Available online at www.sciencedirect.com
ScienceDirect
Journal of Nuclear Materials 367–370 (2007) 725–729
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Mechanical properties of SiC/SiC composite with magnesium–silicon oxide interphase

N. Igawa *, T. Taguchi, R. Yamada, Y. Ishii, S. Jitsukawa

Japan Atomic Energy Agency, Tokai-mura, Ibaraki-ken 319-1195, Japan

Abstract

A SiC fiber reinforced SiC composite with magnesium–silicon-based oxide interphase was fabricated by the chemical vapor infiltration process and its mechanical properties were evaluated. An interphase was prepared on the advanced SiC fiber, Tyranno SA, using the alkoxide method. The tensile strengths of SiC fiber with the Mg–Si–O coating retained at 85% or higher compared to uncoated-unheated fiber after heating below the 1300 °C, while strength were slightly degraded to 80% after heating at 1400 °C. The composite showed ductile failure behavior and the fiber pull-out effect was observed in the fracture surface of the composite.

1. Introduction

Silicon carbide (SiC) exhibits excellent high temperature mechanical properties, dimensional and thermodynamic stability, thermal conductivity, and low induced radioactivity by neutron irradiation. Therefore SiC is considered one of the candidate structural materials for fusion reactors for applications such as first wall and blanket [1–3]. Because the monolithic SiC ceramic is a brittle material, continuous SiC fiber reinforced SiC composites (SiC/SiC), which exhibit high toughness with a pseudo-ductile failure mode, are of more interest for certain fusion reactor designs. The interphase between SiC fibers and SiC matrix is very important to the mechanical properties of SiC/SiC; the roles of the interphase are to allow debonding between fiber and matrix during deformation to branch the cracks to increase fracture toughness, and to prevent chemical reactions between fiber and matrix during processing. Several interphases were investigated in previous studies [4–9] with BN one of the best materials to allow for fiber pull-out. For fusion reactor components, however, BN interphase is not favorable because of its chemical reactivity with oxygen and water, and its residual radioactivity after fusion-neutron irradiation [10]. Oxide interphases become potential candidates to improve those properties since advanced SiC fibers have been produced, such as Hi-Nicalon Type S [11] and Tyranno SA [12] which show the oxidation resistance and excellent performance under neutron irradiation as well as superior mechanical and thermal properties [13–15].

In this study, we investigated a magnesium–silicon-based oxide interphase for SiC/SiC because

---

* Corresponding author. Tel.: +81 29 282 6099; fax: +81 29 284 3909.

E-mail address: igawa.naoki@jaea.go.jp (N. Igawa).

0022-3115/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnucmat.2007.03.091

of its thermal stability under the mild oxidizing environment and a low induced radioactivity under the 14-MeV fusion-neutron environment. We fabricated the Mg–Si–O coating on an advanced SiC fiber, Tyranno SA, using the alkoxide method. This coating is expected to provide a weak interphase due to the coefficient of thermal expansion mismatch between the coating and SiC. Tensile tests for the coated fiber were carried out to estimate the interaction between SiC fiber and the interphase. The SiC/SiC composite with the Mg–Si–O interphase was fabricated by the chemical vapor infiltration method and the mechanical properties of the composite were evaluated by the bending test.

2. Experimental procedure

The SiC fiber used in the present work was Tyranno SA (Ube Industries, Ube, Japan). The fiber diameter and tensile strength of Tyranno SA are 7.5 μm and 2.5 GPa, respectively [12].

The flow chart of the Mg–Si–O coating process used in this study is shown in Fig. 1. The coating solution was prepared by dissolving magnesium ethoxide, $Mg(OC_2H_5)_2$ and tetraethyl orthosilicate, $Si(OC_2H_5)_4$ (Wako Pure Chemical Industries, Ltd., Osaka, Japan) into 2-propanol. The magnesium and silicon concentrations of the coating solution were 0.50 and 0.25 mol/dm^3, respectively. The SiC fibers were dipped into the coating solution and hydrolyzed alkoxides at room temperature, under ca. 70%-humidity for 24 h. The dip-and-hydrolysis process was repeated 5 times. Finally the fibers were heated up to 1400 °C in air. The coating phase was analyzed using an X-ray diffractometer (Horizontal-model No. 3100R, MAC Science Co., Ltd., Yokohama, Japan). Cu Kα radiation of 40 kV and 20 mA was used with a step-scanning technique in the 2θ range from 25° to 80°. The tensile strength of the coated fibers was also measured at room temperature by a single filament method using an Instron-type tensile testing machine (model RTC-1150A, A&D Co., Ltd., Tokyo, Japan). The specimen gauge length was 25 mm and the cross-head speed was 0.5 mm/min.

