Oxidation Protective SiC Coating on Graphite for VHTR Core Support Structure

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1. Introduction

A protective SiC coating on the graphite components can assist in slowing the oxidation down. The prime concern in SiC coated graphite under the irradiation condition would be the coating robustness and the coating/substrate adhesion at the expected elevated temperature.

In this work, functionally gradient electron beam evaporative coating with an ion beam processing was conducted. These efforts are aimed at mitigation of the interface abruptness in order to reduce the residual stresses in the coating layer. Oxidation and thermal cycling tests of the coated specimens were performed and reflected in the process development to obtain more reliable SiC coated graphite.

2. Methods and Results

2.1 Sample preparation

Nuclear grade graphite specimens (IG110, Toyo Tanso) were used for the substrate materials. The SiC film was deposited on the graphite substrate with Ar+ ion beam mixing. Multiple samples held in jigs were both revolved around the jigs' main axis and rotated on their individual axis for a uniform coating on all sample surfaces.

Crucibles for e-beam evaporation were prepared with varied compositions of graphite and SiC (Fig.1). The coating thickness for each composition was determined in consideration of the stresses in the total film thickness of ~ 30 ㎛ [1]

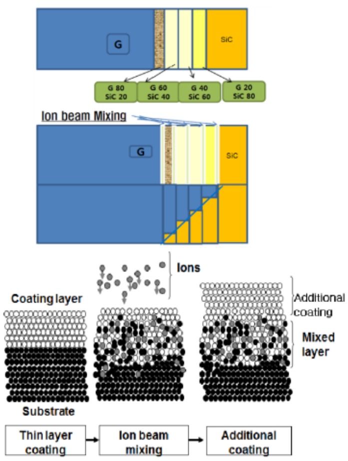

Fig. 1. Schematic of the ion beam mixed functionally graded coating.

2.2 FE-SEM of the coated

The film at the initial stage of coating appears a rough surface after coating (Fig. 2a) and shows a somehow dense and multiply stacked column (Fig. 2b-2c). The column structure is believed to be strain-tolerant against a dimensional change of the graphite caused by the neutron irradiation. However, if the inter-column width is larger, the graphite substrate may be more prone to oxidation.

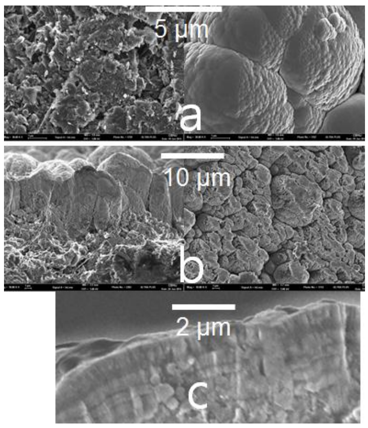

Fig. 2. As-received rough surface after SiC coating (a), the coating morphologies are the multiply stacked columnar growth (b - c).

2.3 Thermal cycling test

After 18 thermal cycles (1000 − 500 °C) in a vacuum, no coating delamination or crack formations at the interface owing to the difference in CTE between the film and substrate are observed, but the film is cracked (Fig. 3), ensuring a strong adhesion.

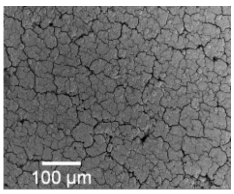

Fig. 3. After annealing at 1000 °, a film crack was formed rather than the delamination, suggesting a strong bonding.

2.4 Interface analysis

As shown in the Si and C Auger depth profiles (Fig.4), the ion beam mixed specimen shows a more broadened interface than the simply coated interface.

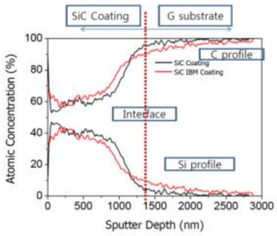

Fig. 4. The ion beam mixed specimen shows a more broadened interface than the simply coated interface.

The ion beam process may produce not only a graded interface but also a modification of the bonding property in the near surface layers of the graphite.

2.5 Oxidation test

When the samples are heated to 600 °C in air for 2 hrs, 45 wt% of the graphite is burnt off, whereas for the coated graphite only 5 wt % at 600 °C in air for 2 h (Fig. 5a). When this sample was heated-up to 1000 °C in air, a vigorous oxidation took place leaving only the SiC cask. A few paths on the SiC cask were found after heating at 1000 °C in air, which may have been caused by mars formed during the sample holding or cracks formed in the film due to the differences in the thermal expansion between the graphite and SiC film, and a film spallation during heating due to a difference in the thermal expansion coefficients between the graphite and SiC film (Fig. 5b). It is thus suggested that careful coating is required for slowing down the oxidation rate.

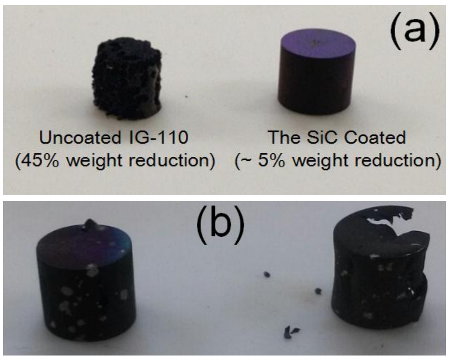

Fig. 5. Samples heated to 600 °C (a) and 1000 °C (b) in air for 2 hrs.

2.6 Oxidation after crack healing

In order to cover the crack lines, ion beam mixed coating followed by heating was repeated. The crack line widths of ~ 1 ㎛ upon heating at 1000℃ (Fig.6a) were greatly reduced as the process was repeated (Fig. 6b).

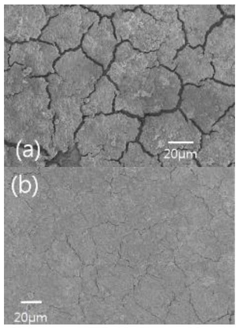

Fig. 6. The wide crack line upon heating at 1000℃ (a) were greatly reduced as the process repeated (b).

The oxidation resistance was gradually improved as the crack line widths were reduced by repeating the ion beam mixed coating process, as can be seen in Fig. 7. This suggests that efforts need to be paid to the samples having minimized crack line widths for retarding the oxidation.

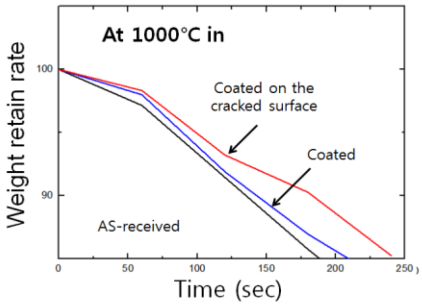

Fig.7. The oxidation resistance gradually improves as the crack line widths are reduced.

3. Conclusions

The film structure at the initial stage of coating shows somewhat dense and multiply stacked columnar. When samples are heated to 600 °C in air for 2 hrs, 45 wt% of the graphite is burnt off, whereas for the coated graphite only 5 wt % at 600 °C in air for 2 h. Vigorous oxidation of graphite took place through the mars and/or crack lines leaving only the SiC coating layer when heating at 1000 °C in air. The oxidation resistance was gradually improved as the crack line widths reduce with repeating the ion beam mixed coating process.

ACKNOWLEDGMENTS

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[1] J.I. Kim, W.-J. Kim, D.J. Choi, J.Y. Park, W.-S. Ryu, Design of a C/SiC functionally graded coating for the oxidation protection of C/C composites, Carbon Vol. 43, p.1749, 2005.