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Effect of Fabrication Process on Internal Friction and Elastic Modulus of SiC/SiC Composites †

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Abstract

Silicon carbide (SiC) based fiber reinforced SiC composites have been fabricated by various methods which are, for example, polymer impregnation and pyrolysis (PIP), hot press (HP) and chemical vapor infiltration (CVI) processes. It is also well known that there are many differences in the density, the elastic modulus and the strength of composites made by those methods. In this paper, the effect of fabrication process on the internal friction and elastic modulus of SiC/SiC composites was studied. From the temperature dependences of internal friction and elastic modulus of composites made by PIP, HP and CVI processes, a suitable method to make the high-damping SiC-based composite with a good elastic property is considered to be the CVI process. In the case of the composite produced by the CVI process at 1323 K, a stable peak of internal friction due to the grain boundary relaxation was obtained at about 1200 K. The temperature of this peak moved to the lower side after heat treatment at 1773 K. Also, it was found that the peak was not affected by the property of the SiC based fiber but by the microstructure of the matrix.

KEY WORDS: (Internal Friction) (Elastic Modulus) (Heat Treatment Temperature) (Temperature Dependence) (Silicon Carbide) (Composite)

1. Introduction

Silicon carbide (SiC) is one of the typical nonoxide ceramics and has superior properties to the oxide ceramics. In particular, SiC is a promising candidate as a high-temperature structural material, due to its potential of excellent high-temperature strength and good heat and oxidation resistance in an oxidizing environment at the temperature up to 1773 K.

SiC fiber reinforced SiC composites (SiC/SiC composites) are also known to be superior in strength-to-weight and stiffness-to-weight ratio, and as a result of R & D efforts in the field of aero space and advanced energy, they are expected as high heat flux component materials, which are, for example, the parts of future gas turbines and ceramic engines[1-5]. It is very difficult, however, to obtain the high-damping property since SiC based composites have much porosity and a complex structure. Moreover, as the fabrication process of SiC based composites, there are various methods, which are, for instance, polymer impregnation and pyrolysis (PIP), hot press (HP) and chemical vapor infiltration (CVI) processes[6-9]. It is well known that there are many differences between the density, the elastic modulus and the strength of the composites made by those methods.

In this study, the effect of fabrication process on the internal friction and elastic modulus of SiC/SiC composites was examined. Another objective of this research was to develop SiC/SiC composites with a stable peak of internal friction up to 1373 K by controlling the condition of CVI process and thermal annealing because SiC/SiC composites fabricated by CVI method have a higher elastic modulus and strength.

2. Experimental

In this research, four types of SiC fibers and three kinds of manufacturing method for SiC based composites were employed. The characteristics of the fibers used are shown in **Table 1**. In the case of CVI method, Nicalon CG, Hi-Nicalon and Tyranno SA fibers were used as reinforcement and the composites were made with seven sheets of flat woven fabrics stacked under pressure. The precursor and carrier gases of raw

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Table 1 Characteristics of SiC fibers used.

Properties		Nicalon CG	Hi-Nicalon	Tyranno LoxM	Tyranno SA
Fiber Diameter	(μm)	14	14	11	10
Number of Filaments	(fil/yarn)	500	500	800	500
Tensile Strength	(GPa)	3.0	2.8	3.5	2.8
Tensile Modulus	(GPa)	220	270	190	420
Elongation	(%)	1.4	1.0	2.0	0.7
Density	(g/cm^3)	2.55	2.74	2.60	3.24
Chemical Composition	Si (wt.%)	56.6	62.4	53.3	67.8
	C	37.1	37.1	31.2	31.3
	O	11.7	0.5	13.0	0.3
	other	-	-	Ti 2.5	Al 2.0

Table 2 Conditions of manufacturing and heat treatment of composites used.

