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Thermal Fatigue Resistance of CVD SiC/C Functionally Gradient Material

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Abstract

A deposit having a composition change from C to SiC was prepared on a graphite substrate by chemical vapour deposition (CVD). Under cyclic heat flow conditions (heat flux, 0·7 MWm^{-2}) at surface temperatures of 1700–1150 K, SiC/C functional graded material (FGM) did not tolerate cracking, however, SiC-coated graphite (SiC NFGM) suffered cracking under the same condition and vertical cracking was observed in the SiC layer. The cracking was considered to be due to the hoop tensile stress between the SiC layer and the graphite substrate caused by heating.

Eine Schicht, deren Zusammensetzung sich von C nach SiC verändert, wurde mittels chemischer Aufdampfung (CVD) auf ein Graphitsubstrat aufgebracht. Unter zyklischen Wärmeflußbedingungen (Wärmestrom 0·7 MWm^{-2}) bei einer Oberflächentemperatur von 1700 bis 1150 K, zeigte SIC/C anwendungsbezogenes Material (FGM) keine Rißbildung, während jedoch in SiC beschichtetem Graphit (SiC NFGM) sich bei den selben Bedingungen Risse bildeten; in der SiC-Schicht entstanden vertikale Risse. Die Rißbildung ist vermutlich auf die durch Erwärmung entstandene Zugspannung zwischen der SiC-Schicht und dem Graphitsubstrat zurückzuführen.

Sur un substrat de graphite on a déposé par dépôt chimique en phase vapeur (DCV), un produit dont la composition varie de C à SiC. Pour des conditions de flux thermique cyclique (flux thermique: 0·7 MWm^{-2}) et pour des températures en surface de 1700 à 1150 K, le matériau dont la composition varie de manière progressive ne présente pas de fissures après les tests mécaniques alors que, soumis aux mêmes conditions, le graphite recouvert de SiC présente des fissures verticales. La fissuration est attribuée aux contraintes de dilatation entre la couche de SiC et le substrat de graphite, causées par la température.

1 Introduction

In recent years, equipment for aerospace applications has required materials resistant to extreme conditions, e.g. high temperatures, and an oxidising atmosphere of >2000 K on one side and a temperature of around 1000 K on the other. However, traditional *in situ* composites with uniform properties are incapable of withstanding large temperature differences. In this respect, the authors have attempted to develop CVD-prepared SiC/C functional graded materials (FGMs) in which the SiC/C composition changes gradually,[1,2] to prepare SiC-C nano-composites[3] and to investigate the thermal properties of such composites.[4] The authors have also investigated the thermal shock resistance[5] and thermal barrier characteristics of SiC/C FGM.[6,7]

In the present work, the fabrication of SiC/C FGM by CVD and its thermal barrier characteristics were investigated.

2 Experimental

An $SiCl_4$–CH_4–H_2 system was used and the substrate was heated in a hot-wall type reaction chamber. The $SiCl_4$ reservoir was kept at 293 K and its vapour was carried into the furnace by bubbling hydrogen carrier gas. The gas flow rate ratio of $SiCl_4$ to CH_4 was controlled by a programme controller. The deposition temperature (T_{dep}) was selected between 1673 and 1773 K. The total gas pressure (P_{tot}) was 1·3 kPa. Deposition time (t_{dep}) was kept at 9 ks.

The experimental set-up for the evaluation of

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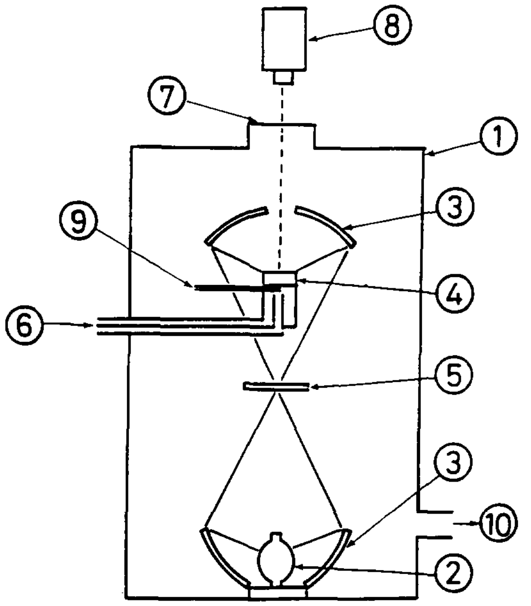

