Si₃N₄-SiC Composite Consolidated by Post-HIPing Process

Yoshio UKYO, Toshio KANDORI and Shigetaka WADA

Toyota Central R & D Labs., Inc., Nakakute, Aichi-gun, Aichi 480-11

ポスト HIP 法による Si₃N₄-SiC 複合体

右京良雄・神取利男・和田重孝

(株)豊田中央研究所, 480-11 愛知県愛知郡長久手町大字長久手字横道41-1

[Received July 9, 1993; Accepted November 16, 1993]

Si₃N₄-SiC particle composites constaining up to 30 wt% SiC were sintered to a neary full density using gas pressure sintering and HIPing process (post-HIPing). Fine SiC powder with an average particle size of about 0.03 μ m, and Y₂O₃ as sintering additive were used. Si₃N₄-30 wt% SiC sintered composite with a strength of about 700 MPa at 1400°C was obtained by post-HIPing.

Key-words : Composite, Si₃N₄, SiC, HIP, Sintering

1. Introduction

Si₃N₄ ceramics have attracted much attention as high temperature structural materials because of their excellent high temperature mechanical properties. For the widespread application of Si₃N₄-based ceramics, it is necessary to attain higher strength up to higher temperature and higher toughness. There is a possibility that the alternation of Si₃N₄ monoliths through SiC dispersoid additions offers a potential of considerable improvement of high temperature strength and fracture toughness.

Extensive studies have been carried out on Si₃N₄-SiC composites.¹⁾⁻⁸⁾ For example, Lange¹⁾ has showed that the strength of the Si₃N₄-SiC composite prepared by hot-pressing with MgO as sintering additive was twice compared with that for monolithic Si₃N₄ at 1400°C. Recently, Niihara et al.⁵⁾⁻⁸⁾ have succeeded in the development of the Si₃N₄-SiC composite with extremely high temperature strength, about 1.0 GPa at 1400°C. They used the amorphous Si-C-N powders prepared by vapor phase reaction as starting materials, and Y₂O₃ as sintering additive.

Dense Si₃N₄-SiC composite can only be obtained by hot-pressing or glass encapsulated HIPing, because the addition of SiC into Si₃N₄ decreases sinterability of the composites. For a wide variety of applications, it is very important to obtain dense Si₃N₄-SiC composites by post-HIPing.

The purposes of this study are to prepare the dense Si₃N₄-SiC composites by post-HIPing and to measure their mechanical properties.

2. Experimental procedures

The SiC powder used in this study (Sumitomo Cement Co., Ltd., T1 grade, obtained by plasma CVD

method) has an average particle size of about 0.03 μ m and a specific surface area of about 30 m²/g. The particle size of this SiC powder is very small compared with that of usual SiC powders. The Si₃N₄ (UBE SNE-10) and the above SiC powder were mixed with 8 wt% Y₂O₃ as sintering additive, and ball-milled well in ethanol using Si₃N₄ jar for 50 h. The contents of SiC were 0, 10 and 30 wt%. The milled and dried powders were die pressed at 20 MPa. The compacts were sintered in a N₂ atmosphere at 1 MPa between 1750 and 1900°C for 4 h, and then HIP treatment was conducted under a pressure of 100 MPa in N₂ at 1750°C for 1 h.

The fracture strength from room temperature to 1400°C was measured in four point loading according to JIS R1601. The microstructure was observed by scanning electron microscopy (SEM).

3. Results and discussions

Figure 1 shows the relationship between sintering temperature and the apparent density of Si₃N₄-30 wt% SiC composites which was measured by Archimedes' method. As shown in this figure, the density increased with increasing sintering temperature and a nearly full dense Si₃N₄-SiC composite was obtained when sintering temperature was 1900°C. It has been known that the dense Si₃N₄-SiC composites with a SiC content such as high as 30 wt% cannot be obtained by pressureless sintering when only Y₂O₃ was used as sintering additive. Although the reason for this has yet been clarified, we consider it to be closely related to the use of very fine SiC powder used. For example, the density of Si₃N₄-30 wt% SiC composite sintered at 1900°C for 4 h was only about 3.0 g/cm³ when SiC powder with an average particle size about 0.3 μ m (Ibiden Ultrafine grade) was used.