Specimen Name	Process	Fiber Type	Manufacturing Temp. (K)	Annealing Condition	Density (g/cm^3)
CV1248H	CVI	Hi-Nicalon	1248	-	2.18
CV1323N	CVI	Nicalon CG	1323	-	2.49
CV1723NA	CVI	Nicalon CG	1323	1723K x 1hr.	2.61
CV1773NA	CVI	Nicalon CG	1323	1773K x 1hr.	2.52
CV1323H	CVI	Hi-Nicalon	1323	-	2.52
CV1323T	CVI	Tyranno SA	1323	-	2.51
PIP1323H	PIP	Hi-Nicalon	-	1323K x 1hr.	1.92
HP2173T	HP	Tyranno LoxM	2173	-	2.48

material were $C_2H_5Cl_3Si$ and H_2, and the deposition of SiC on the sheet was accomplished by pyrolyzing the organic matrix for 100 hours at 1248 or 1323 K according to the following equation. The fabricated SiC/SiC composite was a 120 mm diameter disk and its fiber volume fraction was about 30 %.

$$C_2H_5Cl_3Si + H_2 \rightarrow SiC + CH_4 + 3HCl \qquad (1)$$

For the PIP method, the seven sheets of flat woven fabrics of Ni-Nicalon were stacked and the processes of the impregnation of polycarbosilane, the drying and the pyrolysis were repeated seven times. The pyrolysis was carried out at 1323 K for 1 hour and the fiber volume fraction was about 30 %.

In the HP method, after oxidizing the surface of Tyranno LoxM fibers, the sheets of flat woven fabrics of the oxidized Tyranno were stacked and HP process was conducted at 2173 K in Ar with a 500 atm pressure. The fiber volume fraction was 85 %.

The conditions of manufacturing and heat treatment are summarized into **Table 2**. After surface polishing, the specimen size for measuring the internal friction and the elastic modulus was $1^T \times 4^W \times 60^L$ mm^3. Also, in order to examine the effect of high temperature annealing, the specimen was heat treated at 1723 or 1773 K for 1 hour in a vacuum.

The internal friction measurement was carried out utilizing the lateral vibration type internal friction apparatus by electrostatic capacitance method[10]. The elastic modulus was determined from the resonant frequency and the internal friction values were obtained from free damping behavior under lateral vibration with a resonant frequency of about 2500 Hz. The temperature of this measurement ranged from 293 K (room temperature) to 1373 K and the vacuum quality at room temperature was about 2×10^{-5} Pa.

3. Results and Discussions

3.1 Effect of manufacturing methods

In order to study the effect of manufacturing methods on the internal friction and elastic modulus, the temperature dependences of internal friction and elastic modulus of SiC/SiC composites fabricated by PIP, HP and CVI processes are shown in **Fig.1**. In the case of PIP method, the internal friction was larger and the elastic modulus was smaller than the others. In this method, to densify the composites, several cycles of polymer impregnation are needed. The composites, however, still have much porosity and many micro cracks and it is very difficult to increase the density more than 2.00 Mg/m^3. So these micro cracks are considered to be a reason for the large internal friction and the small elastic modulus.

In contrast, the internal friction of the composite made by HP method remained stably low up to the high temperature region. The elastic modulus slightly

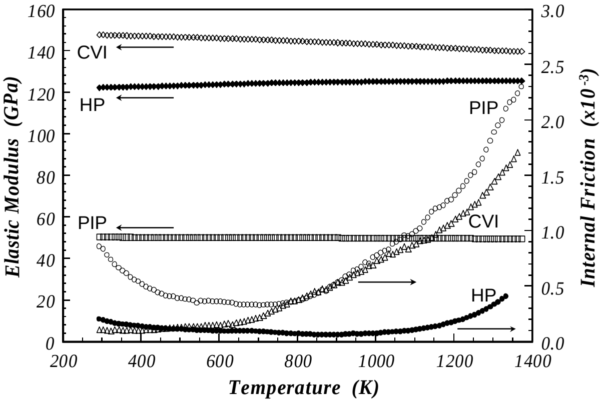


Fig.1 Effect of manufacturing method on elastic modulus and internal friction of SiC/SiC composites.

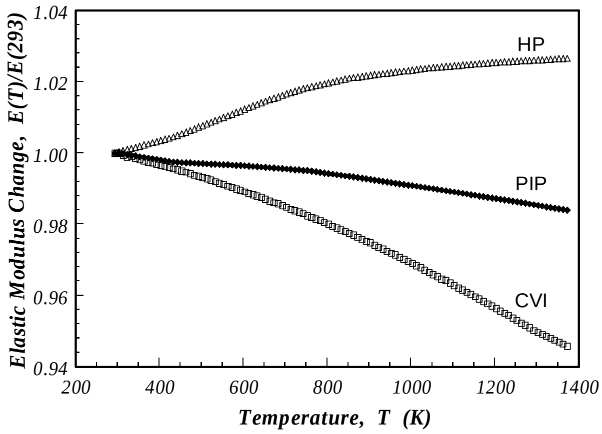


Fig.2 Effect of manufacturing method on normalized elastic modulus of SiC/SiC composites.

