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FABRICATION AND EVALUATION OF SiC/C FUNCTIONALLY GRADIENT MATERIAL

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ABSTRACT

A plate-like deposit having a compositional gradient from carbon (C) to silicon carbide (SiC) (SiC/C FGM) was obtained on a graphite substrate with chemical vapour deposition (CVD) by changing the Si/C molar ratio in input gas during deposition. Cracking due to heat flux was investigated using a CO₂ laser setup. Heat flux which resulted in cracking in SiC-coated graphite (SiC NFGM) was 5.8 MWm⁻² at a surface temperature (T_s) of 1300 K and a reverse surface temperature (T_b) of 1220 K. In the case of SiC/C FGM, heat flux occurred cracking was 7.4 MWm⁻² at a T_s of 1650 K and a T_b of 1080 K. The temperature difference between T_s and T_b was 570 K in FGM and 80 K in NFGM. Thermal stress calculation showed that in cracking caused by hoop stress, the stress was close to the fracture strength of SiC. It was clear that SiC/C FGM has effective thermal shock resistance and thermal barrier characteristics.

1. INTRODUCTION

In recent years, much attention has been focused on the development of high-temperature functional materials, especially in the field of aerospace applications /1/. Functionally gradient materials (FGMs) have been developed which have a compositional or structural distribution from one side to the other side. It is necessary to have both high oxidation resistance and high fracture strength at high temperatures. We have attempted to fabricate an FGM consisting of SiC and C, taking into consideration the high oxidation resistance and high fracture strength of SiC, as well as the low Young's modulus and easy-processing of C /2,3/. An SiC/C FGM which has a compositional gradient from C to SiC has been fabricated with the CVD method by changing the Si/C molar ratio in input gas. The fabrication and thermal properties of SiC-C composites were also investigated. The authors previously clarified the effect of the addition of C on the lower thermal conductivity of SiC-C composites /4/ and on the superior thermal barrier characteristics of SiC-C composites. Furthermore, they clarified the thermal barrier characteristics and thermal shock resistance of SiC/C FGM using a Xenon arc lamp heating method. As a result, it was shown experimentally and by calculation of thermal stress that SiC/C FGM has thermal barrier characteristics because of lower thermal conductivity, as well as thermal stress relaxation due to a lower Young's modulus /5-7/.

The authors have already investigated the thermal shock resistance of SiC/C FGM using the CO₂ laser (50 W) method [8,9]. However, the difference between SiC-single-layer-coated graphite (SiC NFGM) and SiC/C FGM was not clarified of the lower laser power (50 W) of this method. In the present study, we report the thermal shock resistance of SiC/C FGM having a graphite substrate (30 mm in diameter, 5 mm in thickness) using the 5 kW-CO₂-laser-heating method, and the effect of laser spot diameter.

2. EXPERIMENTAL

An SiCl₄-CH₄-H₂ system was used for the preparation of the SiC NFGM and SiC/C FGMs. The temperature of the SiCl₄ reservoir was kept at 293 K, and SiCl₄ gas was carried into the reactor by bubbling of hydrogen gas. The flow rate of hydrogen gas and SiCl₄ gas was $5.0 \times 10^{-6} \text{ m}^3 \text{ s}^{-1}$ and $1.7 \times 10^{-6} \text{ m}^3 \text{ s}^{-1}$, respectively. The flow rate of CH₄ gas was continuously changed from 6.7×10^{-6} to $1.3 \times 10^{-6} \text{ m}^3 \text{ s}^{-1}$. The gas flow rates were controlled by a programmable controller. The deposition temperature was 1673 K, and total gas pressure was 6.5 kPa. A compositional distribution in the depth direction of the SiC/C FGM was investigated by EPMA equipment. Specimens thus fabricated were irradiated by 50W- and 5 kW-CO₂ laser in air and evaluated as to thermal shock resistance.

3. RESULTS AND DISCUSSION

SiC/C FGMs were fabricated on graphite substrates (30 mm in diameter, 5 mm in thickness). Figure 1 shows a continuously changed composition of Si in a specimen having an SiC/C graded layer with a thickness of 1.8 mm. Other SiC/C FGMs having thicknesses of 0.8 mm and 2.1 mm and SiC NFGM having a 0.3 mm SiC layer were fabricated. Figure 2 illustrates the laser heating setup (CO₂ laser, Mitsubishi; ML50C) used for the thermal shock resistance test. The difference of thermal shock resistance between SiC

Fig. 1. Compositional distribution of SiC/C FGM prepared at a T_{dep} of 1673 K and a P_{tot} of 1.3 kPa.

