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Thermal diffusivity/conductivity of Tyranno SA fiber- and Hi-Nicalon Type S fiber-reinforced 3-D SiC/SiC composites

R. Yamada *, N. Igawa, T. Taguchi

Japan Atomic Energy Research Institute, Tokai-mura, Naka-gun, Ibaraki-ken 319-1195, Japan

Abstract

Thermal diffusivity measurements for 3D-SiC/SiC composites reinforced with either Tyranno SA or Hi-Nicalon Type S fibers were performed by laser flash method. The 3D weaves with several X:Y:Z fiber configurations were directly woven from each fiber bundle, and the SiC composites were fabricated by either isothermal CVI or PIP/CVI process. For the 3D-Tyranno SA-CVI and -PIP/CVI composites the measured thermal conductivity values were 40–50 W/mK and 35–40 W/mK at RT, respectively, and about 24 and 18 W/mK at 1000 °C, respectively. For the 3D-Hi-Nicalon Type S-CVI composites, the values were about 36 and 20 W/mK for RT and 1000 °C, respectively. These fairly high thermal conductivity values were attributable to their high fiber and matrix densities. The influence of the 3D-fiber configuration on the overall composite thermal conductivity was observed in the temperature range less than 600 °C.

1. Introduction

SiC fiber-reinforced SiC composites (SiC/SiC) have potential for a structural material for the blanket wall of advanced fusion reactors [1–4]. Under heavy thermal loads of the reactors, it is essential to reduce thermal stresses in the wall so that SiC/SiC composites are expected to have as high a thermal conductivity (K_c) as possible [1,2]. When conventional fibers such as Nicalon fiber were used, K_c-values of SiC/SiC composites fabricated with chemical vapor infiltrated (CVI) or polymer impregnated and pyrolized (PIP) process were not high enough, around 10 W/mK for CVI or less than that for PIP even at RT [3–7]. It is known that recently developed SiC fibers, i.e., Tyranno SA and Hi-Nicalon Type S, have achieved higher fiber thermal conductivity (K_f) than previous fibers [8–11]. We have already reported that 3D-SiC/SiC composites reinforced with Tyranno SAC preforms, which were obtained by sintering 3D-weaves of amorphous Si–Al–C–O fiber, exhibited K_c-values higher than the conventional ones due to the high fiber density and crystallized SiC structure of the sintered preforms [12]. The reason for using the amorphous Si–Al–C–O fiber that is the starting material of sintered SA fiber [8,9] was that it was easy to weave the 3D preforms due to the low stiffness and tensile modulus.

As for the next step, in this study, we have employed 3D-SiC preforms directly woven with Tyranno SA fiber and measured their thermal diffusivity, and compared their K_c-values with the previous ones. In addition, we also have made 3D-SiC/SiC composites with 3D fabric preforms directly woven with Hi-Nicalon Type S fiber and compared their K_c-values to those of the Tyranno SA-SiC/SiC composites.

2. Experimental

2.1. Specimen preparation

3D-preforms were directly woven with either Tyranno SA fiber (10 μm diameter) or Hi-Nicalon Type S fiber (13 μm diameter) by Shikibou Ltd., Japan. The X:Y:Z fiber configurations of 3D-preforms were selected as 1:1:0.09, 1:1:0.2, 1:1:0.45, and 1:1:1.23 for Tyranno

* Corresponding author. Tel.: +81-29 282 5403; fax: +81-29 282 6556.
E-mail address: reiji@popsvr.tokai.jaeri.go.jp (R. Yamada).

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SA fiber, and 1:1:0.22, 1:1:0.44, and 1:1:1.2 for Hi-Nicalon Type S fiber, where the Z-direction was taken through the thickness of the specimen. For fabricating 3D-SiC/SiC composites with Tyranno SA fiber, isothermal CVI (performed at Mitsui Engineering & Shipbuilding Co., Ltd., Japan) and PIP/CVI (performed at Kawasaki Heavy Industries Ltd., Japan) were adopted.

For the PIP/CVI composites, the role of CVI was to protect the preforms with CVD SiC layer from chemical attacks of reactive species produced in PIP processing. The CVD thicknesses on the fiber estimated from weight gains were rather thick (3.3–3.8 μm) compared to the previous study (1.7–2.4 μm) in which we noted it as PIP [12]. In this study, we accurately note it as PIP/CVI. In addition to increasing CVI thickness, the number of repeated PIP process was increased to 10 times, whereas the previous one was 6 times. These difference resulted in making SiC/SiC composite densities higher, i.e., about 2.71–2.81 g/cm^3. The porosity of the PIP/CVI composites decreased from 80% to 30% by CVI processing and from 30% to <10% by 10 times PIP processing. The fiber volumes were about 34% for all PIP/CVI composites. A 100–150 nm carbon coating was applied to the SA preforms prior to PIP/CVI process to improve mechanical properties.