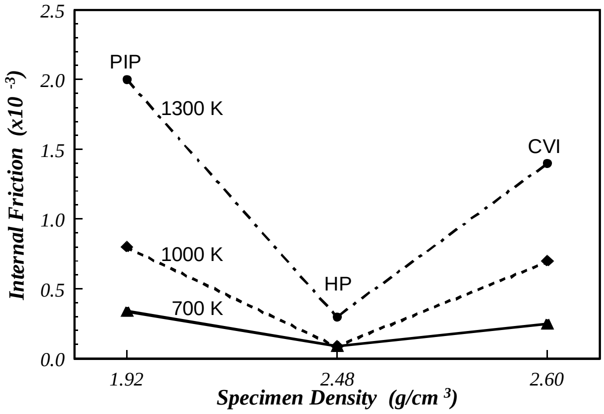


Fig.3 Effect of specimen density on temperature dependence of internal friction in SiC/SiC composites.

increased with increasing temperature. In this HP process, a high load was applied to make the matrix from SiO_2 obtained by oxidizing the fiber surface. Then, although the density of composites became higher, the cross sectional shape of fiber changed from a circle to a polygon and the fiber volume fracture became larger than the theoretical value. So it is considered that there is a large residual strain in the composite and the elastic modulus is affected by this residual strain.

For CVI process, the elastic modulus decreased with increasing temperature although the elastic modulus in all the measuring temperature region was much larger than the others. In this method, SiC is produced by a chemical reaction with high temperature and is deposited on the fiber surface. Thus, the SiC matrix is highly densified without residual strain. So the high density without residual strain is considered to be a reason for the negative temperature dependence of elastic modulus. On the other hand, the internal friction was relatively high, although this result was smaller than that of PIP method in all the measuring temperature area.

In order to identify the effect of temperature on the elastic modulus, the elastic modulus change is normalized and presented in **Fig.2**. With increasing temperature, the normalized elastic moduli of the composites manufactured by HP, PIP and CVI processes increased, slightly decreased and largely decreased, respectively. This behavior was the same as our previous result for C/C composites[11-13)]. In this result, it was reported that the temperature dependence of the elastic modulus of C/C composites was positive when the microstructure was turbostratic and there was large residual strain. The dependence, however, changed from positive to negative with increasing the degree of carbonization and with decreasing the residual strain by the heat treatment at high temperature. So it was found that the residual strain increased in the order of HP, PIP and CVI methods.

The internal friction at the same temperature is compared and summarized into **Fig.3**. From these results of both the internal friction and the elastic modulus, it was concluded that a suitable method to make a high-damping SiC base composite with a good elastic property might be the CVI method.

3.2 Effect of manufacturing temperature in CVI method

The temperature dependences of internal friction and elastic modulus of CV1248H specimen are shown in **Fig.4**. In the 1st run, which was carried out up to 800 K, a peak I of the internal friction was obtained. After 1st run, the specimen was cooled back to room temperature keeping a vacuum quality, then a 2nd run was carried out up to 1373 K. In the 2nd run, the peak I disappeared. From our previous researches for C/C composites[10,11)], this peak seems to be caused by the gas desorption behavior from the inside and the surface of specimen. Namely, for most specimens, the peak I could be measured in the 1st run. In the 2nd run, the internal friction monotonically increased with increasing the temperature. Since this result was repeated, this was

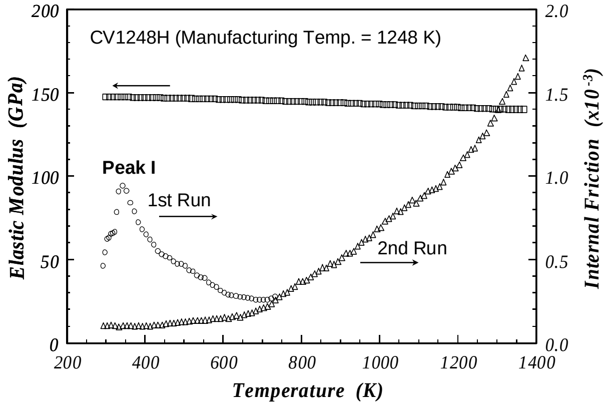


Fig.4 Temperature dependence of elastic modulus and internal friction in SiC/SiC_{CVI} composite.