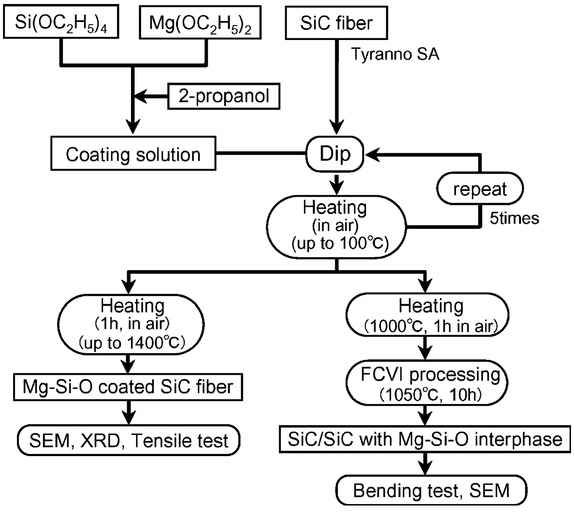

Fig. 1. Flow chart of fabrication and evaluation process for Mg–Si–O coating on the SiC fiber and SiC/SiC with Mg–Si–O interphase.

The SiC/SiC with the Mg–Si–O interphase was fabricated by a forced flow chemical vapor infiltration method (FCVI). A two dimensional-plain weave of Tyranno SA fiber fabrics was used. After the 5 times dip-and-hydrolysis process, the coated fabric was heated at 1000 °C in air for 1 h. Seven layers of the coated fiber fabrics with a fabric layer orientation of [0°/90°] were restrained in a furnace. The preform dimension was 55 mm in diameter and 2.5 mm in thickness. The precursor for the SiC matrix was methyltrichlorosilane (MTS, CH_3SiCl_3, Shin-Etsu Chemicals Co., Ltd., Tokyo, Japan). The SiC matrix was formed at 1050 °C with the MTS flow rate of ca. 0.5 g/min carried by 1 dm^3/min of H_2 under 0.05 MPa for 10 h. The perform was flipped 4 times during the FCVI process for matrix uniformity. After cutting and polishing the specimens, 3-point bending test was carried out at room temperature with a span length of 20 mm and the cross-head speed of 0.1 mm/min using an Instron-type bending machine (model AG-50kNG, Shimadzu Corp., Kyoto, Japan). The size of the specimen for the bending test was about $25 \times 4 \times 2$ mm^3. The surface of the coating fibers and SiC/SiC composite were examined using a scanning electron microscopy (SEM, model JXA-840A, JEOL Ltd., Tokyo, Japan).

3. Results and discussion

Fig. 2 shows the SEM photographs of uncoated and Mg–Si–O coated SiC fiber fabrics after heating at 1000 °C. The uncoated fibers have a smooth surface. From the photos, it was found that the surface of the fibers was fully coated and with a relatively rough surface. Fig. 3 shows the X-ray diffraction patterns of the coated Tyranno SA SiC fibers as a function of coating fabrication temperature. For

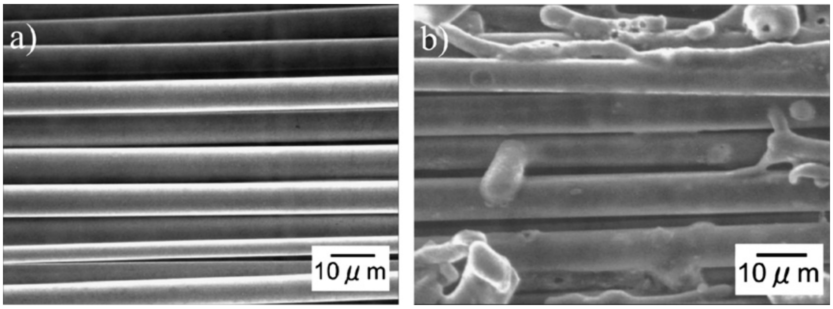

Fig. 2. SEM microphotographs of: (a) uncoated and (b) coated Tyranno SA fiber after heating at 1000 °C in air.