For the CVI SiC/SiC composites, the Tyranno SA specimen densities ranged from 2.82–2.88 g/cm^3 and their fiber volumes were about 34%. The carbon coating was also applied to the SA preforms prior to CVI, whose thicknesses were estimated to be 0.2–0.3 μm. The Hi-Nicalon Type S specimen densities were above 2.9 g/cm^3, and their fiber volumes were about 40%. A nominal 120 nm carbon was directly coated on the Hi-Nicalon Type S fiber prior to weaving the preforms.

2.2. Thermal diffusivity measurement

The specimen size was nominally 10 mm in diameter and 2.5–3.5 mm in thickness. Through-the-thickness thermal diffusivity was measured in vacuum using the laser flash method (TC2000, Shinku-Riko, Inc.). The details of the measurement were described in the previous paper [12].

When the incident laser power for thermal diffusivity measurement is not uniformly distributed in the lateral direction, a measured thermal diffusivity value depends on specimen thickness even if the specimen is homogeneous. When the power distribution has a peak in the center, a thermal diffusivity value becomes larger than the true value. Using reference specimens of pure Al and SUS304, the ratio of the reference values of thermal diffusivity to the values measured in our laser flash system at RT was determined as a function of specimen thickness. It is revealed that the ratio was nearly 80% for the severest case, i.e., 2–3 mm specimen thickness. We have corrected each measured thermal diffusivity value by multiplying the ratio as a function of specimen thickness.

3. Results and discussion

In Fig. 1 are shown the measured composite thermal diffusivity and conductivity values for 3D-Tyranno SA-CVI SiC/SiC composites. The thermal diffusivity values increased with increasing Z-directional fiber configuration, namely $X{:}Y{:}Z = 1{:}1{:}0.09$, 1:1:0.2, 1:1:1.23 in order, except $X{:}Y{:}Z = 1{:}1{:}0.45$. The tendency became evident in lower temperature region. The K_c-values also exhibited a similar dependence. It should be noted that although the $X{:}Y{:}Z = 1{:}1{:}0.09$ specimen had the highest specimen density, it had the lowest values of the thermal diffusivity and conductivity. It is also noticeable that the $X{:}Y{:}Z = 1{:}1{:}0.45$ and $X{:}Y{:}Z = 1{:}1{:}1.23$ specimen, which had similar specimen densities, exhibited a large difference in their thermal diffusivity/conductivity. In Fig. 2 are given the measured composite thermal diffusivity and conductivity values for 3D-Tyranno SA-PIP/CVI SiC/SiC composites. Similar to the result of the CVI composites, the $X{:}Y{:}Z = 1{:}1{:}0.09$ specimen exhibited the lowest values, although it had the highest density. The $X{:}Y{:}Z = 1{:}1{:}1.23$ specimen with the lowest density had the lowest values when the $X{:}Y{:}Z = 1{:}1{:}0.09$ specimen was excluded.

The PIP/CVI composites reached approximately 80% of the K_c-values of the CVI ones. These are fairly high in comparison with the previous PIP SiC/SiC composites whose densities ranged 2.18–2.57 g/cm^3 [12]. The increase in both the CVD SiC layer thickness and the number of PIP cycles caused the specimen densities to be higher (2.71–2.81 g/cm^3), and resulted in higher thermal

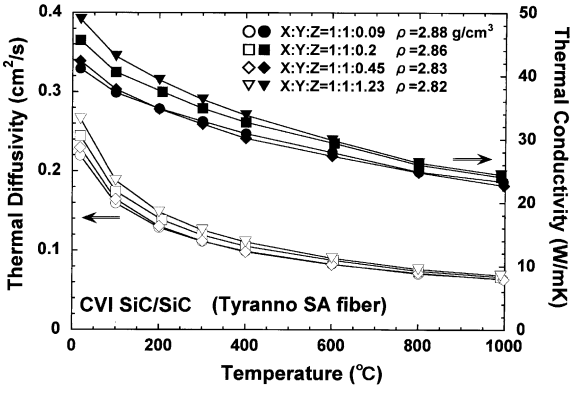

Fig. 1. Through-the-thickness thermal diffusivity/conductivity for CVI SiC/SiC composites reinforced with 3D-Tyranno SA weaves: ○ □ ◇ ▽ thermal diffusivity; ● ■ ◆ ▼ thermal conductivity.

