Fabrication and Life Prediction of SSiC Ceramic Joint Joined with Silicon Resin YR3370

YUAN Xiao-kun\textsuperscript{*}, XU Bing-she\textsuperscript{b}

\textsuperscript{a}School of Materials Science and Engineering, Beijing University of Aeronautics and Astronautics, Beijing 100083, China
\textsuperscript{b}College of Materials Science and Engineering, Taiyuan University of Technology, Taiyuan 030024, China

Received 16 May 2006; accepted 31 October 2006

Abstract

Joints between sintered silicon carbide (SSiC) were produced using a polysiloxane silicon resin YR3370 (GE Toshiba Silicones) as joining material. Samples were heat treated in a 99.99% nitrogen flux at temperatures ranging from 1100 °C to 1300 °C. Three point bending strength of the joint reached the maximum of 179 MPa as joined at 1200 °C. The joining layer is continuous, homogeneous and densified and has a thickness of 2 μm -5 μm. The joining mechanism is that the amorphous silicon oxycarbide (Si\textsubscript{x}O\textsubscript{y}C\textsubscript{z}) ceramic pyrolyzed from silicon resin YR3370 acts as an inorganic adhesive to SSiC substrate, which means the formation of the continuous Si-C bond structure between Si\textsubscript{x}O\textsubscript{y}C\textsubscript{z} structure and SSiC substrate. Life prediction of the ceramic joint can be realized through the measurement of the critical time of the joint after the cyclic loading test.

Keywords: silicon resin; YR3370; SiC ceramic; ceramic joining; joint life

1 Introduction

Silicon carbide ceramic and silicon carbide matrix composite are promising high temperature structural materials because of their high strength at elevated temperatures, high anticorrosion, high rigidity and low specific gravity, and are broadly used in advanced fields including aeronautical and astronautic vehicles, power plants and automobile brakes. Advanced ceramic joining is an issue with a view to fabricate ceramic components with complicated shapes and reduce the cost of manufacture, also to repair damaged ceramic parts and prolong the operational life\textsuperscript{[1]}.

Silicon resin is a promising material for ceramic and ceramic matrix composite joining because it is comparatively cheap and commercially available. The use of silicon resin for joining is applicable at low temperatures and can save great amount of energy, it can overcome limiting factors in other joining methods and decrease the deleterious chemical reactions. For example, joining reaction bonded silicon carbide (RBSiC) with polysiloxane silicon resin SR350 can gain a maximal three point bending strength of 220 MPa\textsuperscript{[2]}. While the evaluation of joining effect mainly comes through the joint strength, there is little concern about the joint life.

In this paper, joints between sintered silicon carbide (SSiC) were produced using a polysiloxane silicon resin YR3370 (GE Toshiba Silicones) as joining material, and the joint life was taken into account besides the joint strength to form a comprehensive evaluation of the joining effects. A ceramic joint life prediction method was brought for-
ward through the measurement of the critical time of the joint after the cyclic loading test, which could provide economic accounting and safety designing bases to ceramic joining.

2 Experiments

2.1 Fabrication of the joints

The SSiC ceramic specimen (FCT Fine Ceramics Technologies) is $\varnothing10 \text{ mm} \times 20 \text{ mm}$ in size with a density of 2.9 g/cm$^3$, an open porosity less than 2.0%, and a flexural strength of 280 MPa at room temperature. The specimen was polished on one side using a 1500# diamond millstone and ultrasonically cleaned with grain alcohol before joining.

The polysiloxane used for joining is silicon resin YR3370 (GE Toshiba Silicones). It is a transparent brittle solid with light yellow color, its molecular weight is about 7 kg/mol and has a density of 1.036 g/cm$^3$ at 25$^\circ$C, it has a glass transition temperature of around 86$^\circ$C and its pyrolysis in inert atmosphere yields amorphous Si$_x$O$_y$C$_z$ ceramic, with a weight loss of about 19% [3]. The silicon resin was dissolved in grain alcohol to yield a saturated solution, then 2.5 wt% silane crosslinking agent Silquest A-1100 (GE Toshiba Silicones) was ultrasonically dispersed into the saturated solution. The viscous saturated solution was homogeneously applied to the surface of the SSiC ceramic to be joined, and the samples were overlapped to obtain a joint with sandwich structure.

