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Effects of the curing methods on the fabrication of polycarbosilane derived SiC_f/SiC composite

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1. Introduction

Silicon carbide has potential advantages for structural applications in the next generation energy system-VHTR, GFR and the fusion reactor due to its unique properties such as a good irradiation resistance and thermo-mechanical properties, less severe waste generation due to neutron activation and improved plant conversion efficiencies by higher operating temperatures [1-3]. Among the several fabrication processes for SiC_f/SiC composites, the polymer impregnation and pyrolysis (PIP) process is the only method derived from polymeric precursors [4]. In the PIP process, the careful control of the oxygen content is important to avoid the property degradation at a high temperature because polymeric precursors are used as source materials of SiC ceramics. During the polymer precursor conversion process, unintended oxygen may be introduced for a cross-linking with producing the Si-O-Si bonds at the curing step [5]. High oxygen content affects the degradation of the high temperature stability in SiC ceramics. Therefore, a decrease of the oxygen content is desirable to obtain SiC ceramics with the high temperature stability [6]. One of the methods to reduce the oxygen content of polymer derived SiC ceramics is the irradiation curing process by gamma ray or electron beam [6, 7]. Polymer derived SiC ceramics with the low oxygen content prepared by the electron beam curing showed the improved thermal stability at a higher temperature [7].

In this study, the electron beam (EB) and the thermal oxidation curing methods were applied to make SiC_f/SiC composite using a polymer precursor, polycarbosilane (PCS) by the PIP process. And the evaluations of the curing effects, the pyrolysis behaviors and a high temperature stability were performed.

2. Experimental Procedures

PCS (NIPUSI Type A, Nippon Carbon Co., Japan) was used as a polymeric precursor. A plain weave fabric of Tyranno-SA^{TM} (Ube Co., Japan) was used as a reinforced substrate. Slurry was prepared by mixing PCS dissolved in hexane with beta SiC powder. Tyranno-SA fabric was immersed into the slurry to impregnate the slurry around the fiber and into the inter fiber space. Five layers of the slurry impregnated fabrics were stacked as a green preform. And then, they were dried in air and vacuum for 12 h, respectively. Dried green preforms were pressed at 7.5 kg/cm^2 with heating up to 300^{o}C. The applied pressure was released after cooling down to ambient temperature. The obtained preceramics were cured and pyrolized to converse to SiC ceramics. In this study, two different types of the curing process, thermal oxidation and electron beam curing were applied to compare the yield and oxygen content with different curing methods. The thermal oxidation curing was performed at 240^{o}C in air. The electron beam curing was carried out using 1 MeV electron beam generated by the electron accelerator (EB Tech, Co.) at ambient temperature under He atmosphere. The full doses were 10 and 15 MGy and a dose rate was 95 KGy/s. The pyrolysis of both preceramics was preformed at 1300^{o}C in Ar atmosphere.

Before making SiC/SiC composite, the conversions to SiC-based ceramics of only PCS with an oxidation or EB curing were also carried out to understand the oxygen behavior during the curing process, respectively. The conditions of curing and pyrolysis of PCS were same with those of SiC_f/SiC composite.

The yield was evaluated by measuring the weight changes. The oxygen content of conversed SiC-based ceramics was measured by the Oxygen-nitrogen determinater (TC-136, LECO, America). The weight losses of conversed SiC-based ceramics by both processes were analyzed to estimate the high temperature stability after a heat treatment at 1600^{o}C in a vacuum.

3. Results and Discussion

Fig. 1 shows the yield and the oxygen content of conversed SiC based ceramics from PCS using the thermal oxidation and EB curing method, respectively. Irrespective of the types of the curing methods, a high yield of 82% was obtained. This suggested that the larger amount of SiC could be contained in the EB cured specimens because oxygen did not cross-link with Si during EB curing. The oxygen content of the conversed specimens by the oxidation and EB curing were 21 % and less than 8 %, respectively. The oxygen contents of the conversed products could be remarkably decreased using the EB curing method. But the amount of the full does did not largely effect on the yield of the conversion during EB curing.

Fig. 2 shows the yield and the oxygen content of conversed SiC_f/SiC composites from PCS+SiC slurry

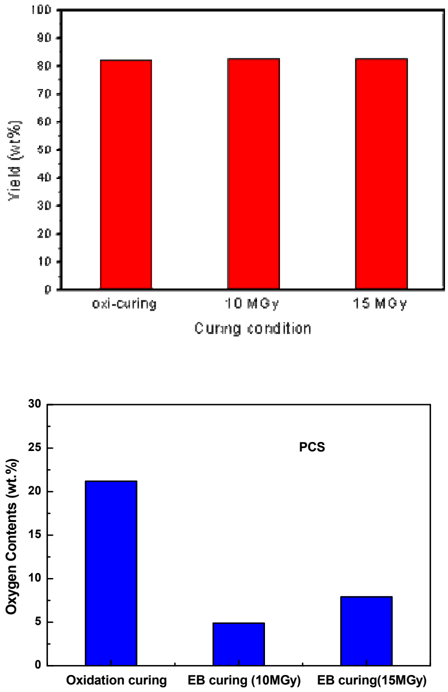

Fig. 1. Effect of the types of the curing methods on the yield and oxygen contents of conversed SiC based ceramics from only PCS.