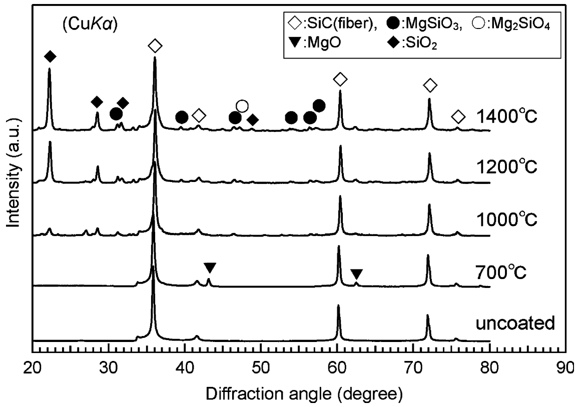

Fig. 3. X-ray diffraction patterns of Mg–Si–O coated Tyranno SA fibers as a function of coating fabrication temperature.

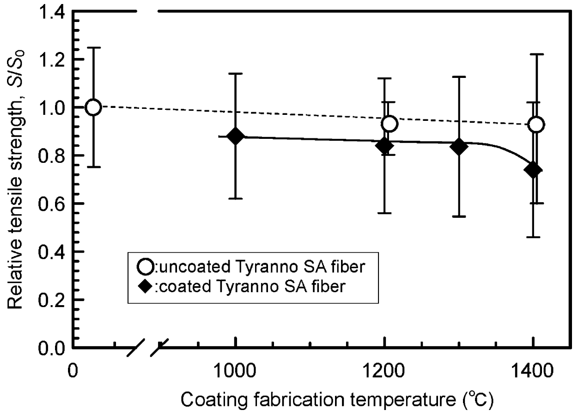

Fig. 4. Relative tensile strength of Tyranno SA fiber as a function of temperature.

comparison, uncoated-unheated fiber fabrics are also shown in the figure. The XRD patterns of the uncoated fibers showed five peaks around 2θ values of 35.7°, 41.4°, 60.4°, 71.8° and 76.0° which were attributed to β-SiC. While the widths of diffraction peaks attributed to conventional Hi-Nicalon SiC fiber were narrowed by the heating [16], no change of the peak shape and position were observed for Tyranno SA SiC fiber in the present work. This indicates that the advanced SiC fiber is much stabler than the conventional fibers under those conditions. In addition to these β-SiC peaks, the phase of MgO was observed in the coated fiber at 700 °C. While the peaks attributed to the MgO phase decreased with increasing temperature, peaks attributed to $MgSiO_3$, Mg_2SiO_4 and SiO_2 appeared and increased with the heating temperature.

In Fig. 4, the tensile strengths of the coated Tyranno SA SiC fibers are shown as a function of coating fabrication temperature. For comparison, those of uncoated fibers are also shown as a function of heating temperature in this figure. The tensile strength of as-received Tyranno SA, S_0, was 2.5 GPa. The tensile strength, S, of uncoated Tyranno SA fibers retained at 95% of S_0 after 1400 °C heating. The tensile strengths of coated Tyranno SA fibers retained 85% or higher of S_0 after heating below 1300 °C, while those of the coated fibers were degraded to 80% of S_0 after heating to 1400 °C. Since the strength of the coated conventional Hi-Nicalon SiC fiber was remarkably degraded to 55% of S_0 even at 1000 °C [16], this indicates that the advanced SiC fiber, Tyranno SA, has much higher oxidation resistance. Among the various fabrication processes of SiC/SiC, FCVI is an excellent technique for producing a crystalline β-SiC matrix with high purity, stoichiometry and a low thermal stress. Since the fabrication temperature of FCVI process is about 1100–1200 °C [17], the degradation of the tensile strength of Mg–Si–O coated Tyranno SA SiC is relatively small, and suggests that this is a viable method for producing a new oxidation resistant interphase.

