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Densification Mechanism of NITE-SiC and SiC_f/SiC Composites

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ABSTRACT: *Nano Infiltration Transient Eutectic Phase - Silicon Carbide (NITE-SiC) and SiC/SiC composite have been fabricated by a Hot Pressing (HP) process, using SiC powder with an average size of about 30nm. Alumina (Al_2O_3) and Yttria (Y_2O_3) were used for additives meterials. These mixed powders were sintered at the temperature a of 1300℃, 1650℃, 1800℃ and 1900℃ under an applied pressure of 20MPa. And unidirection and two dimension wooven structures of SiC/SiC composites were prepared starting from Tyranno SA fiber. Densification of microstructure gives an effect to density . Specially, Densification Mechanism basically is important from the sintering which use the HP. In this study, the densification of NITE-SiC and SiC/SiC composite mechanism by a press displacement appears investigated. The mechanism on the densification of each sintering temperature was investigated. The each step is shows a with each other different mechanism quality.*

1.Introduction

Silicon carbide is a preferred ceramic material for many applications in harsh environmental condition because of its resistance to high temperature, aggressive chemicals and thermal expansion coefficient at elevated temperatures (Yano et al., 2003, She et al., 1999). Liquid phase sintering(LPS) processes are recognized as attractive methods for making high density SiC_f/SiC composites with good thermal conductivity and a higher fracture toughness (Suyama et al., 2002). And the use of yittria or other rate earth oxides and Al_2O_3 or AlN as sintering additive, which form, together the SiO_2 existing on the surface of the starting SiC powder, a liquid phase during the sintering, reducing the sintering temperature to values less than 2000℃ (Ihle et al., 2005).

However, a major R&D forcus for SiC_f/SiC composite is the production of high purity SiC matrix and its densification in the intra bundle (katoh et. al., 2004, kotani, et. al., 2001).

Therefore, in this study is to search the densification mechanism of NITE-SiC and SiC_f/SiC composite fabricated by hot pressing method, and the change of the microstructure particles is observed. Also, densification of the inter-fiber bundle that to making the SiC_f/SiC composites used as reinforcement material is observed.

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2. Experiments

2.1 Fabrication of NITE-SiC and SiC_f/SiC composite.

The SiC powder (Marketech International Inc., USA) with average size of about 30nm was used for NITE-SiC material. Typical composition of this nano-powder is as follows: SiC > 95%, Free C 1~2% and O 1~1.5%. Al_2O_3 (High Purity Chemical, Japan) and Y_2O_3 (High Purity Chemical, Japan) were used as sintering additives. The mixtures of SiC nano-powder and additives were blended in acetone in planetary ball mill for 12 h, then dried with an evaporator, crushed and screened.

The mixed powder was sintered by hot pressing in argon atmosphere. Process temperature was varied from 1300℃ to 1900℃. Process pressure was continuously 20 MPa. Table 1 shows the batch composition and processing condition of NITE-SiC.

To make SiC_f/SiC composite which was densificated, the work performed that the mixture powder infiltrate between inter-fiber bundle.

The fiber was used Tyranno SA fiber coated carbon of 0.25μm. The fiber was used unidirection and two dimension woven structures. The volume ratio of fiber was 15%.

2.2 Characterization of microstructure

Sintered density was measured by Archimedes' method. Field Emission Scanning electro microscope (JEOL, JSM-6700F) was used for microstructure of NITE-SiC. Also, to investigate crystatal size and density used Multi-Purpose High Resolution X-ray Diffractiometer(X'Pert PRO MPD).

3. Results and Discussions

3.1 Sintering process

Fig. 1 is sintering process used experiment. As Fig. 1 sintering temperature was raise rising 20℃/min until 1150℃ and 10℃/min until 1900℃. To investigate the sintering characteristic about holding time, we had a holding time of 30 and 60 minute at 1900℃. First, we made the monolithic SiC then we could know that specimen, having a holding time, was appear highly density. So, we did make NITE-SiCf/SiC below 1900℃. Because tyranno SA was damaged with increase temperature. we have brought a result of different study. It was showed No. 7 of Fig. 1. No. 7 was made by sintering temperature 1800℃ and continuously 20 MPa. Fig. 5 (a) and (b) are microstructure of No. 7 specimen.