Fig. 1. Schematic diagram of high-temperature difference set-up. (1) vacuum chamber, (2) xenon arc lamp, (3) mirror, (4) specimen, (5) shutter, (6) liquid nitrogen, (7) window, (8) radiation thermometer, (9) thermocouples, (10) pump.

thermal barrier characteristics is shown in Fig. 1. A test specimen (30 mm in diameter, 10 mm in thickness) was heated by a xenon arc lamp having a maximum heat input of 30 kW. The bottom surface of the specimen was cooled with liquid nitrogen passing through a copper holder. The pressure inside the set-up was below 10^{-2} torr. The thermal barrier characteristics under forced temperature differences were investigated. The heat flux was 0·7 MWm^{-2}. The surface temperature (T_s) of the specimen was measured by a radiation thermometer (error level, 5%), and the temperature of the reverse side (T_b) was measured by thermocouples (error level, 1%). The specimen was 30 mm in diameter and supported with a copper holder. Three thermocouples were installed in the holder to measure the temperature of the reverse side and the heat flux.

Effective thermal conductivity (K_{eff}), as reflected by thermal barrier characteristics, is measured at steady heating. K_{eff} may be represented by the following equation:

$$K_{eff} = qt/(T_{ws} - T_{wb})$$

where q is the heat flux within the specimen; t is the thickness of a specimen, T_{ws} is the average surface temperature: and T_{wb} is the temperature of the reverse side as measured by extrapolation of the holder temperature.

Thermal stress was calculated by an isoparametric finite element method. Values used for the calculation were kept constant over the whole temperature range. Values in the intermediate composition were extrapolated from observed values, assuming that the surfaces without heat input were insulated.

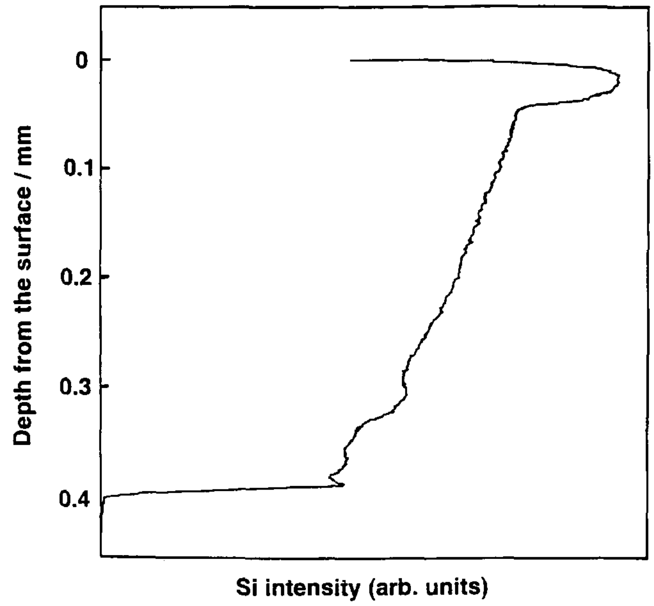

Fig. 2. Compositional distribution of SiC/C FGM prepared at a T_{dep} of 1673 K, and a P_{tot} of 1·3 kPa.