Fig. 5 shows typical cross-sections of Tyranno SA/SiC composite with the interphase. Pores were observed among the fibers, and a grey circle was observed around the fibers, which are the interphase of Mg–Si–O. From this figure, one can see that those gray circles have almost uniform thickness of less than one-micron and the interphase fully coated each fiber. A typical load–displacement curve for a bending test for the coated Tyranno SA/SiC composite is shown in Fig. 6. The load–displacement curve of Hi-Nicalon/SiC composite is also shown in Fig. 6 for comparison [18]. For the Hi-Nicalon/SiC composite, a little ductile failure behavior was observed after the maximum load. The Tyranno SA/SiC composite also showed ductile failure behavior. The load of both composites did not reach zero after the large deformations. The maximum load of Tyranno SA/SiC composite was higher than that of Hi-Nicalon/SiC composite and it is clear that the Tyranno SA/SiC composite has high oxygen-resistance as described above. Fig. 7 shows typical fracture surfaces of the composites after bending test. Fiber pull-out was observed in both composites and this pull-out effect caused the pseudo-ductile failure mode, which indicates that the Mg–Si–O layer has potential for good properties as an interphase of SiC/SiC.

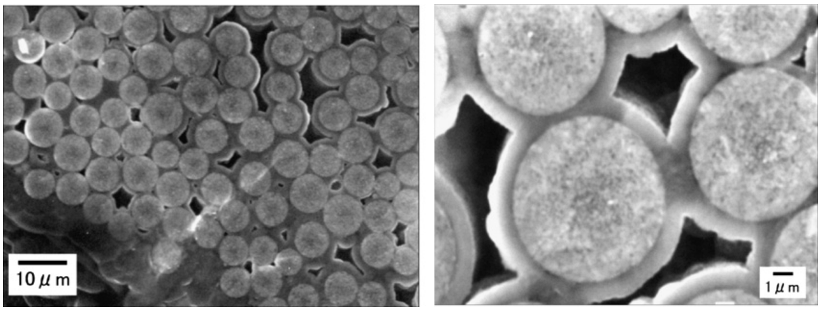

Fig. 5. SEM cross-sections of Tyranno SA/SiC composites with Mg–Si–O interphase.

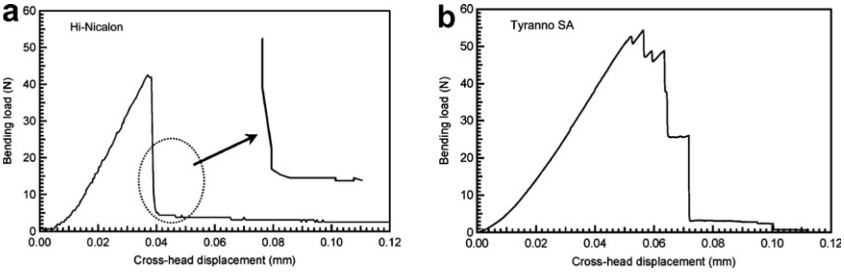

Fig. 6. Typical bending load–displacement curves of: (a) Hi-Nicalon/SiC and (b) Tyranno SA/SiC composites with Mg–Si–O interphase.

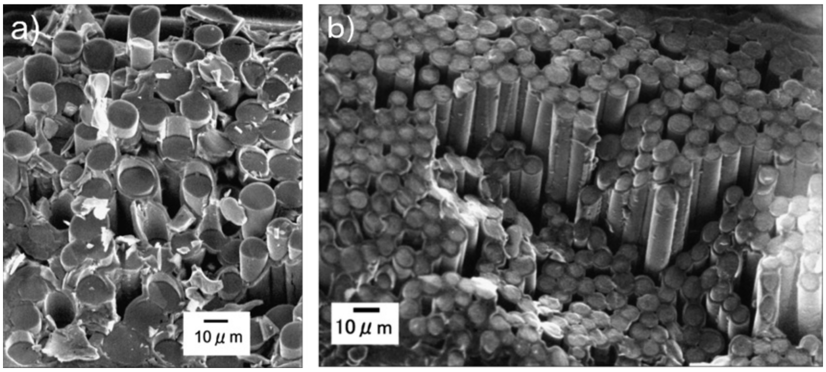

Fig. 7. Fracture surfaces of: (a) Hi-Nicalon/SiC and (b) Tyranno SA/SiC composites with Mg–Si–O interphase.