To comparison, we made a NITE-SiCf/SiC composites at sintering temperature 1780℃. Pressure is the same as 20MPa.

3.2 Density of NITE-SiC and NITE-SiC$_f$/SiC

Fig. 2 is measure density of specimen at each temperature. As Fig. 2 in the case of measurement at each temperature,

Table 1 Processing Condition

Specimen ID	Sintering Additive (X wt%)	Process Temperature(℃) - Holding Time
SiC - 1	Al_2O_3:Y_2O_3 = 6:4	1300
SiC - 2	Al_2O_3:Y_2O_3 = 6:4	1650
SiC - 3	Al_2O_3:Y_2O_3 = 6:4	1800
SiC - 4	Al_2O_3:Y_2O_3 = 6:4	1900
SiC - 5	Al_2O_3:Y_2O_3 = 6:4	1900 - 30min
SiC - 6	Al_2O_3:Y_2O_3 = 6:4	1900 - 60min
SiC$_f$/SiC - 7	Al_2O_3:Y_2O_3 = 6:4	1800 - 60min
SiC$_f$/SiC - 8	Al_2O_3:Y_2O_3 = 6:4	1780 - 60min

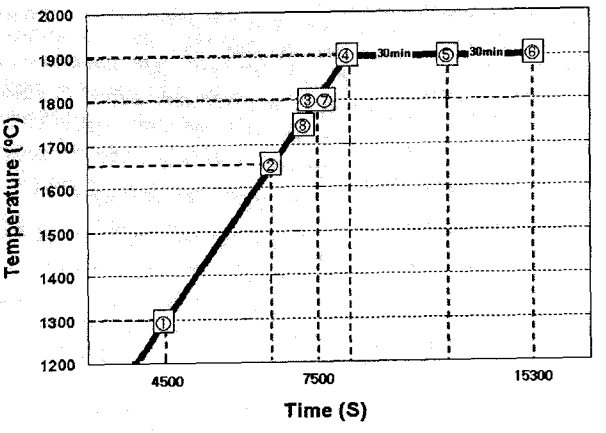

Fig. 1 sintering process

density tended to the more sintering process progress the more get higher. But the density had low-value as a whole. Then again, the specimen which holding time of 30minute and 60minute were show relative density above 95%.

From side of density, when it is interpreted densification mechanism, the density on specimen which holding time of 30minute and 60minute had only 0.0149g/cm^3 difference. So, it was over the possibility of doing there will be densification process. Fig. 3 is measure density of NITE-SiC$_f$/SiC which includes the fiber. In order to compare the result of this test, we have brought a result of different study. SiC$_f$/SiC-7 is density of specimen which adds the fiber to unidirection at 1800℃ under 20MPa. The specimen was manufactured to add fiber of unidirection and two dimension woven structures at press pressure of 20MPa. The specimen which adds the fiber at unidirection was show density of 2.64g/cm^3. The specimen which adds the fiber at two dimension woven structures was show density of 2.58g/cm^3. The density resulted differently in

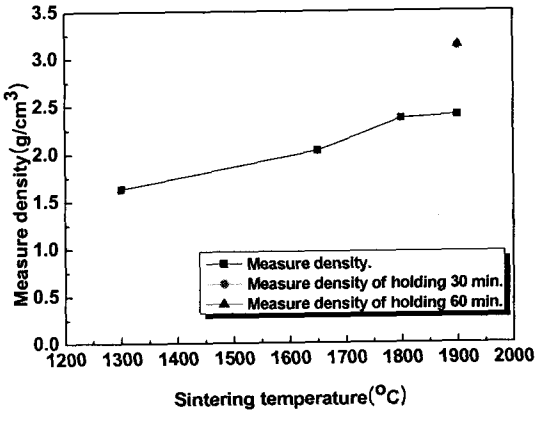

Fig. 2 Measure density

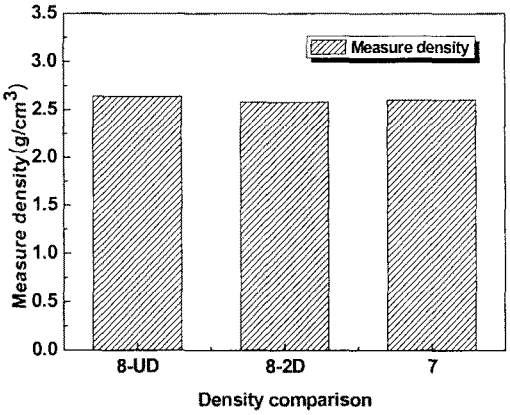

Fig. 3 Measure density of various conditions

constant ratio of mixed additives. It thought two dimension woven structures-fiber is more difficult to fill gaps than unidirection fiber. Also, the result of density have similar density of other research.

