Mechanical Properties of In-situ Prepared SiC Whisker- and Nano Particle-Reinforced Si_3N_4

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Abstract

A trial of in-situ preparation of SiC whisker- and nano particle-reinforded Si_3N_4 was carried out. Bending strength showed higher values by adding the nano particles 'after' the in-situ whiskerizing reaction than 'before',although the improvement was not so remarkable. With regard to fracture toughness, a slightly higher value was observed in the case of adding the particles 'before' than 'after' the in-situ reaction, where the form of whiskers look constricted due to the co-existence of particles. Relative density was not so high enough that HIP treatment would be needed for any more effective improvement.

1.Introduction

In-situ preparation of SiC whiskers in Si_3N_4 matrix together with SiC nano particles (denoted as SiC_{np+w}/Si_3N_4) has never been tried so far. SiC whiskers are, in general, known as a toughness improving reinforcer, while nano particles can play a role to improve the strength. In addition, in-situ preparation of the whiskers may solve the environmental problem accmpanied by direct handling of them. In this article, monolithic specimens as well as those of in-situ prepared SiC_w/Si_3N_4 were prepared for comparison.

2.Experiment

2.1 Preparation of composites

As typical SiC nano particles,'T-1' prepared by Sumitomo Osaka Cement Co.,Ltd.,was employed. Whiskers used for comparison were 'TWS-400' by Tokai Carbon Co.,Ltd. All the other materials used, and every procedure,such as mixing, whiskerizing, homogenizing and hot-pressing, are as described elsewhere[1]. A flow sheet to fabricate the composite is as shown in Fig.1.

As seen in Fig.1, there are 3 ways to add the nano particles:

(1)After in-situ whiskerizing reaction,at the same time when sintering aid is mixed prior to sintering;'after in-situ method'

(2)Before in-situ reaction at the time when raw materials are mixed by ball milling;'before in-situ method'

(3)At the same time when all the raw materials with sintering aid is mixed, followed by hot-pressing,where the whiskerizing reaction is carried out at sintering stage;'only hot-pressing (HP) method'

For each method above, 2 volume-ratios such as Si_3N_4:SiC_w:SiC_{np}=80:10:10 and 80:15:5, totally 6 runs, were practiced.
In order to obtain the volume-ratio of 80:10:10 in 'after in-situ method', a mixture of Si_3N_4:C:Fe_2O_3=92.4:7.4:0.15 by weight was employed for in-situ reaction, followed by ball-milling of a mixture, consisting of reacted powder:nano particles:Y_2O_3:Al_2O_3=79.9:8.9:8.0:3.2 by weight. To 80:15:5(vol.), each ratio above was 91.0:8.9:0.15 and 84.3:4.4:8.1:3.2 by weight respectively.
In the case of 'before in-situ method' for 80:10:10(vol.), the ratio of Si_3N_4:C:Fe_2O_3:SiC_{np} was 84.1:6.8:0.15:9.0 by weight, followed by in-situ reaction. After the reaction, only the sintering aid was mixed at the weight-ratio of 88.8:8.0:3.2. Similarly for volume-ratio 80:15:5, each weight-ratio was 87.0:8.5:0.15:4.4 and 91.6:5.1:3.3.
For 'only HP method', all the materials, Si_3N_4, C, Fe_2O_3, SiC_{np}, Y_2O_3 and Al_2O_3, 75.6:6.1:0.13:8.1:7.2:2.9(weight) for 80:10:10(volume) were mixed. For volume ratio of 80:15:5, weight ratio of 78.4:7.6:0.13:3.9:7.0:2.8 was used. During whiskerization the pressure was not applied, 1600°C x 2h. Hot pressing was carried out under 33MPa(uniaxial), 1850°C x 1.5h for sintering.
For comparison, monolithic material(Si_3N:Y_2O_3:Al_2O_3=93.0:5.2:2.0 by weight), as well as physically mixed composite consisting of 80:10:10 by volume as above mentioned (Si_3N_4:SiC_w:SiC_{np}:Y_2O_3:AlO=70.9:8.9:8.9:8.0:3.2 by weight), was used.