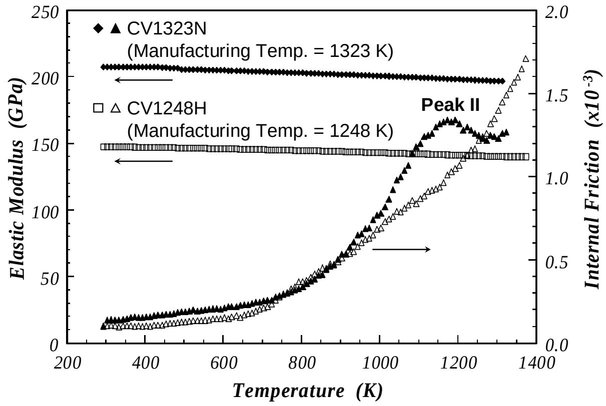


Fig.5 Effect of manufacturing condition on elastic modulus and internal friction of SiC/SiC_{CVI} composites.

found to be a true property of the composite. On the other hand, the elastic modulus slightly increased with increasing test temperature.

For a comparative study, the results of CV1248H and CV1323N are shown in **Fig.5**. In the case of CV1323N, a peak II was obtained at about 1200 K and the height of this peak was 1.34 x 10^{-3}. In the higher temperature region, the internal friction increased again. Since there were two kinds of differences between CV1248H and CV1323, which were the difference of manufacturing temperature and the difference of fiber type, it was very important to reveal those effects on this peak II. So, first of all, the effect of fabrication temperature was examined in the next section.

On the other hand, the elastic modulus of CV1323N became about 30 % higher than that of CV1248H. This increment seems to be caused by a 14 % rise in the volume density and a 23 % increase in the tensile modulus of fiber.

3.3 Effect of thermal annealing in CV1323N

In order to examine the effect of manufacturing temperature on the peak II of internal friction, CV1323N

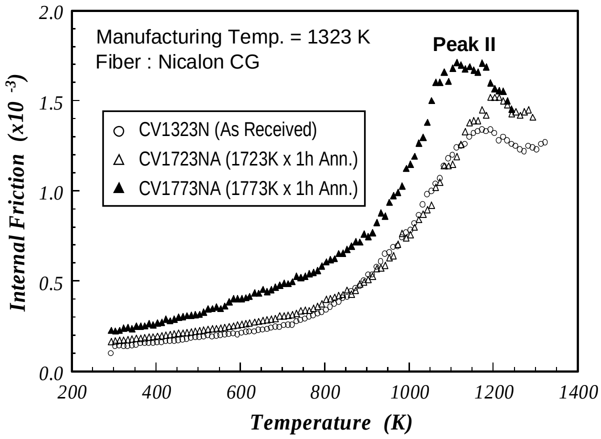


Fig.6 Effect of heat treatment temperature on internal friction of SiC/SiC_{CVI} composites.