Kijima⁹⁾ has obtained the dense SiC sintered body by hot-pressing the mixtures of fine SiC powder and commercial SiC powder. In this case, it is thought that fine SiC powder worked as sintering additive. On the other hand, the addition of SiC powder into Si₃N₄ decreases the sinterability even if fine SiC powder is used. For example, the dense Si₃N₄-SiC composite could not be obtained when the amount of Y₂O₃ was lower than 8 wt%. This means that fine

Fig. 1

Fig. 2

Fig. 3

Fig. 1. Relation between sintering temperature and density of Si_3N_4 -SiC composite containing 30 wt% SiC particle before and after post-HIPing.

Fig. 2. Effect of HIP treatment on strength of Si_3N_4 -30 wt% SiC composite sintered at 1850°C (□) and 1900°C (■).

Fig. 3. Temperature dependence of strength of Si_3N_4 -SiC particle composites.

Fig. 4. Microstructure of Si_3N_4 -SiC particle composite observed by SEM.

SiC powder used does not act as sintering additive. The roles of fine SiC powder in the sintering of SiC and Si_3N_4 are completely different.

HIP treatment were then conducted on the Si_3N_4 -30 wt% SiC composites sintered at various temperatures. The density of all the composites increased by HIP treatment, and a full dense Si_3N_4 -30 wt% SiC composite was found to be obtained when sintering temperature was higher than 1850°C.

Figure 2 shows the change of the strength of Si_3N_4 -30 wt% SiC composite sintered at 1850°C (□) and 1900°C (■) by HIP treatment. As shown in this figure, remarkable improvement was observed in the strength of Si_3N_4 -30 wt% SiC composite sintered at 1850°C by HIP treatment. It is considered that the remarkable improvement in the strength of Si_3N_4 -30 wt% SiC composite sintered at 1850°C is attributed to the increasing of the density by HIP treatment. Figure 3 shows the temperature dependence of the strength of Si_3N_4 -SiC composites with 0, 10 and 30 wt% SiC content, which were obtained by sintering at 1850°C for 4 h and post-HIP treatment. The change of strengths of all the specimens were very small up to 1400°C and the composites with 30 wt% SiC showed highest strength of about 700 MPa at 1400°C. Figure 4 shows microstructures of these composites observed by SEM. The figures revealed that the grain growth of Si_3N_4 was suppressed by the addition of SiC. It is not clear where SiC particles exist because the grain size is too small and transmission electron microscopy (TEM) observation with high magnification is required to be conducted. However, we estimate that most of the SiC particles exist in Si_3N_4 grains as pointed out by Niihara et

al.,⁵⁾ and that this is one of the important factors for excellent high temperature strength. Evaluation of the high temperature properties such as oxidation resistance or creep resistance are now being conducted.

4. Conclusion

- (1) Si_3N_4 -SiC composites containing up to 30 wt% SiC particle were consolidated to a nearly full density using conventional gas pressure sintering, when very fine SiC particle with an average particle size about 0.03 μm was used.
- (2) Si_3N_4 -SiC composites containing up to 30 wt% SiC particle with a strength about 700 MPa at 1400°C was obtained using post-HIPing.

References

- 1) F. F. Lange, *J. Am. Ceram. Soc.*, **56**, 445-50 (1973).
- 2) S. T. Buljan, J. G. Baldoni and M. L. Huckabee, *Am. Ceram. Soc. Bull.*, **66**, 347-52 (1987).
- 3) K. Ueno and W. Toibana, *Yogyo-Kyokai-Shi*, **91**, 491-97 (1983).
- 4) P. D. Shalek, J. J. Petrovic, G. F. Hureley and F. D. Gac, *Am. Ceram. Soc. Bull.*, **65**, 351-56 (1986).
- 5) K. Niihara, T. Hirano, A. Nakahira, K. Ojima, K. Izaki and T. Kawakami, "Proceedings of MRS International Meeting on Advanced Materials, Vol. 5," Materials Research Society, Pittsburgh (1989) p. 107.
- 6) K. Niihara, K. Izaki and T. Kawakami, *J. Mater. Sci. Lett.*, **10**, 112-14 (1990).
- 7) K. Niihara, K. Suganuma, A. Nakahira and K. Izaki, *J. Mater. Sci. Lett.*, **9**, 598-99 (1990).
- 8) K. Niihara, T. Hirano, A. Nakahira, K. Suganuma, K. Izaki and T. Kawakami, *J. Japan Soc. Powder and Powder Metall.*, **36**, 169-72 (1989).
- 9) K. Kijima, Proceedings of the Special Symposium on Advanced Materials, The Joint Committee for Advanced Materials Research (1990) p. 147.