3.2 Low Activation Joining of SiC/SiC Composites for Fusion Applications: Tape Casting TiC+Si Powders

— C. H. Henager, Jr., R. J. Kurtz, N. L. Canfield, Y. Shin, W. G. Luscher, J. T. Mansurov, T. J. Roosendaal, and B. A. Borlaug (Pacific Northwest National Laboratory[1])

OBJECTIVE

This work discusses the latest developments in TiC + Si displacement reaction joining at PNNL based on new work to produce tape-cast powders for improved SiC-joints.

SUMMARY

The use of SiC composites in fusion environments likely requires joining two plates using reactive joining or brazing. One promising reactive joining method uses solid-state displacement reactions between Si and TiC to produce Ti_3SiC_2 + SiC. We continue to explore the processing envelope for these joints to produce optimal joints to undergo irradiation studies in the High Flux Isotope Reactor (HFIR). One noted feature of the older joints, produced with tape-calendared powders of TiC + Si, were the large, unavoidable void regions they contained. Although the joints were very strong, voids are undesirable. In addition, the tapes made for these joints were produced approximately 20 years ago and had aged. Therefore, we embarked on an effort to produce new tape-cast powders of TiC and Si that can replace our aged tape-calendared materials.

PROGRESS AND STATUS

Introduction

SiC is an excellent material for fusion reactor environments, including first wall, plasma-facing, and breeder materials. SiC is low-activation, temperature-resistant, and radiation damage-tolerant relative to most materials. In the form of woven or braided composites with high-strength SiC fibers, it has the requisite mechanical, thermal, and electrical properties to be a useful and versatile material system for fusion applications, especially since microstructural tailoring during processing allows for control over the physical properties of interest [1-6]. However, it is difficult to mechanically join large sections of SiC using conventional fasteners. As a result, the analog of welding is being pursued for these ceramic materials [2, 4-15]. This paper reports on the current status of the production of tape-cast powders to be used for strong SiC-joining materials with fewer voids than observed in previous joints made with tape-calendared powders. The status of the unirradiated strength testing and HFIR irradiation testing of the new, tape-cast joints is also discussed.

Experimental Procedures

Tapes are cast using a prepared powder, binder, solvent, and plasticizer mixture that consists of a slurry preparation and tape-casting operation using blended TiC+Si powders (all high purity and d<10 μm), ethanol and methyl ethyl ketone (MEK) as solvents, polyvinyl butyral resin (PVB) as binder, n-butyl benzyl phthalate (BBP) as a plasticizer, and EMPHOS PS-21A organophosphate ester as a dispersant. The powder loading was optimized at approximately

[1] Battelle Memorial Institute under Contract DE-AC06-76RLO 1830 operates PNNL for the U.S. Department of Energy.

55% by volume for the dry tape after casting and drying. The initial TiC+Si powders were blended with a 3:2 molar ratio. The final tape thickness was varied and joints were made from tapes that were 43 µm, 90 µm, and 195-µm thick (green tape thicknesses). The tapes were smooth and easy to handle.

A single tape thickness was selected, from optical microscopy and quantitative metallographic analysis of SiC-phase content in the joints, for the production of CVD SiC miniature torsion test specimens. The CVD SiC test specimens were joined at 1708 K under 40 MPa pressure for four hours. Compared to previous processing times and temperatures, we increased the temperature by 10 K and increased the hold time at temperature and pressure to 4 h from 2 h. This was done to improve joint composition homogeneity from some preliminary high-resolution TEM results. These will be reported in a future semiannual report. Up to 20 specimens have been fabricated for unirradiated strength testing at PNNL and HFIR irradiation testing.

Results

Table 1 shows the data for the three tapes that were produced, including tape/joint thickness and SiC-phase content. Characteristically, SiC preferentially nucleates at the Hexoloy/joint interface and complicates the quantitative metallography since the joint SiC-phase is attached to the Hexoloy SiC-phase. Difficulty in distinguishing between the SiC-phase in the joint and Hexoloy SiC make it difficult to determine the actual interface location.

Table 1: New Tape Cast Joint Dimensions and SiC-phase Content

Tape Designation	Tape Thickness (µm)	Joint Thickness (µm)	SiC-phase Area Fraction (%)
3TiC+2Si-5 (43)	43	22	31
3TiC+2Si-5 (90)	90	47	32
3TiC+2Si-5 (195)	195	92	33

The optical images and quantitative data of the new, tape-cast joints matched the images and data recorded from the production of a previous set of joints, made with an older, tape-calendared material. In fact, the new joints appear to be superior because the large void regions found in the old joints are not observed. These voids may have been the result of air entrapment in the tape-calendared joining material since mass loss data from the new tapes match data from the older tapes but the new tapes are much thinner. However, despite having a thinner tape, the new joints are not thinner, indicating that the total amount of binder and plasticizer (and perhaps trapped air) were reduced but the total amount of powders per unit area were not.