3 Results and discussion

The specimen of SiC/C FGM in the film thickness of 0·4 mm on the graphite substrate (30 mm in diameter, 10 mm in thickness) obtained by deposition at 1773 K and 1·3 kPa had many pores throughout the film. The specimen resulting from deposition at a T_{dep} of 1673 K and at a P_{tot} of 1·3 kPa had pores near the substrate (average porosity; 30%). SiC-coated carbon substrate in the film thickness of 0·4 mm was fabricated by the CVD method using an $SiCl_4$–CH_4–H_2 system at a T_{dep} of 1773 K and at a P_{tot} of 1·3 kPa.

The CVD conditions having a compositional distribution resulting in minimum thermal stress were 1773 K and 1·3 kPa, and 1673 K and 1·3 kPa. Figure 2 shows the compositional distribution of SiC/C FGM in the film thickness of 0·4 mm prepared at a T_{dep} of 1673 K and at a P_{tot} of

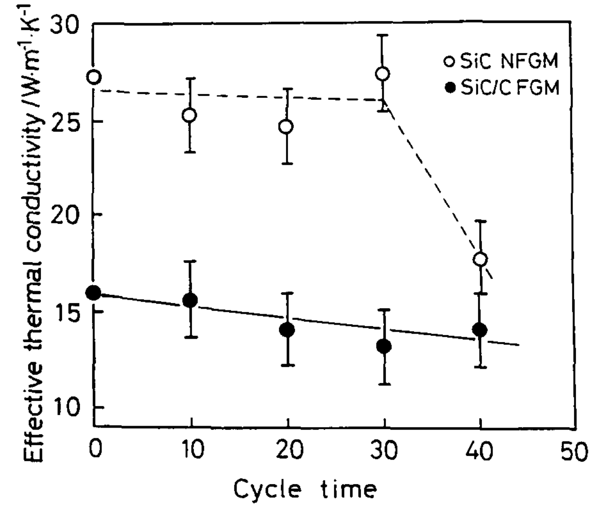

Fig. 3. Dependence of the effective thermal conductivity on the cyclic thermal exposures (heat flux, 0·7 MWm^{-2}).

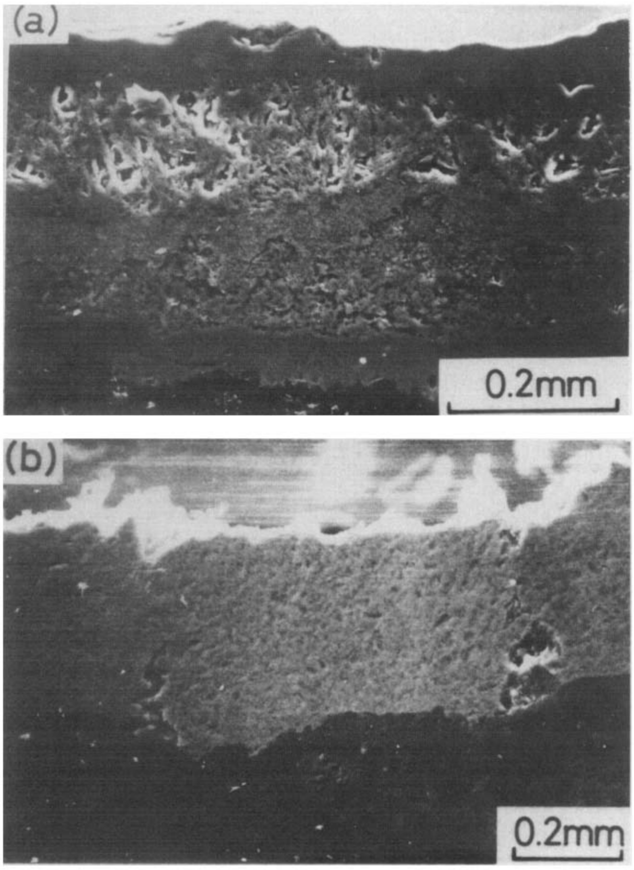

Fig. 4. Cross-sectional surfaces of SiC NFGM and SiC/C FGM after cyclic thermal exposure. (a) SiC/C FGM, (b) SiC NFGM.