Fig. 2. Schematic diagram of laser heating setup. (1)CO₂ laser, (2)specimen, (3)stainless steel, (4)firebrick, (5)radiation thermometer, (6)thermocouple(0.2 mm in diameter), (7)thermo-couple for laser trigger, (8)AE sensor.

NFGM and SiC/C FGM because of lower laser power (50W) was clarified. The square of AE peak height substituted for the area of AE signal as an AE energy. AE energy obtained by 50 W-laser heating is shown in Fig. 3, and SEM photographs of the heated surfaces of the specimens are shown in Fig. 4. No cracks were observed in either specimen. AE energy was very low, being equal to the noise level. The results show the high thermal shock resistance of SiC film.

Other thermal shock tests, therefore, were conducted on some specimens using a 5 kW-CO₂ laser. Figure 5 demonstrates the relationship between laser power density, laser spot diameter and cracking. With an increase in laser spot diameter above 25 mm, the heating situation was close to the overall heating of the specimen (30 mm in diameter). The critical laser power density shows other thermal shock resistance characteristics of the materials. This result demonstrates the high thermal shock resistance of SiC NFGM and SiC/C FGM compared with Al₂O₃, Macor and ZrO₂-NiCrAlY-coated stainless steel (SUS304) FGM. Compared with the critical laser power density of SiC NFGM and SiC/C FGM, SiC/C FGM has thermal shock resistance superior to that of SiC NFGM.

Fig. 3. Relationship between AE total energy and laser power density in the laser spot diameter of 1 mm. (a),sintered-ZrO₂(Y₂O₃); (b),plasma sprayed-ZrO₂/NiCrAlY/Cu FGM; (c),CVD-SiC(SiC NFGM) and (d), SiC/C FGM.

Fig. 5. Relationship between laser power density and spot diameter. open circle, no cracking; closed circle, cracking

Fig. 4. SEM photographs of the surface of (a);SiC NFGM and (b);SiC/C FGM after the irradiation of laser in the diameter of 1 mm.

Figure 6 shows the relationship between laser power density and the temperature difference of the heated and reverse sides. In the case of SiC NFGM, cracking caused by laser power density was 5.8 MWm^{-2} , and was 7.4 MWm^{-2} in SiC/C FGM. Increasing the thickness of the coated layer from 0.8 mm to 1.8 mm caused the temperature difference in SiC/C FGM to increase from 390 to 560 K. Heat flux caused cracking was 7.4 MWm^{-2} in both SiC/C FGMs. Results of local heating test without cracking for the specimen of 30 mm in diameter are shown in Table I. Figure 7 demonstrates the relationship between laser power density and the calculated surface temperature of the specimen. Cracking occurred on the SiC NFGM at a temperature of 1270 K. However, no cracking occurred on the SiC/C FGM with a film thickness of 1.8 mm at a surface temperature of 1500 K. Figure 8 shows the cracking in the specimens after laser irradiation. Cracks were observed around the specimens in both SiC NFGM and SiC/C FGM. SEM photographs of the sur-

Table I. Results of local heating test without cracking for the specimen of 30 mm in diameter.

Parameter	SiC NFGM	SiC/C FGM
Surface temperature (K)	1060	1510 1450
Reverse side temperature (K)	1000	1170 970
Temperature difference (K)	60	340 480
Film thickness (mm)	0.3	0.8 1.8
Heat flux (MWm^{-2})	4.6	6.4 6.4
Laser spot diameter (mm)	23.5	24.5 24.5

Fig. 7. Relationship between laser power density and surface temperature under thermal shock resistance test in the film thickness of $\blacktriangle$ 0.3, $\bullet$ 0.8, $\blacktriangledown$ 1.8 and $\blacksquare$ 2.0 in mm. open circle, no cracking; closed circle, cracking

Fig. 6. Relationship between laser power density and temperature difference under thermal shock resistance test in the film thickness of $\blacktriangle$ 0.3, $\bullet$ 0.8, $\blacktriangledown$ 1.8 and $\blacksquare$ 2.0 in mm. open circle, no cracking; closed circle, cracking

Fig. 8. Schematic diagram of the cracking in specimen under local heating.(1)specimens (SiC NFGM and SiC/C FGM), (2)crack

faces of laser-irradiated SiC NFGM and SiC/C FGM are shown in Fig. 9. Figure 9(a) shows a crack on the SiC NFGM with a film thickness of 0.3 mm and a heat flux of 5.8 MWm^{-2} . In this case, the reverse surface temperature was 1220K, and the calculated surface temperature was 1300 K. Figure 9(b) shows a crack on the SiC/C FGM with a film thickness of 1.8 mm and a heat flux of 7.4 MWm^{-2} . In both cases, cracks run through the SiC grains. These SEM photographs show that the cracks occurred due to similar fracture stress modes. The reverse surface temperature of SiC/C FGM was 1080 K, and the calculated surface temperature was 1650 K.

