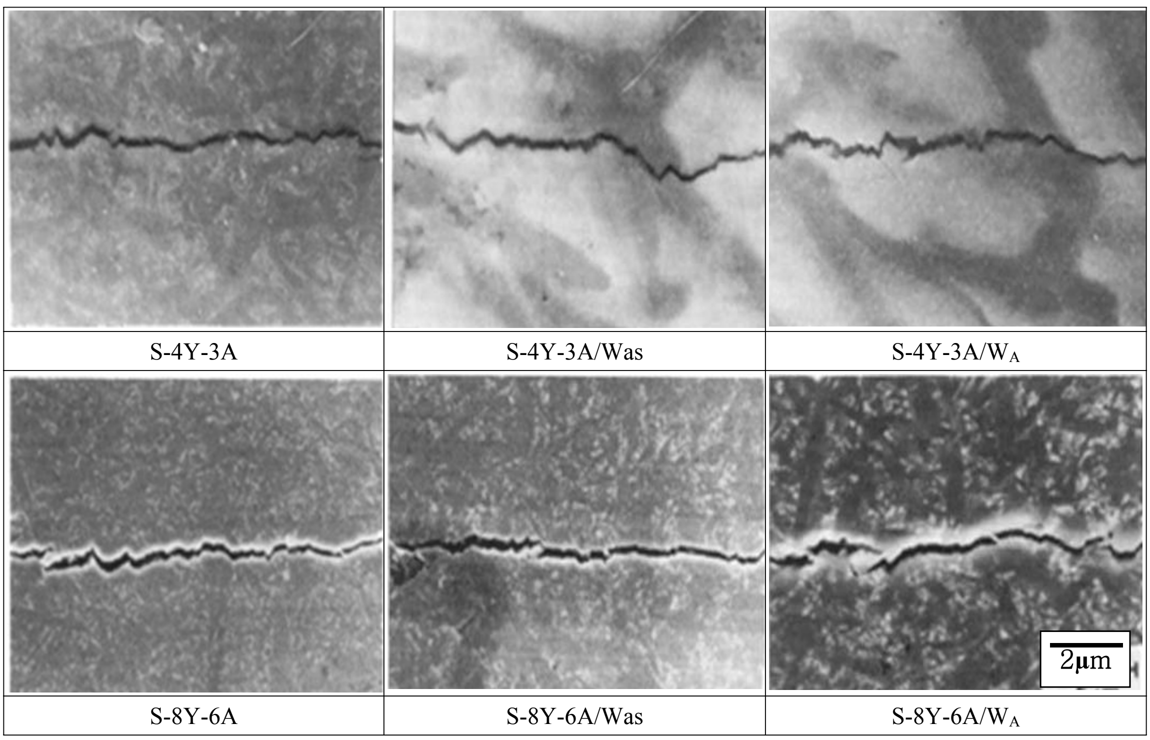
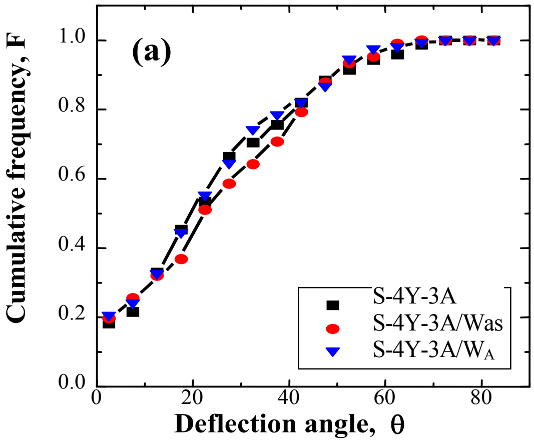


Fig. 5. SEM photographs of crack induced by Vickers indenter in each specimen.



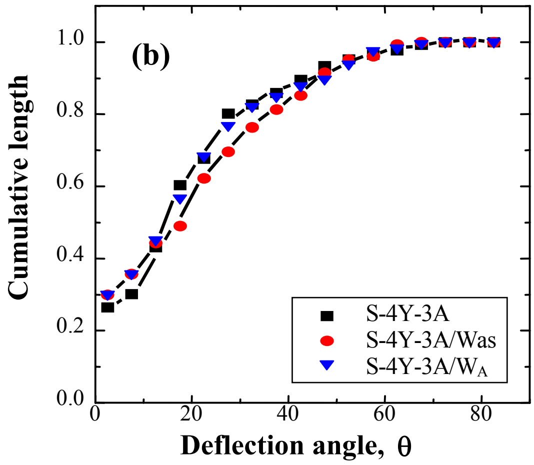


Fig. 6. Cumulative frequency(a) and length(b) of deflection angle of crack segment to main crack direction for monolith(S-4Y-3A) and two kinds of SiC(w)/Si_3N_4 composites(Was, and W_A)

compared with as-received one. Investigation for the frequency and length of deflected crack provides indirect information on interfacial strength between matrix and whiskers. The lower the degree of crack deflection, the stronger the interfacial strength. Thus, alumina coated SiC(w)/Si_3N_4 composite is considered to have relatively strong interfacial strength between matrix and whiskers.

4. Conclusions

The effects of alumina coated whiskers on the mechanical properties of SiC whisker reinforced Si_3N_4 ceramic composite were investigated. The results obtained can be summarized as follows.

(1) By the addition of alumina coated SiC whiskers instead of as-received ones, the fracture toughness of composite was about 6.7 $MPam^{1/2}$ which was slightly lower than as-received SiC whisker reinforced composite.

(2) The alumina coated SiC(w)/Si_3N_4 composite showed low degree of crack deflection and pull-out in fracture surface compared with as-received one. Therefore, alumina coated layers were considered to form relatively strong interfaces with matrix.

(3) The β-Si_3N_4 grains in alumina coated SiC whisker/Si_3N_4 composite tended to orient more parallel to hot-press direction, and their size were decreased, differing from those in as-received SiC(w)/Si_3N_4 composite.

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