1·3 kPa. A plate-like deposit having a compositional gradient from C to SiC was fabricated by controlling the Si/C ratio in the gas phase during deposition.

Figure 3 shows the effect of cyclic heating on the decrease of the effective thermal conductivity of SiC NFGM and SiC/C FGM. A decrease of effective thermal conductivity of SiC NFGM was observed from 30 to 40 cycles of heating. SEM photographs of the cross-sectional surface of SiC NFGM and SiC/C FGM after cyclic heating are shown in Fig. 4.[7] Vertical cracking was observed in the SiC NFGM. This cracking is thought to be due to the thermal stress under cyclic heating. On the other hand, SiC/C FGM did not suffer such cracking under the same conditions.

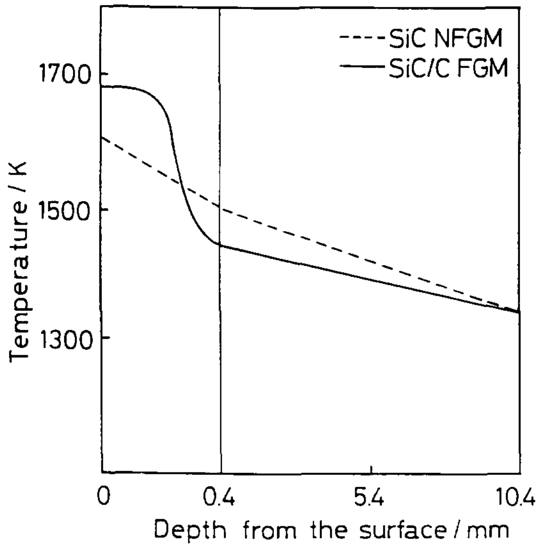

Fig. 5. Temperature distribution of SiC/C FGM and SiC NFGM under steady thermal exposure.

Figure 5 shows the calculated temperature distribution of SiC/C FGM and SiC NFGM under steady thermal exposure with a heat flux of 0·7 MWm^{-2}. The surface temperature and the reverse side temperature of the SiC layer (0·4 mm in thickness) were 1600 K and 1500 K, respectively (ΔT, 100 K), and the temperature difference of the SiC/C FGM layer (0·4 mm in thickness) were 1700 K and 1460 K, respectively (ΔT, 240 K). The difference of the surface temperatures between them might be caused by the thermal conductivity difference between SiC/C FGM layer and SiC layer. .

For the purpose of investigating thermal fatigue resistance, steady-state thermal stress analysis was attempted. Table 1 shows the values of SiC-C composites for the calculation of thermal stress.[8] The calculation was attempted using two-dimensional isoparametric steady-state method.[1] In the case of two-dimensional calculation, one can note the stress field by axial, radial and

Table 1. Values for the calculation of thermal stress

	SiC content (vol %)							
	100	*86·3*	*61·0*	*45·0*	*35·0*	*25·0*	*12·6*	*Substrate*
Thermal conductivity ($Wm^{-1} K^{-1}$)	56·7	46·0	29·3	20·4	15·4	10·7	5·42	10
Specific heat ($kJ\ kg^{-1} K^{-1}$)	0·78	0·78	0·78	0·78	0·78	0·78	0·78	0·7
Density ($10^3\ kg\ m^{-3}$)	3·21	3·02	2·66	2·44	2·30	2·16	1·99	1·82
Young's modulus (GPa)	257	107	57·5	48·3	38·9	29·2	20·1	30
Thermal expansion ($10^{-6} K^{-1}$)[a]	1·8	1·8	1·8	1·8	1·8	4·0	9·15	4·0
Thermal expansion ($10^{-6} K^{-1}$)[b]	2·16	2·16	2·16	2·16	2·16	2·16	0·8	4·0
Poisson's ratio	0·3	0·3	0·3	0·3	0·3	0·3	0·3	0·3

[a] Perpendicular to the substrate.
[b] Parallel to the substrate.