Joints produced from the 43-μm tapes were exceptionally uniform. For these reasons, it was selected as the best tape to produce SiC miniature test specimens with. Hexoloy SiC specimens were initially produced to test the joining process. Figure 1 displays an optical micrograph of a Hexoloy SiC joint in its entirety, made from tape with a starting thickness of 43 μm. After processing, the joint has an average thickness of 25.60 μm (40.47% reduction). Figure 2 displays SEM images of the Hexoloy SiC joint at 2000x (Figure 2a) and 4000x (Figure 2b), respectively.

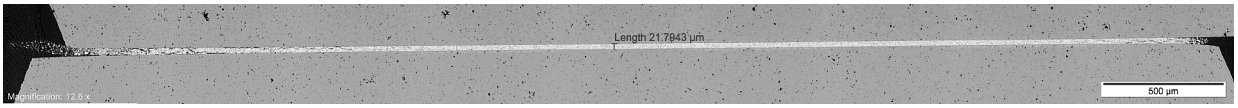
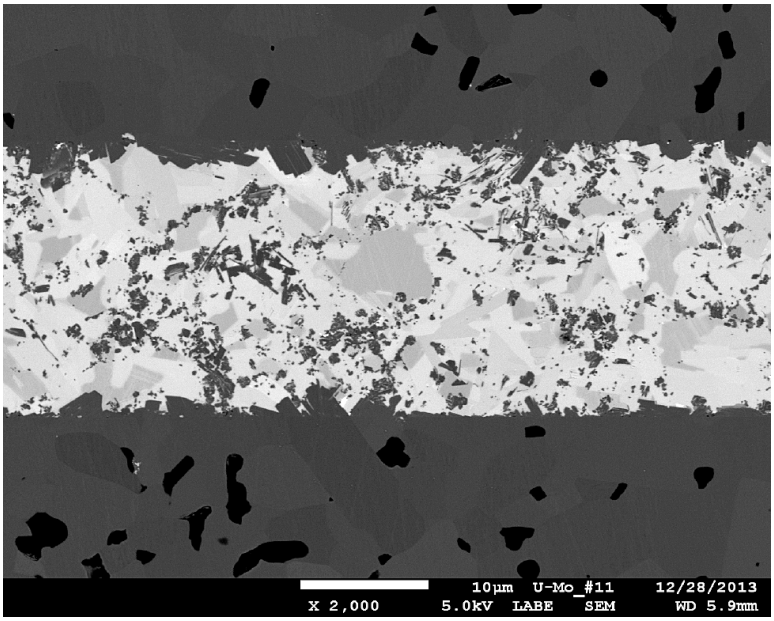
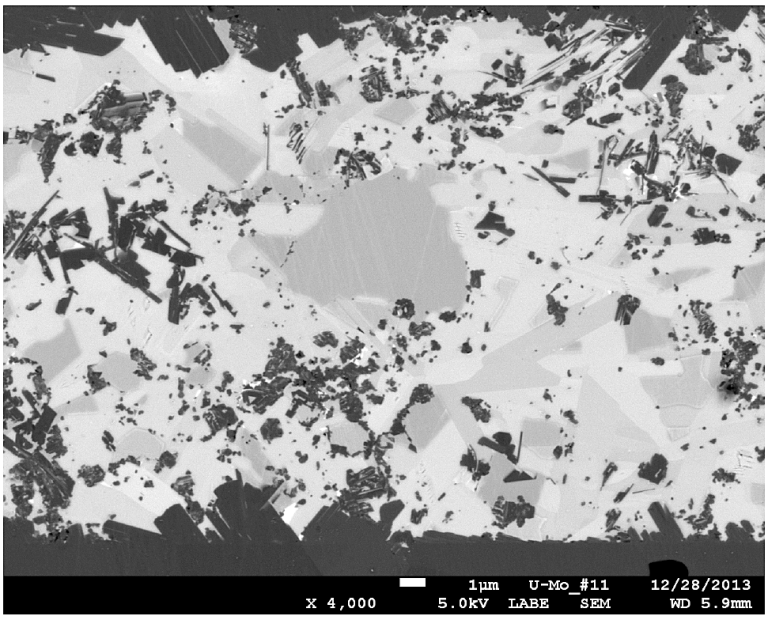


Figure 1. Optical micrograph with a 500-μm dimension marker showing the full width of the Hexoloy SiC miniature test specimen produced using 43 μm tape-cast tape. The bulk of the tape cast joint reveals a noted lack of the large pores that were a feature of previous joints. The overall joint thickness is also comparable to the previous joints.



(a)



(b)

Figure 2. SEM images of the joint in a Hexoloy SiC miniature test specimen produced using 43-µm tape-cast tape. The images were taken at (a) 2000x and (b) 4000x magnification. The dark phases are SiC and the light-colored phases are Ti_3SiC_2. Note the large blocky SiC at the top and bottom surfaces of the joined regions. These SiC features are nucleated and grown in from the surface of the Hexoloy polished surfaces.