using the thermal oxidation and EB curing method, respectively. As the same with the PCS conversion, the types of the curing methods did not largely effect on the yield of SiC_f/SiC composite prepared by the PIP process. A high yield of about 97% was obtained. But using the EB curing method, the oxygen contents of conversed composites could be remarkably decreased.

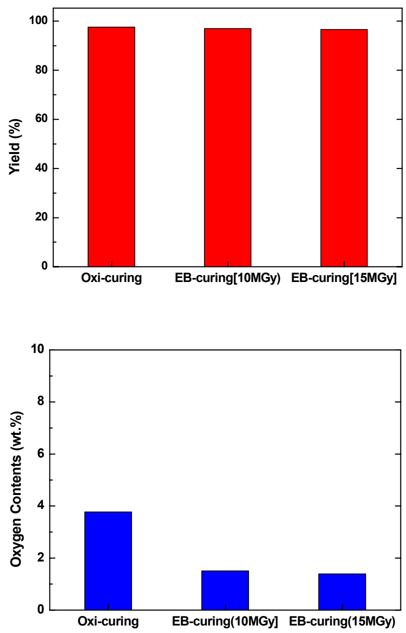

Fig. 2. Effect of the types of curing methods on yield and oxygen content of SiC_f/SiC composites by PIP.

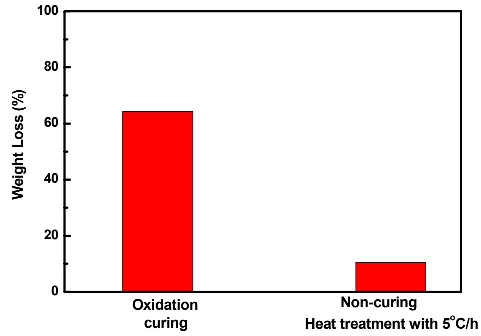

Fig. 3. Weight losses of Si-based ceramic prepared by conversion of PCS after heat treatment at 1600^{o}C.

Fig. 3 shows the thermal stability test results of conversed SiC based ceramic. The larger amount of the weight loss was observed in the oxidation cured specimen after heat treatment at 1600^{o}C (compared to non-cured one). This was caused to the volatilization of oxygen at a higher temperature (<1300^{o}C) according to the following reactions: SiO_2(s)+3C(s) → SiC(s)+2CO(g) or SixCyOz → xSiC+zCO+(y-x-z)C. A large amounts of weight losses in conversed Si-based ceramics seemed to be resulted in the property degradation at a high temperature.

4. Conclusion

The higher oxygen content was detected in the oxidation cured specimen by crosslink of Si-O-Si bond A large amount of oxygen in conversed Si-based ceramics seemed to be resulted in the property degradation at a high temperature. The types of the curing methods, oxidation curing or EB curing did not largely effect on the yield of the conversed products. But using the EB curing method, the oxygen contents of the conversed products could be remarkably decreased.

REFERENCES

[1] T. Burchell and J. Klett, in VHTR Materials R&D Collaboration Meeting, May 4~6, Idaho, USA (2004).
[2] A. Kohyama, T. Hinoki, T. Mizuno, T. Kunugi, M. Sato, Y. Katoh and J.S. Park, Pro. of ICAPP '05, Paper 5282, May 15-19, Seoul, Korea (2005).
[3] B.Riccardi, L.Giancarli, A.Hasegawa, Y.Katoh, A. Kohyama, R.H. Jones, L.L. Snead, J. Nucl. Mater., 329-333, 56-65 (2004).
[4] M.Kotani, Y.Katoh and A. Kohyama, Ceram. Trans., vol. 143, pp 97-103 Pub. By The Am.Ceram.Soc., Ohio, USA (2002)
[5] Y. Hasegawa, J. Mater. Sci., 24, 1177-1190 (1989).
[6] M. Takeda, Y. Imai, H. Ichikawa, T. Ishikawa, T. Seguchi and K. Okamura, Ceram. Eng. Sci. Proc., 12[7-8], 1007-1018 (1991).
[7] M. Takeda, Y. Imai, H. Ichikawa, T. Ishikawa, N. Kansi, T. Suguchi and K. Okamura, Ceram. Eng. Sci. Proc., 13[7-8], 209-217 (1992).