3.3 Microstructure of SiC_f/SiC

Fig. 4 is microstructure of sintering temperature, respectively. (a) 1300℃ and (b) 1650℃ in Fig. 4 are surface microstructure in sintering process at sintering temperature, respectively. From surface microstructure of (a),(b) in Fig. 4, particles shape in the picture of (a) are similar as raw material shape. Also, we could see particle in white color, this is sintering additives for liquid formation.

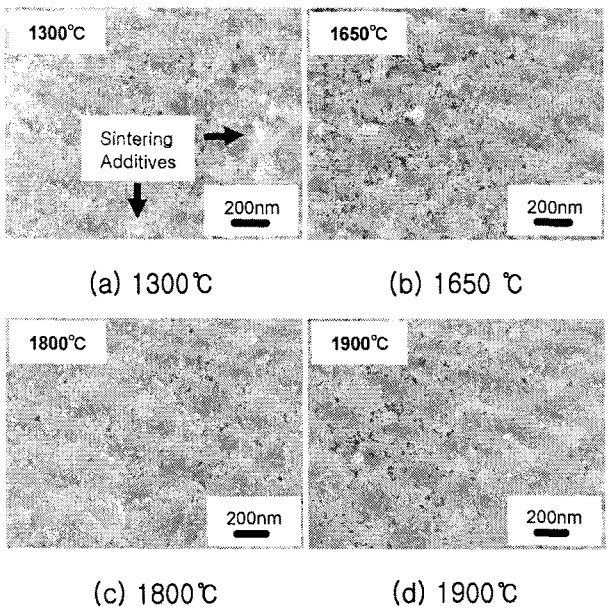

Fig. 4 microstructure of various sintering temperature.

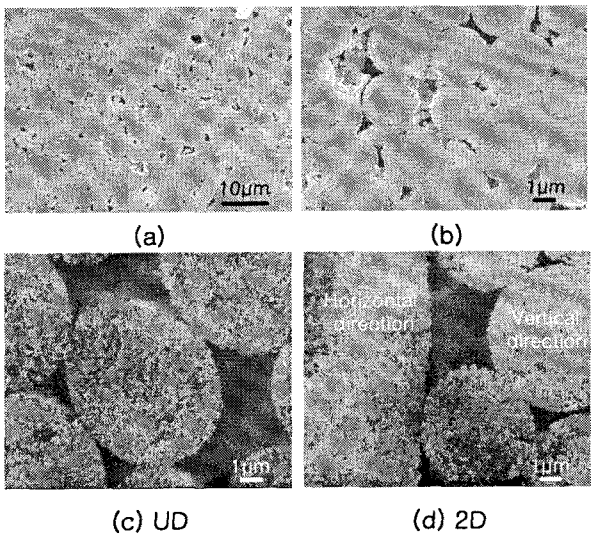

Fig. 5 Microstructure of NITE-SiC_f/SiC composite

From this, we can say that not making a liquid formation of sintering temperature at the sintered temperature. Fig .4 is a graph of particles size accumulation at sinitering temperature 1300℃and (b) 1650℃ that using the CAD program. Graphs are appeared microstructure of 1300℃ larger than 1650℃.

There are confirmed from density. Density was 2.4013g/cm^3 at sintering temperature 1900℃ and another specimen wsa appeared 3.1248g/cm^3 that having holding time of 30 minute sintering temperature 1900℃. As a result of density difference, we did think that sintering is highly advancing within 30 minute. From this part at the end of sintering process, we were thinking that particle growth was quickly appeared by grain boundary migration because it was prevented by pores.