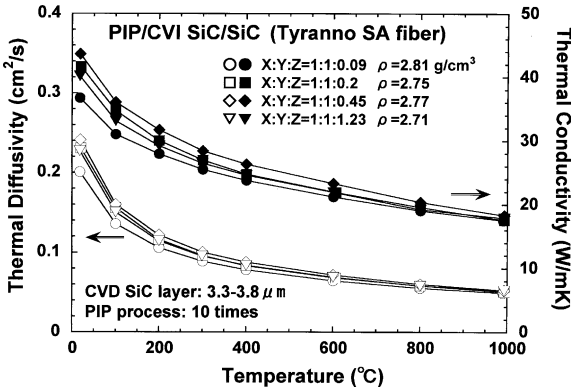

Fig. 2. Through-the-thickness thermal diffusivity/conductivity for PIP/CVI SiC/SiC composites reinforced with 3D-Tyranno SA weaves: ○ □ ◇ ▽ thermal diffusivity; ● ■ ◆ ▼ thermal conductivity.

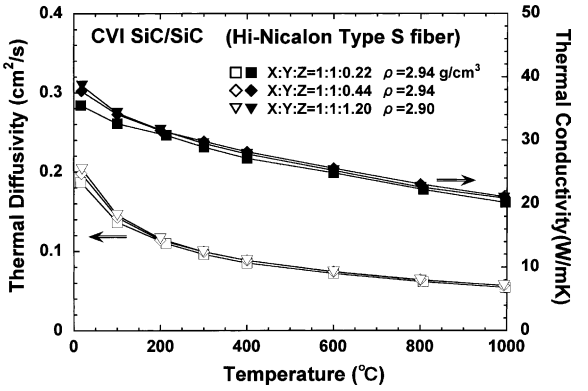

Fig. 4. Through-the-thickness thermal diffusivity/conductivity for CVI SiC/SiC composites reinforced with 3D-Hi-Nicalon Type S weaves: □ ◇ ▽ thermal diffusivity; ■ ◆ ▼ thermal conductivity.

diffusivity and conductivity than the previous ones. The K_c-values of SiC/SiC composites at RT obtained from the present and the previous study are given in Fig. 3. It is indicated that the K_c-values increase with increasing specimen density and that most of the measured K_c-values increase with increasing ratio of Z-directional fiber configuration when specimen densities are above 2.71 g/cm^3. The CVI composites reinforced with Tyranno SAC weaves had the highest K_c-values, where the SAC preforms, which were obtained by sintering 3D-weaves of amorphous Si–Al–C–O fiber at a temperature exceeding 1900 °C, have a better thermal conductive condition than the Tyranno SA weaves directly woven from SA fiber. In addition, the carbon coating was applied to the SA weaves at <1300 °C, which leads to a poor quality of pyrolytic carbon. The graphitization of pyrolytic carbon generally starts at >1500 °C [13,14]. If a pyrolytic carbon with low thermal conductivity as well as imperfect thermal contact at the interface exists, they would worsen the overall thermal conductivity [15,16].

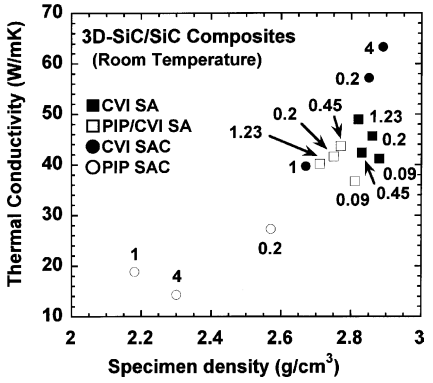

Fig. 3. Measured thermal conductivity values at RT vs. specimen density. Specimens: ■ CVI-SiC/SiC composites with 3D-Tyranno SA weaves, □ PIP/CVI-SiC/SiC composites with 3D-Tyranno SA weaves, ● CVI-SiC/SiC composites with 3D-Tyranno SAC weaves, ○ PIP-SiC/SiC composites with 3D-Tyranno SAC weaves. The numbers in the figure stand for Z values in the 3-D fiber configuration of $X{:}Y{:}Z = 1{:}1{:}Z$.