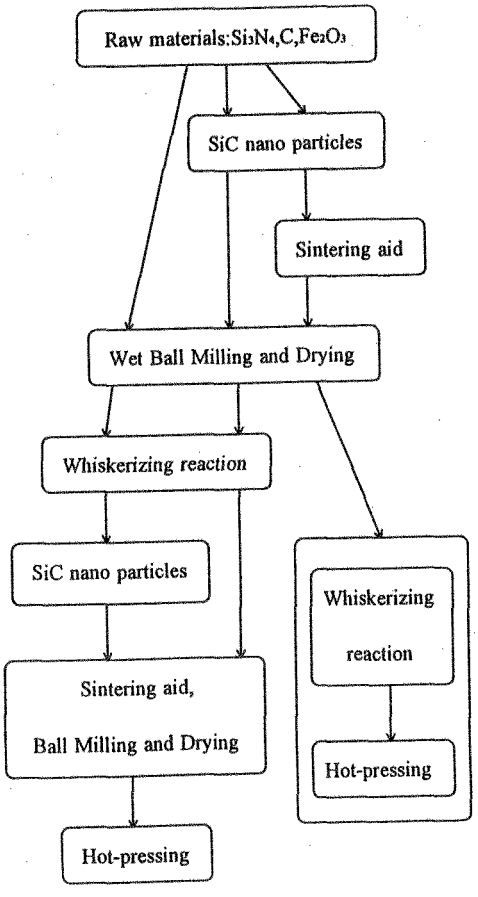
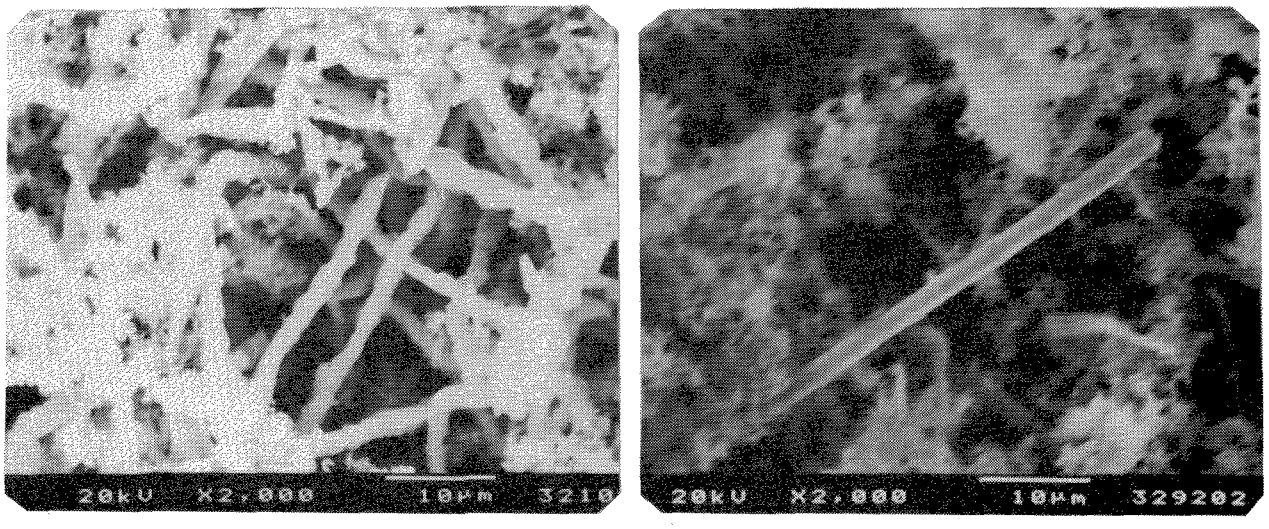


Fig. 1. A flow sheet to fabricate the SiC_{np+w}/Si_3N_4 composite

2.2 Determination of bending strength and fracture toughness
Size and shape of specimen were in accordance with JIS R-1601. 4 point method was applied for strength and V-notch was machined for toughness. Apparent density was determined by Archimedes method.

3.Result and discussion

3.1 S E M observation
Except the specimens obtained by 'only HP method',all of reacted materials contain such typical whiskers as seen in Fig.1;however, the whiskers obtained by 'before in-situ method' partly show constricted shape, differing from those by 'after in-situ method'. This might be due to an effect of nano particles during the in-situ process, since such a shape cannot be observed in all the whiskers by the latter method.



(a)'Before in-situ method' (b)'After in-situ method'
Fig. 2. S E M of SiC_{np+w}/Si_3N_4 prior to HP

3.2 Density
As summarized in Table 1, density of all the specimens except monolithic one was around only 94%, differing from the result obtained elsewhere[1], 98 to 101%. Even the value of composite specimens without nano particles were also only about 94%. To improve the density, HIP would play a role.