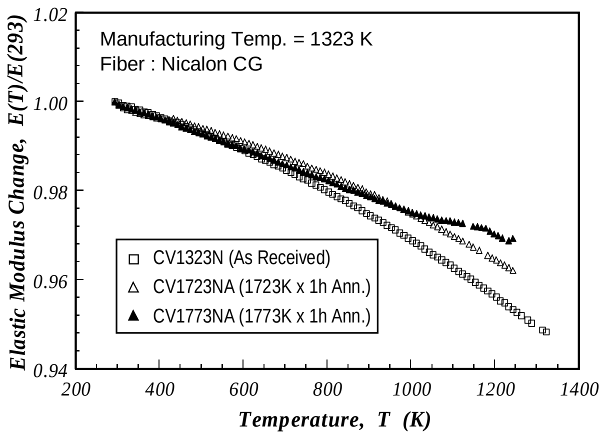
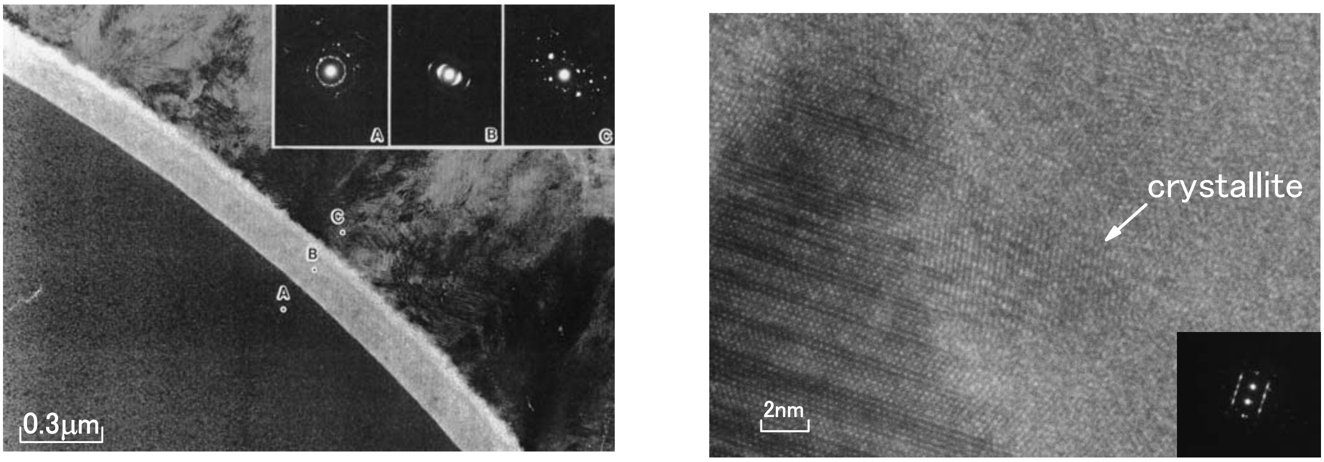


Fig.7 Effect of heat treatment temperature on normalized elastic modulus of SiC/SiC_{CVI} composites.

was thermally annealed at 1723 or 1773 K for 1 hour, because a recrystallization of SiC starts at about 1773 K. The temperature dependences of internal friction in those specimens are shown in **Fig.6**. Although the result of the specimen heat treated at 1723 K (CV1723NA) was almost the same as that of the as received specimen (CV1323N), the height of the peak slightly increased. On the other hand, in the case of the composite annealed at 1773 K (CV1773NA), the internal friction in the heat-up process became higher than that of CV1323N and the height of the peak increased up to 1.71 x 10^{-3}. Moreover, the temperature of this peak shifted to the lower temperature side.

Figure 7 shows the temperature dependences of normalized elastic modulus. The decrement of normalized elastic modulus decreased with increasing heat treatment temperature. As mentioned in section 3.1, the matrix of CV1323N was produced by CVI method where SiC crystals were stacked keeping the atomic order with a fine atomic alignment. So the temperature dependence of normalized elastic modulus showed the negative behavior.



(a) Near interface between fiber and matrix (b) Magnified image of matrix

Fig.8 Characteristics of SiC fibers used.

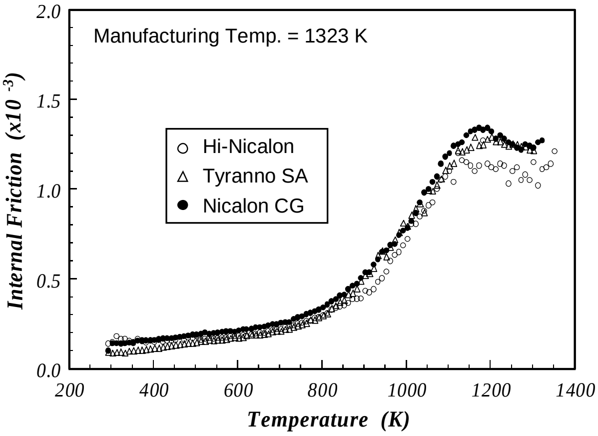


Fig.9 Effect of fiber type on internal friction of SiC/SiC_{CVI} composites.