Fig. 4 illustrates the measured composite thermal diffusivity/conductivity values for 3D-Hi-Nicalon Type S-CVI SiC/SiC composites, whose specimen densities were higher than those of 3D-Tyranno SA-CVI SiC/SiC composites. The obtained K_c-values were about 80% of the Tyranno SA composites, and appeared higher than the expected since the K_f-value of Hi-Nicalon Type S is considerably lower than that of Tyranno SA (see Table 1). It can be ascribed to the difference in the matrix densities of two composites. Youngblood et al. have also reported that the Hi-Nicalon Type S-SiC composites with higher bulk density obtained higher K_c-values than Tyranno SA SiC composite [17]. The small effect of Z-directional fiber configuration on the K_c-values for the case of Hi-Nicalon Type S is also understandable because the fiber volumes are almost the same here and the matrix thermal conductivity (K_m) plays a more significant role in increasing K_c than K_f does. Since high density Hi-Nicalon Type S-CVI SiC/SiC composites have high K_c-values, they might be a good candidate material for advanced fusion reactors, since they also exhibit good radiation resistance [18].

There is no way to directly measure the K_m-values. We can estimate them by using simple models: When matrix and fiber bundle are assumed to be configured either in parallel or in series to the heat flow, K_c-values are given as

$$K_c = V_f K_f + (1 - V_f) K_m \text{ (parallel)}, \quad (1)$$

$$1/K_c = V_f/K_f + (1 - V_f)/K_m \text{ (series)}, \quad (2)$$

Table 1
Matrix thermal conductivity values estimated by the simple parallel or series model of heat flow using the measured composite thermal conductivity values and fiber volumes[a]

	Measured composite TC (W/mK)	Fiber TC (W/mK)[b]	Matrix TC (W/mK) by parallel model	Matrix TC (W/mK) by series model
Tyranno SA CVI				
(RT)	45	64	35	39
(1000 °C)	24	32	20	21
Tyranno SA PIP/CVI				
(RT)	38	64	25	31
(1000 °C)	18	32	11	15
Hi-Nicalon Type S CVI				
(RT)	36	18	48	108
(1000 °C)	20	9	27	107

[a] The fiber volumes were chosen 34% for Tyranno SA-CVI and Tyranno SA-PIP/CVI, and 40% for Hi-Nicalon Type S-CVI composites.

[b] The fiber thermal conductivity values at RT supplied by fiber producers from Ref. [8–11]. There are no data at 1000 °C so that a half of RT values were assumed.

where V_f is the fiber volume, and both porosity and volume of carbon layer were neglected since the former was <10% for both PIP/CVI and CVI composites, and the latter was estimated <2%. In Table 1 are given the K_m-values estimated from the measured K_c-values and the K_f-values specified by the fiber producers [8–11]. It appears that K_m calculated by parallel method is more consistent in Table 1. We have previously calculated thermal diffusivity values of SiC/SiC composites by using a finite-element method [12,19]. That method reveals that when using highly thermal conductive SiC fiber, such as Tyranno SA fiber, the Z-directional composite thermal diffusivity of SiC/SiC composites with a low K_m-value is increased with increasing either the total fiber volume or the Z-directional fiber volume [12] and that the 3-D fiber network with a high K_f-value could enhance the Z-directional heat flow via heat-piping [19]. The results of the parallel model and FEM calculations correspond to the experimental results that in 3D-SiC/SiC composites the component with higher thermal conductivity mainly contributed to the K_c through the Z-directional heat flow via the 3D-heat-pipe effect.

The K_c-values obtained in this study were fairly high (nearly 20 W/mK at 1000 °C), but still are not high enough when the degradation of K_c due to radiation damage is taken into account, which would lead to a decrease of probably 50% from unirradiated values at 1000 °C [5]. There may be a possibility that K_m-value would increase up to 60–70 W/mK at RT similar to the K_f-value of Tyranno SA fiber by means of increasing matrix density, but the increase of K_m-value at 1000 °C may not be highly expected. Reactor designs have revealed that the mechanical strength and thermal conductivity of the first wall are simply related [1,2]. Therefore, if the K_c-value cannot meet the design requirements, for example, an irradiated value of 15 W/mK for DREAM, which would correspond to 30 W/mK for unirradiated value, the design value for mechanical strength must be increased. The present allowable stress is 200 MPa [1,2], and may need to be increased 50%, i.e., to 300 MPa. When a safety factor of 2 is desired, then 600 MPa will be required for the tensile strength. In the near future, this value might be realized in some combinations of preforms and fabrication processing [18].

