Fabrication of Multi-Layerd SiC Composite Tube for LWR Applications

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1. Introduction

SiC ceramics and SiC-based composites have recently been studied for LWR fuel cladding applications because of good mechanical/physical properties, neutron irradiation resistance and excellent compatibility with coolant under severe accident [1,2]. A multi-layered SiC composite tube as the nuclear fuel cladding is composed of the monolith SiC inner layer, SiC/SiC composite intermediate layer, and monolith SiC outer layer. Since all constituents should be highly pure, stoichiometric to achieve the good properties, it has been considered that the chemical process is a wellsuited technique for the fabrication of the SiC phases. In this study, the chemical vapor deposition (CVD) and chemical vapor infiltration (CVI) methods were employed for the fabrication of the composite tubes.

2. Fabrication of SiC and SiC Composite Tube

2.1 Selection of substrate material

In terms of purity, stoichiometry and geometric requirements such as straightness and ovality of a SiC inner layer, nuclear grade graphite is a suitable substrate material. Among a number of nuclear grade graphite, IG-110 graphite was selected because it is isotropic, contains little impurities, does not react with SiC, and has similar thermal expansion coefficient to SiC.

2.2 Deposition of monolith SiC inner layer

SiC was deposited onto the IG-110 graphite rod with 80 mm length and 8.5 mm in diameter by a low pressure chemical vapor deposition (LPCVD) method at 1300° C. Methyltrichlorosilane (MTS) was used as a source gas. Ar and H_2 were used as dilution and carrier gases. Fig. 1 shows monolith SiC inner layer deposited on the IG-110 graphite rod.

Fig. 1. Deposition of monolith SiC as an inner layer of the multi-layered SiC composite tube.

The SiC inner layer deposited had good uniformity in thickness. As shown in Fig. 2, thickness of SiC was about 300 µm after 5 hours of deposition. Standard deviation of a SiC inner layer within 50 mm was $8.3 \mu m$.

Fig. 2. Thickness variation of a SiC inner layer.

2.3 Fabrication of SiC/SiC composite interlayer

The CVI-processed SiC_f/SiC composite with a pyrolytic carbon (PyC) interphase was employed as an intermediate layer. Fully crystallized SiC fibers (Tyranno SA3) were wound by hoop or helical methods as shown in Fig. 3.

Fig. 3. Helical-wound and hoop-wound SiC fibers.

PyC as an interphase material of the composite was deposited on SiC fibers by pyrolysis of $CH₄$ at 1100 $^{\circ}$ C. As shown in Fig. 4, PyC interphase with 170 nm was uniformely deposited over the whole region of the composite.

Fig. 4. PyC as an interphase material deposited on SiC fibers.

Finally SiC as a matrix phase was infiltrated in the PyC-coated SiC fiber perform by the CVI method using MTS at 1000° C. Although the chemical reaction rather than mass-transport is the rate-limiting step at 1000° C, matrix infiltration preferentially occurred in an outer region of the composite layer for the hoop-winding specimens, as shown in Fig. 5(a). On the other hand, density gradient of SiC matrix was not observed for the helical-winding specimens owing to the lower fiber density. When fiber density of the composite is high, strength can be enhanced but mass transport of reactant gases into the composite perform is more difficult. As a result, majority of reactants are consumed at the surface and matrix density is not uniform in depth. However, the helical-wound composite inherently had large matrix pores between each layer as shown in Fig. 5(b).

Fig. 5. Infiltration behaviors of (a) hoop- and (b) helicalwinding SiC/SiC composite specimens.

2.4 Deposition of monolith SiC outer layer

The matrix infiltration was directly followed by the CVD deposition of a SiC outer layer at the same deposition conditions. The matrix infiltration and coating of the outer layer were carried out for 28 hours. After SiC matrix infiltration was complete within 5 hours, outer layer was deposited over 300 µm as shown in Fig. 6.

Fig. 6. Multi-layered SiC composite fabricated by CVD/CVI methods.

3. Summary

The multi-layered SiC composite was fabricated by chemical vapor deposition/infiltration methods for LWR fuel cladding. The monolith SiC inner layer and PyC interphase of the SiC/SiC composite had good uniformity in thickness. Matrix density of the SiC/SiC composite as an intermediate layer was also considerably high. Helical-winding SiC/SiC composite was more effective to minimize matrix density gradient.

Acknowledgement

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MEST) (No. 2012M2A8A5009818).

REFERENCES

[1] G. Newsome, L.L. Snead, T. Hinoki, Y. Katoh, and D. Peters, Evaluation of neutron irradiated silicon carbide and silicon carbide composite, Journal of Nuclear Materials, Vol. 371, p. 76, 2007.

[2] L. Hallsadius, S. Johnson, and E. Lahoda, Cladding for high performance fuel, Progress in Nuclear Energy, Vol. 57, p. 71, 2012.