For the purpose of investigating crack formation, non-steady state thermal stress analysis was attempted. Heat flux was fixed at 3.6 MWm^{-2} , in which no cracking occurred in either SiC NFGM nor SiC/C FGM. Thermal stress on the surface of the specimens at 1 second after laser irradiation was calculated. As shown in Fig. 10, surface axial stress is close to zero in both cases of SiC NFGM and SiC/C FGM. The SiC layer or the SiC/C FGM layer did not detach from the graphite substrate supported by the lower axial stress. Axial stress in the depth direction was also close to zero. Such absence of detachment was thought to be due to the lower difference in thermal expansion coefficients between films and the substrate. Figure 11 demonstrates the radial stress on the surface of the specimens under laser heating. The radial stress on the surface of the specimens is compressive, and the radial stress is maximum on the surface. The maximum radial stress of SiC NFGM is 500 MPa close to the fracture stress of SiC, of which SiC/C FGM is 220 MPa lower than that of SiC NFGM. Calculation of radial stress showed the compressiveness extending over the whole specimen.

Figure 12 demonstrates the hoop stress on the surface of the specimens. This figure shows that the hoop stress in the part which was not heated by laser is tensile. The hoop tensile stress is maximum on the

Fig. 9. SEM photographs of the crack on the heated surface of specimen

- (a) SiC NFGM (heat flux, 5.8 MWm^{-2})
- (b) SiC/C FGM (heat flux, 7.4 MWm^{-2})

Fig. 10. Calculated axial stress on the surface of SiC NFGM and SiC/C FGM at a heat flux of 3.6 MWm^{-2} .

Fig. 11. Calculated radial stress on the surface of SiC NFGM and SiC/C FGM at a heat flux of 3.6 MWm^{-2} .

Fig. 12. Calculated hoop stress on the surface of SiC NFGM and SiC/C FGM at a heat flux of 3.6 MWm^{-2} .

surface of the specimens. SiC NFGM has a higher hoop tensile stress close to the fracture strength of SiC (650 MPa). Cracking around SiC NFGM at a heat flux of 5.8 MWm^{-2} was thought to be due to the outer hoop tensile stress caused by local heating. Cracking around SiC/C FGM at a heat flux of 7.4 MWm^{-2} was also caused by similar hoop tensile stress.

A thermal shock resistance test using laser heating was previously reported for a $\text{ZrO}_2(8\% \text{Y}_2\text{O}_3)/\text{NiCrAlY/SUS304 FGM}$ /10-12/. Calculation showed biaxial compressive stress in the heated part of $\text{ZrO}_2/\text{NiCrAlY/SUS304 FGM}$. It is expected that non-elastic deformation of ZrO_2 occurred in the heated part, and the tensile stress appeared in the some parts of ZrO_2 after cooling. Cracking of ZrO_2 film was thought to be due to tensile stress accompanying this non-elastic deformation /10/. In contrast to $\text{ZrO}_2/\text{NiCrAlY/SUS304 FGM}$, SiC/C FGM does not suffer cracking in the heated part, and cracking occurs due to hoop tensile stress in the outer portion of the specimens. These results and calculations show that the SiC/C FGM suffers cracking under local heating.

As mentioned above, it is clear that SiC/C FGM has both effective thermal shock resistance and thermal barrier characteristics under local heating.

4. CONCLUSIONS

A plate-like deposit having a compositional gradient from C to SiC was obtained on a graphite substrate (30 mm dia. x 5 mm thick) using an $\text{SiCl}_4\text{-CH}_4\text{-H}_2$ gas system by changing the Si/C molar ratio in input gas. Heat flux of 5.8 MWm^{-2} caused cracking in SiC NFGM (SiC-coated graphite) and 7.4 MWm^{-2} in SiC/C FGM (SiC/C FGM-coated graphite). Increasing the thickness of the SiC/C FGM layer from 0.8 mm to 1.8 mm caused the temperature difference between the surface and the reverse surface of the SiC/C FGM to increase from 390 to 560 K. Laser power density of 7.4 MWm^{-2} caused cracking in several types of SiC/C FGM. SiC/C FGM which contained C dispersion in the CVD layer had both effective thermal shock resistance and thermal barrier characteristics.

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