4. Conclusion

Mg–Si–O coating on an advanced SiC fiber, Tyranno SA, was prepared as an interphase of SiC/SiC composites by the alkoxide method. The surface on the fiber was coated completely by 5 times dipping in the coating solution of $[\mathrm{Mg}]_\mathrm{T} = 0.50\ \mathrm{mol/dm}^3$ and $[\mathrm{Si}]_\mathrm{T} = 0.25\ \mathrm{mol/dm}^3$. The coating consisted of a mixture of MgO, SiO_2, $MgSiO_3$, and Mg_2SiO_4 after heating at 1400 °C in air. Compared with uncoated fiber, the tensile strength of a coated fiber was retained at 85% or higher of S_0 after heating below 1300 °C, while those of the coated fibers were degraded to 80% of S_0 after heating at 1400 °C. A composite with the Mg–Si–O interphase was fabricated by FCVI process. This composite showed ductile failure behavior in a bending test and fiber pull-out effect was observed in the fracture surface of the composites. Those results indicate that the Mg–Si–O layer has potential for good properties as an interphase of SiC/SiC.

References

[1] P. Fenici, A.J.F. Rebelo, R.H. Jones, A. Kohyama, L.L. Snead, J. Nucl. Mater. 258–263 (1998) 215.
[2] A. Hasegawa, A. Kohyama, R.H. Jones, L.L. Snead, B. Riccardi, P. Fenici, J. Nucl. Mater. 283–287 (2000) 128.
[3] B. Riccardi, L. Giancarli, A. Hasegawa, Y. Katoh, A. Kohyama, R.H. Jones, L.L. Snead, J. Nucl. Mater. 329–333 (2004) 56.
[4] T. Taguchi, N. Igawa, S. Jitsukawa, T. Nozawa, Y. Katoh, A. Kohyama, L.L. Snead, J.C. McLaughlin, Ceram. Trans. 144 (2002) 69.
[5] T. Taguchi, N. Igawa, R. Yamada, M. Futakawa, S. Jitsukawa, Ceram. Eng. Sci. Proc. 22A (2000) 533.
[6] T. Taguchi, T. Nozawa, N. Igawa, Y. Katoh, S. Jitsukawa, A. Kohyama, T. Hinoki, L.L. Snead, J. Nucl. Mater. 329–333 (2004) 572.
[7] Y. Katoh, L.L. Snead, T. Nozawa, T. Hinoki, A. Kohyama, N. Igawa, T. Taguchi, Mater. Trans. 46 (2005) 527.
[8] S. Suyama, Y. Itoh, T. Kameda, A. Sayano, in: A. Kohyama, R.H. Jones, P. Fenici (Eds.), Proceedings of the 2nd IEA/JUPITER Joint International Workshop on SiC/SiC Ceramic Composites for Fusion Applications, The Japanese Society of Materials for Advanced Energy Systems, 1997, p. 89.
[9] W.Y. Lee, E. Lara-Curzio, K.L. More, J. Am. Ceram. Soc. 81 (1998) 717.
[10] Y. Seki, I. Aoki, N. Yamano, T. Tabara, Fusion Technol. 30 (1996) 1624.
[11] Takeda, A. Urano, J. Sakamoto, Y. Imai, J. Nucl. Mater. 258–263 (1998) 1594.
[12] T. Ichikawa, Y. Kohtoku, K. Kumagawa, T. Yamanura, T. Nagawasa, Nature 391 (1998) 773.
[13] T. Nozawa, T. Hinoki, L.L. Snead, Y. Katoh, A. Kohyama, J. Nucl. Mater. 329–333 (2004) 544.
[14] 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.
[15] Y. Katoh, A. Kohyama, T. Hinoki, L.L. Snead, Fus. Sci. Technol. 44 (2003) 155.
[16] N. Igawa, T. Taguchi, R. Yamada, S. Jitsukawa, Ceram. Eng. Sci. Proc. 21B (2000) 237.
[17] N. Igawa, T. Taguchi, L.L. Snead, Y. Katoh, S. Jitsukawa, A. Kohyama, J. Nucl. Mater. 307–311 (2002) 1205.
[18] N. Igawa, T. Taguchi, R. Yamada, Y. Ishii, S. Jitsukawa, Ceram. Eng. Sci. Proc. 26 (2005) 27.