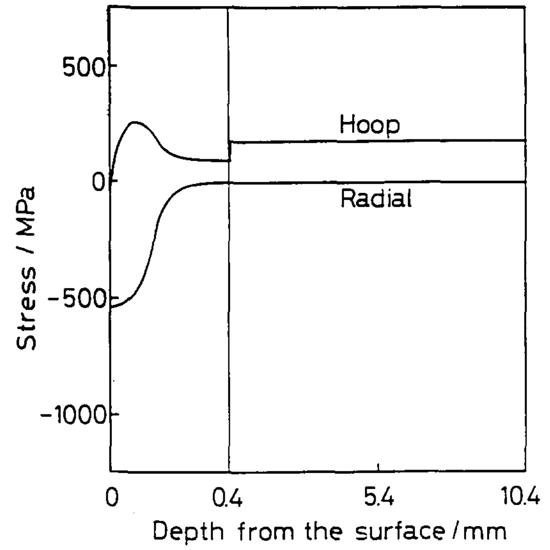

Fig. 6. Calculated hoop and radial stresses in SiC/C FGM.

hoop directions. In the present work, very low axial stress appears because of the low thermal expansion mismatching between the substrate and CVD SiC-C film. Figure 6 shows the calculated hoop (σ_{qq}) and radial (σ_{rr}) stresses in the SiC/C FGM. The σ_{rr} in the SiC/C FGM layer was compressive, and the σ_{qq} had a lower tensile stress than the fracture strength of SiC (650 MPa). Figure 7 shows the calculated hoop and radial stresses in the SiC NFGM. The σ_{qq} between the SiC layer and the substrate had a tensile stress nearly equal to the fracture strength of SiC (650 MPa). Cracking (Fig. 4b) was thought to be due to the hoop tensile stress between the SiC layer and the substrate which occurred at heating.

It is clear that SiC/C FGM has effective thermal barrier characteristics and thermal stress relaxation under conditions of temperature difference.

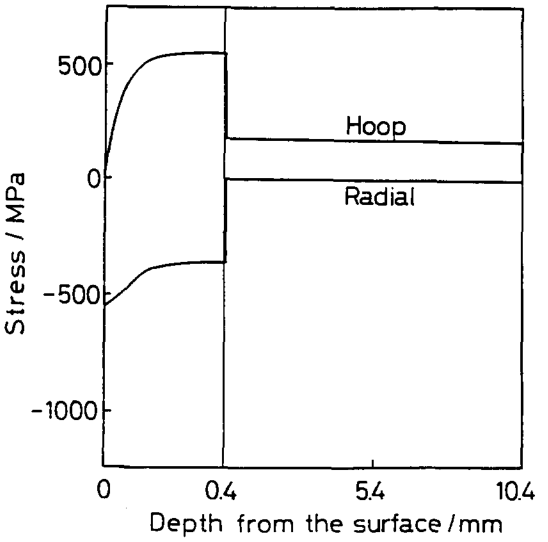

Fig. 7. Calculated hoop and radial stresses in SiC NFGM.

4 Conclusions

A plate-like deposit having a compositional gradient from C to SiC was obtained on a graphite substrate by chemical vapour deposition (CVD). The CVD conditions used were as follows: an $SiCl_4$–CH_4–H_2 system, deposition temperatures of 1673–1773 K, and a total gas pressure of 1·3 kPa. SiC/C functional graded material (FGM) has thermal barrier characteristics superior to those of SiC non-FGM (NFGM). Under cyclic high temperature heat flow conditions (heat flux, 0·7 MWm^{-2}) at surface temperatures of 1700–1150 K, SiC/C FGM did not suffer cracking and showed superior resistance to thermal fatigue. SiC NFGM suffered cracking under the same conditions and vertical cracking was observed in the SiC layer. Such cracking was thought to be due to the hoop tensile stress between the SiC layer and the substrate caused by heating.

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