Fig. 5 (a) and (b) is a cutting plane microstructure of fiber direction. About Fig. 7 (a), infiltrated mixture powder was changed matrix between fiber during sintering process. From

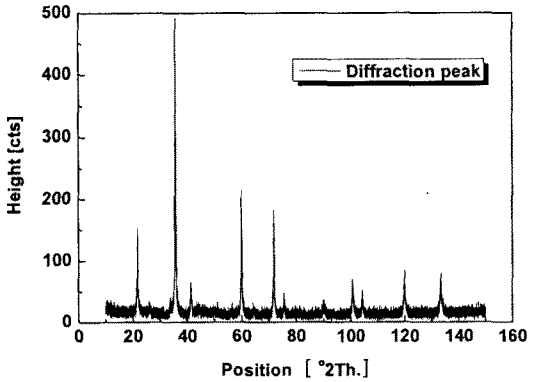

Fig. 6 XRD analysis result of SiC_f/SiC composite

(b), we could know densificatied part with pores. Fig. 5 (c) and (d) is a cutting plane microstructure unidirection and two dimension woven structures, respectively.

Mixture powder did not infiltrate. So, pores remain bewteen fiber from this picture. We could say that this state is main cause a drop of the density. In accordance with we need study that reduced pores and improved densification.

3.4 Partical Size Analays of XRD

Fig. 6 is X-ray diffraction result of NITE-SiC_f/SiC composites. A result of X-ray diffraction was appeared pattern SiC, SiO_2, $Al_2O_3 \cdot SiO_2$ and $C_{19}Y_{15}$ of NITE-SiC_f/SiC composites.

Particle size was evaluated by Scherrer's method about X-ray diffraction result.

$$Particle\ \ size = \frac{X \times 0.9}{F \times \cos(\theta)} \quad (1)$$

From this, X is X wavelength. F is Full Width of Half Maximum(FWHM). A result of measurement of particle size was 4.741×10-10m.

Also, particle density(d_x) was evaluated by X-ray diffraction result.

$$d_x = \frac{M \times Z}{N_a \times V} \quad (2)$$

N_a is Avogadros's number(6.02×10^{23}). V is volume of particle unit lattice. M is molecular weight. And Z is chemical unit. Fig. 8 is picture density of particles that appeared by X-ray diffraction result.

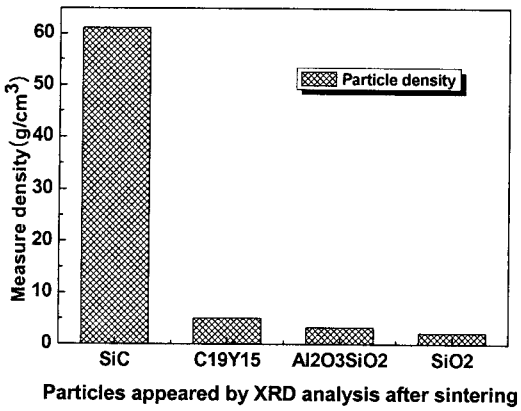

Fig. 7 Measure density of particles appeared by XRD

From Fig. 7, particle density of SiC was most larged as 61.135g/cm^3.

As this reason, Many SiC powder remain sintered specimen of sintering temperature 1780℃ because of SiC powder did not melt below 2000℃.

4. Conclusions

In this study, densification mechanism is analyze that NITE-SiC ceramics and NITE-SiC_f/SiC composites were made by NITE mothod.

(1) It produced press displacement slope on difference slope of three way during sintering process in manufaturing NITE-SiC. Early stage of sintering came out densification by press pressure. Middle stage of sintering was made liquid phase of sintering additives and final stage of sintering was occurred sudden growth of particle.

(2) In result of measured density at each temperature, the density tended to the more increasing temperature the more get higher, but the density had low-value as a whole. Also, the specimen which maintains time of 30min and 60min at 1900℃ were show relative density above 95%.

(3) Particle was changed a polygon shape from a circle by particle growth and diffusion with advance sintering process.

(4) XRD result of sintered SiC_f/SiC was appear SiC, SiO_2, $Al_2O_3 \cdot SiO_2$ and $C_{19}Y_{15}$ pattern. Also, SiC particle density was most large in SiC_f/SiC composites.

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