The Huaxiang HZS-25 vacuum furnace was refitted to facilitate the circulating of N$_2$ current. The joints were loaded with an axial pressure of 20 kPa and were placed in a 99.99% nitrogen flux. The joints were then preheated firstly at 180$^\circ$C for 1 h to let the silicon resin YR3370 crosslinked and solidified, then with a heating rate of 5$^\circ$C/min, the joints were preserved for 1 h at 1 100$^\circ$C to 1 300$^\circ$C to let the silicon resin YR3370 pyrolyzed, and the joints were naturally cooled at last.

IR spectra of silicon resin YR3370 pyrolyzed at different temperatures were observed by Fourier transform infrared spectroscopy FTIR Nicolet 750/950. Three point bending strength of the joint was measured by material testing machine Instron 5544, with a cross head speed of 0.5 mm/min and a span of 30 mm. Microstructure of the joint was observed by scanning electron microscopy JEOL JSM-6700F.

2.2 Measure of the critical time of the joint in cyclic loading

A batch of SSiC joints was fabricated under the same optimal condition (the joints were heat treated at the joining temperature of 1 200$^\circ$C for 1 h), and the joints may be considered with the same three point bending strength (about 179 MPa). The joints went through two kinds of cyclic loading test on material testing machine Instron 5544: (a) dynamic fatigue test: the joints were brought to axial compressive load with constant monotonically increasing rates ranging from $10^2$ N/s to $10^6$ N/s; and (b) cyclic fatigue test: the joints were brought to axial compressive load with a sinusoidal pattern at frequency of 10 Hz. The critical time of the joint was measured by Merlin analytic system of Instron 5544.

3 Results and Discussion

3.1 Joining effect

An example of the microstructure of the joint is shown in Fig.1. The joining layer appears to be fairly continuous, dense, and homogeneous and shows no obvious opening or crack, and has a thickness range from 2 μm to 5 μm. At the same time, the joining layer is well adherent to SSiC substrates, and there is distinct interface but no
obvious reaction layer between them. Three point bending strength of these joints range from 130 MPa to 179 MPa and achieve the maximum at 1 200 ℃. At the same time, some mixed fractures are formed when the joints undergo the three point bending strength test and indicate good joining effect.

3.2 Joining mechanism

During the crosslinking stage at low temperature, the silicon resin YR3370 releases most of gas including hydrogenium, water and ethanol. Sufficient crosslinking time at low temperature can make the evolution of gaseous species complete before the development of closed porosity in the structure. At the same time, because silane crosslinking agent Silquest A-1100 can crosslink the molecular chains and inhibit the shrinkage of silicon resin YR3370, the joining layer can has fairly continuous, dense, and homogeneous structure [4].

In the pyrolysis stage at high temperature, the silicon resin YR3370 forms amorphous Si$_x$O$_y$C$_z$ ceramic, which has covalent bonded network structure. The dominant gaseous species SiO and CO can play a major role in material transport and the formation of continuous bonding between amorphous Si$_x$O$_y$C$_z$ joining layer and SSiC substrate [5]:

\[
\begin{align*}
\text{Si} (s) + \text{SiO}_2 (s) & \rightarrow 2\text{SiO} (g) \\
\text{C} (s) + \text{SiO}_2 (s) & \rightarrow \text{SiO} (g) + \text{CO} (g) \\
2\text{Si} (s) + \text{CO} (g) & \rightarrow \text{SiC} (s) + \text{SiO} (g) \\
\text{SiO} (g) + 2\text{C} (s) & \rightarrow \text{SiC} (s) + \text{CO} (g)
\end{align*}
\]

The X-ray diffraction patterns of the pyrolyzate of silicon resin YR3370 pyrolyzed at temperatures ranging from 1 100 ℃ to 1 400 ℃ for 1 h show continuous broad diffraction peaks, which indicates that the pyrolyzate of silicon resin YR3370 is amorphous and has covalent bonded network structure [4]. Fig.2 shows the IR spectra of silicon resin YR3370 pyrolyzed at different temperatures.