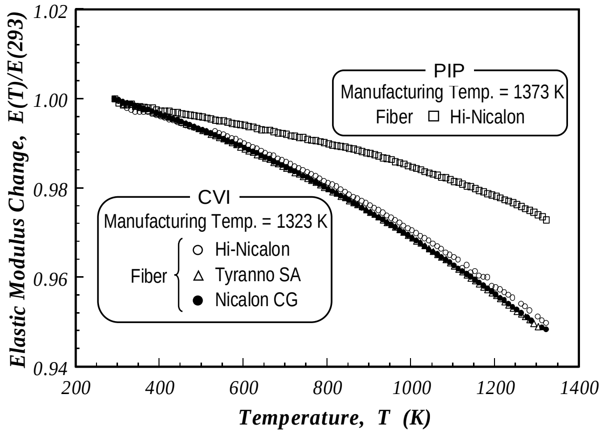


Fig.10 Effect of fiber type on normalized elastic modulus of SiC/SiC composites.

TEM images in CV1323N are shown in **Fig.8**. In Fig.8(a), the points of A, B and C mean the SiC fiber, a carbon coating on the fiber surface and the matrix, respectively[14]. Figure 8(b) shows the magnified image of the matrix. The partial formation of crystallites was observed in Fig. 8(b) where the size ranged from 5 to several tens of nm.

For CV1723NA, the relocations of atoms with growing crystallites would come from the thermal annealing. So the reason for the decrease of decrement in the normalized elastic modulus is considered to be a low residual strain induced by the relocations of atoms.

In the case of CV1773NA, a change in the temperature dependence of normalized elastic modulus was obtained at about 1000 K and this temperature was corresponding to the peak II of internal friction. Above 1200 K, the tendency of normalized elastic modulus change moved back to the behavior of the initial decrement. The growth of crystallites would occur in the wide range of the matrix by the heat treatment at 1773 K. So the shifted peak II seems to be caused by the slip of grains at the grain boundary induced by a resonance at the temperature of the peak. In other words, it is considered that the peak II of internal friction would be caused by the grain boundary relaxation and then the temperature dependence of normalized elastic modulus also might change around the peak II. Moreover, Goldsby has also reported the same peak of internal friction in SiC/SiC composites[15].

3.4 Effect of fiber type

In order to examine the effect of fiber type on the internal friction of composites, Nicalon CG, Hi-Nicalon and Tyranno SA were employed as the reinforcement. The temperature dependences of internal friction in those fiber reinforced SiC composites fabricated by the CVI method are shown in **Fig.9**. For all cases, those dependences showed a similar behavior and the temperature for the peak was also the same. Although the tensile strengths of fibers varied from 2.8 to 3.0 GPa and the density of Tyranno SA, 3.24 Mg/m^3 was about 20 % higher than others, the densities of composites were almost the same, which was about 2.5 Mg/m^3. So it was found that the temperature dependence of internal friction was not controlled by the property of fiber but by the microstructure of matrix.

The temperature dependences of normalized elastic modulus are shown in **Fig.10**. These results fitted in with each other and it was found that the reason of these dependences seems to be the same. The result of CV1248H also agreed with those dependences, although the manufacturing temperature of CV1248H was different from those of the others. On the other hand, the result of PIP1323H fabricated by the PIP method was very different from the others as shown in Fig.10. So it was found that the effect of fabrication process on the temperature dependence of elastic modulus was very large. In the cases of composites manufactured by the CVI process, the temperature dependences of normalized elastic modulus showed the same behavior although the tensile strength, the density and the tensile modulus of fibers were different from each other. Therefore, these dependences are considered to be governed by the matrix due to small micro cracks in the matrix. Moreover, since the composites fabricated by the PIP method might have many micro cracks in the matrix, the dependence is considered to be controlled by residual strain in the matrix.

From these results, it was found that the internal friction was largely affected by the microstructure of the matrix and not by the fiber properties. Also, it is expected that a SiC/SiC composite may have more high-damping property by crystallizing the matrix.

4. Conclusions

The effects of manufacturing methods of SiC based composites on their internal friction and elastic modulus were studied and the conclusions can be summarized as follows.

(1) From the results of the internal friction and elastic modulus of composites fabricated by the PIP, HP and CVI methods, a suitable method for making a high-damping SiC base composite is considered to be the CVI method.

(2) By using the CVI process with a manufacturing temperature of 1323 K, a stable peak of internal friction due to the grain boundary relaxation was obtained at about 1200 K.

(3) The stable peak shifted to the lower temperature side and the height of the peak increased following the heat treatment at 1773 K.

(4) The internal friction of composite fabricated by the CVI process was found to be largely affected by the microstructure of the matrix and not by the fiber properties.

(5) SiC/SiC composite is expected to have more high-damping property by crystallizing the matrix.

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