4. Summary and conclusions

The thermal diffusivity/conductivity of 3D-CVI and -PIP/CVI SiC/SiC composites was measured by a laser flash method. The preforms of the 3D-SiC weaves were directly woven with Tyranno SA fiber or Hi-Nicalon Type S fiber. For the Tyranno SA weaves the 3D-fiber configurations (X:Y:Z) were 1:1:0.09, 1:1:0.2, 1:1:0.45, 1:1:1.23, and for the Hi-Nicalon Type S ones they were 1:1:0.2, 1:1:1, 1:1:4.

The specimen densities slightly decreased with increasing Z-directional fiber fraction. These decreases diminished both thermal diffusivity and thermal conductivity values, especially for larger Z values. When densities are similar, specimens with larger Z values appear to have higher thermal diffusivity/conductivity, especially in the low temperature range (less than 600 °C). From the viewpoint of thermal properties, therefore, SiC/SiC composite with high density and large Z-directional fiber fraction would be preferable when the mechanical properties are not taken into account.

The thermal conductivity value required by fusion designs is still high when the degradation of thermal conductivity due to radiation damage is taken into account. Both thermal and mechanical properties need to be optimized to achieve the fusion designs.

References

[1] S. Nishio, S. Ueda, I. Aoki, R. Kurihara, T. Kuroda, H. Miura, T. Kunugi, Y. Seki, T. Nagashima, M. Ohara, J. Adachi, S. Yamazaki, I. Kawaguchi, T. Hashimoto, K. Shinya, Y. Murakami, H. Takase, T. Nakamura, Fus. Eng. Design 41 (1998) 357.

[2] S. Ueda, S. Nishio, Y. Seki, R. Kurihara, J. Adachi, S. Yamazaki, J. Nucl. Mater. 258–263 (1998) 1589.

[3] L.L. Snead, R.H. Jones, A. Kohyama, P. Fenici, J. Nucl. Mater. 233–237 (1996) 26.

[4] A. Hasegawa, A. Kohyama, R.H. Jones, L.L. Snead, B. Riccardi, P. Fenici, J. Nucl. Mater. 283–287 (2000) 128.

[5] D.J. Senor, G.E. Youngblood, C.E. Moore, D.J. Trimble, G.A. Newsome, J.J. Woods, Fus. Technol. 30 (1996) 943.

[6] R.H. Jones, D. Steiner, H.L. Heinisch, G.A. Newsome, H.M. Kerch, J. Nucl. Mater. 245 (1997) 87.

[7] R. Yamada, T. Taguchi, N. Igawa, J. Nucl. Mater. 283–287 (2000) 574.

[8] T. Nakayasu, M. Sato, T. Yamamura, K. Okamura, Y. Katoh, A. Kohayama, Ceram. Eng. Sci. Proc. 20 (4) (1999) 301.

[9] T. Ishikawa, S. Kajii, T. Hisayuki, Ceram. Eng. Sci. Proc. 21 (4) (2000) 323.

[10] A. Urano, J. Sakamoto, M. Takeda, Y. Imai, Ceram. Eng. Sci. Proc. 19 (3) (1998) 55.

[11] A. Urano, A. Saeki, M. Takeda, A. Yokoyama, Ceram. Eng. Sci. Proc. 20 (3) (1999) 85.

[12] R. Yamada, N. Igawa, T. Taguchi, S. Jitsukawa, J. Nucl. Mater. 307–311 (2002) 1215.

[13] B.T. Kelly, The thermal conductivity of graphite, in: Chemistry and Physics of Carbon, vol. 5, Marcel Dekker, New York, 1969, p. 119.

[14] D.B. Fischbach, The kinetics and mechanism of graphitization, in: Chemistry and Physics of Carbon, vol. 7, Marcel Dekker, New York, 1971, p. 1.

[15] H. Bhatt, K.Y. Donaldson, D.P.H. Hasselman, R.T. Bhatt, J. Am. Ceram. Soc. 75 (1992) 334.

[16] G.E. Youngblood, D.J. Senor, R.H. Jones, J. Nucl. Mater. 307–311 (2002) 1120.

[17] G.E. Youngblood, D.J. Senor, R.H. Jones, in: 15th Topical Meeting on the Technology of Fusion Energy, Washington DC, 12–13 April, 2002; Nuclear Technol., submitted for publication.

[18] R.H. Jones, L. Giancarli, A. Hasegawa, Y. Katoh, A. Kohyama, B. Riccardi, L.L. Snead, W.J. Weber, J. Nucl. Mater. 307–311 (2002) 1057.

[19] R. Yamada, N. Igawa, Taguchi, in: Advanced SiC/SiC Ceramic Composites: Developments and Applications in Energy Systems, Ceramic Transactions, vol. 144, The American Ceramic Society, 2002, p. 289.