Currently, eight (8) CVD SiC miniature test specimens have been fabricated for HFIR irradiation testing at ORNL from this new tape. Future work includes mechanical testing, optical microscopy, and SEM analysis of these test specimens. The resulting data and images will be compared to that collected from the older, tape-calendared joints.

ACKNOWLEDGEMENTS

This research was supported by Office of Fusion Energy Sciences, U.S. Department of Energy (DOE) under Contract DE-AC05-76RL01830.

REFERENCES

[1] Y. Katoh, L. L. Snead, C. H. Henager Jr, A. Hasegawa, A. Kohyama, B. Riccardi, and H. Hegeman, "Current status and critical issues for development of SiC composites for fusion applications," *J. Nucl. Mater.* **367-370 A**, 659-671 (2007).

[2] C. H. J. Henager, Y. Shin, Y. Blum, L. A. Giannuzzi, B. W. Kempshall, and S. M. Schwarz, "Coatings and joining for SiC and SiC-composites for nuclear energy systems," *J. Nucl. Mater.* **367-370**(1), 1139-43 (2007).

[3] P. Yvon, and F. Carre, "Structural materials challenges for advanced reactor systems," *J. Nucl. Mater.* **385**(2), 217-222 (2009).

[4] T. Nozawa, T. Hinoki, A. Hasegawa, A. Kohyama, Y. Katoh, L. L. Snead, C. H. Henager Jr, and J. B. J. Hegeman, "Recent advances and issues in development of silicon carbide composites for fusion applications," *J. Nucl. Mater.* **386-388(C)**, 622-627 (2009).

[5] H.-C. Jung, Y.-H. Park, J.-S. Park, T. Hinoki, and A. Kohyama, "RD of joining technology for SiC components with channel," *J. Nucl. Mater.* **386-388(C)**, 847-851 (2009).

[6] S. Nogami, A. Hasegawa, L. L. Snead, R. H. Jones, and K. Abe. Effect of He pre-implantation and neutron irradiation on mechanical properties of SiC/SiC composite. in Proceedings of the 11th Conference on Fusion Research, December 7, 2003 - December 12, 2003. 2004. Kyoto, Japan: Elsevier.

[7] M. Ferraris, P. Appendino, V. Casalegno, F. Smeacetto, and M. Salvo, "Joining of ceramic matrix composites for fusion application," Adv. Sci. Tech., 33(10th International Ceramics Congress, 2002, Part D), 141-152 (2003).

[8] B. Riccardi, C. A. Nannetti, T. Petrisor, and M. Sacchetti, "Low activation brazing materials and techniques for SiCf/ SiC composites," J. Nucl. Mater. **307-311**(Pt. B), 1237-1241 (2002).

[9] R. H. Jones, L. Giancarli, A. Hasegawa, Y. Katoh, A. Kohyama, B. Riccardi, L. L. Snead, and W. J. Weber, "Promise and challenges of SiC_f/SiC composites for fusion energy applications," *J. Nucl. Mater.* **307-311**, 1057-1072 (2002).

[10] R. L. Bruce, S. K. Guharay, F. Mako, W. Sherwood, and E. Lara-Curzio, "Polymer-derived SiCf/SiCm composite fabrication and microwave joining for fusion energy applications," IEEE/NPSS Symposium on Fusion Engineering, 1078-8891 (2002).

[11] M. Zucchetti, M. Ferraris, and M. Salvo, "Safety aspects of joints and coatings for plasma facing components with composite structures," *Fusion Eng. Des.* **58-59**, 939-943 (2001).

[12] A. R. Raffray, R. Jones, G. Aiello, M. Billone, L. Giancarli, H. Golfier, A. Hasegawa, Y. Katoh, A. Kohyama, S. Nishio, B. Riccardi, and M. S. Tillack, "Design and material issues for high performance SiC_f/SiC-based fusion power cores," *Fusion Eng. Des.* **55**(1), 55-95 (2001).

[13] C. A. Lewinsohn, R. H. Jones, T. Nozawa, M. Kotani, Y. Katoh, A. Kohyama, and M. Singh, "Silicon carbide based joining materials for fusion energy and other high-temperature, structural applications," Ceramic Engineering and Science Proceedings, 2001, 22(4, 25th Annual Conference on Composites, Advanced Ceramics, Materials, and Structures: B, 2001), 621-625.

[14] C. A. Lewinsohn, M. Singh, T. Shibayama, T. Hinoki, M. Ando, Y. Katoh, and A. Kohyama. Joining of silicon carbide composites for fusion energy applications. Colorado Springs, CO, USA: Elsevier (2000).

[15] P. Colombo, B. Riccardi, A. Donato, and G. Scarinci, "Joining of SiC/SiC_f ceramic matrix composites for fusion reactor blanket applications," *J. Nucl. Mater.* **278**(2), 127-135. (2000).