3.3 Strength and toughness
Fig.3 and 4 illustrate the change in strength and toughness as a function of whisker content with regard to processing.
As observed in Fig.3, strength of the specimens obtained by 'After in-situ method' show a slight improvement, differing from those by 'Before in-situ and HP only methods'. It may suggest that the role of nano particles was played in the case when its addition was done after the in-situ reaction. If the density can be improved, the effect might be more remarkable, since only the density of monolithic one was much higher than the others, as seen in Table 1.
In contrast with the above result, a slight increase of fracture

Table 1 Relative density of all the specimens

Samples	Vf(S:w:p)%	Relative density
Monolithic Si_3N_4	(100:0:0)	98.3
"After In-situ" method	(80:10:10)	94.0
	(80:15:5)	94.6
"Before In-situ" method	(80:10:10)	94.0
	(80:15:5)	94.6
"HP only" method	(80:10:10)	93.7
	(80:15:5)	93.6
SiC_w/Si_3N_4(established)	(80:10:10)	93.5
SiC_w/Si_3N_4(In-situ)	(80:20:0)	93.7

toughness was observed only in 'Before In-situ method', as shown in Fig.4. It might be related with a basic difference of whisker shape as seen in Fig 2. The improvement of density is needed for the exact comparison also in this discussion on toughness.
Anyway, this first trial of co-existence of whiskers and nano particles would have a positive significance that all of the specimens obtaind have regular mechanical properties, suggesting no remarkable defect within them.

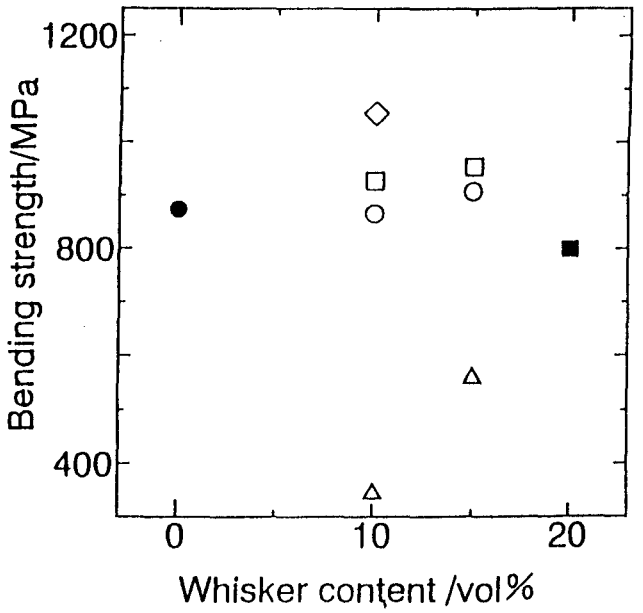


Fig.3 Bending strength of SiC_w/Si_3N_4 composite as a function of SiC_w content.
● :Monolithic Si_3N_4 □ : "After In-situ" method
○ : "Before In-situ" method △ : "HP only" method
◇ :SiC_w/Si_3N_4(established) ■ :SiC_w/Si_3N_4(In-situ)

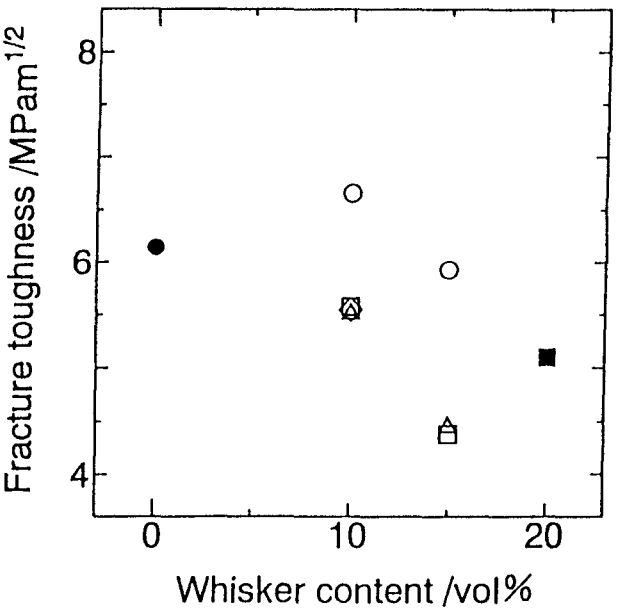


Fig.4 Fracture toughness of SiC_w/Si_3N_4 composite as a function of SiC_w content.
● :Monolithic Si_3N_4 □ :"After In-situ"method
○ :"Before In-situ"method △ :"HP only"method

Reference

1) S.Yamada, Y.Koyama, T.Tada, E.Yasuda and T.Akatsu, "Eng. Ceram. '96":Higher Reliability through Processing,239-49(1997), Kluwer Acad.Pubs.(Netherlands)

Received September 25, 1997
Accepted January 27, 1998