Fig.2  IR spectra of YR3370 and it's pyrolyzate at different temperatures

As has been discussed before, the pyrolyzate of silicon resin YR3370 is amorphous Si$_x$O$_y$C$_z$ ceramic that has covalent bonded network structure, which should has theoretic molecular ratio of SiO$_2$.C$_{x}$. (x=0-2). Due to the disordered arrangement of molecular tetrahedron including (Si)O$_4$, C(Si)O$_3$, C$_2$(Si)O$_2$, C$_3$(Si)O and C$_4$(Si), and because the Si-O bond in Si-O-Si bridge can be easily skewed, the Si$_x$O$_y$C$_z$ ceramic has an amorphous structure. Fig.3 shows the structure simulation of the joint between SSiC substrate based on the inorganic adhesive bonding of amorphous Si$_x$O$_y$C$_z$ ceramic to SSiC substrates.

3.3 Life prediction of the joint

The evaluation of joining effect and quality between ceramic or ceramic matrix composite mainly comes through the joint strength. To the author’s opinion, the joint life should be taken into account to provide economic accounting and safety designing bases to ceramic joining.

The joint may undergo heat or stress cycle dur-
ing its application course, and the stresses may include tensile stress, compressive stress, shear stress, lap shear stress, torsional stress or peel stress. In this paper, the critical time when the joint is fractured during the axial cyclic loading is measured to determine the joint life in the cyclic compressive loading. The corresponding experimental method is shown in Section “2.2”, and the related loading patterns are shown in Fig.4.

![Fig.4 Patterns of cyclic loading](image)

Fig.4 Patterns of cyclic loading

Fig.5 shows the microstructure of the joint after the cyclic loading test, the amorphous Si$_x$O$_y$C$_z$ joining layer being fractured because of the crack growth and developing in its structure during the cyclic compressive loading.

![Fig.5 Joint after cyclic loading](image)

Fig.5 Joint after cyclic loading

Lee et al. [7] studied the fracture course of the ceramic lamina between two substrates because of the crack growth and developing during cyclic compressive loading. The axial compressive load $P$ produces a fracture stress $\sigma$ in ceramic lamina as follows

$$\sigma = \frac{P}{1.35d^2 \log \left( \frac{E_c}{E_s} \right)}$$  \hspace{1cm} (1)

where $d$ is the substrate thickness, $E_c$ and $E_s$ are Young’s modulus of ceramic lamina and that of substrates respectively.

The critical time $t_{Rd}$ in the dynamic fatigue test is

$$\sigma^n t_{Rd} = A(N + 1)$$ \hspace{1cm} (2)

The critical time $t_{Rc}$ in the cyclic fatigue test is

$$\sigma^n t_{Rc} = 2AN^{0.47}$$ \hspace{1cm} (3)

where $A$ and $N$ are characteristic parameters independent of load, time and thickness of lamina.

In this paper, the joint can be regarded as the amorphous Si$_x$O$_y$C$_z$ lamina located between two SSiC substrates. If the characteristic parameters of amorphous Si$_x$O$_y$C$_z$ lamina fitted from dynamic fatigue test and cyclic fatigue test respectively are the same, then the critical time of the joint in the cyclic loading can be calculated from Eqs.(2) and (3), and the ceramic joint life can be predicted.

In Eq.(1), the fracture stress $\sigma$ of the amorphous Si$_x$O$_y$C$_z$ ceramic shows a linear relationship to the axial compressive load $P$. Take compressive load $P$ as negative in calculation, with $d$=20 mm, $E_c$=97.9 GPa for amorphous Si$_x$O$_y$C$_z$ ceramic[8] and $E_s$=390 GPa for SSiC substrate. The critical times of the joint in the dynamic fatigue test and the cyclic fatigue test are shown in Fig.6.

![Fig.6 Critical time of the joint](image)

Fig.6 Critical time of the joint

The $\log t_{Rd}$ and $\log t_{Rc}$ both show a linear relationship to $\log \sigma$, according to Eqs.(2) and (3). Best fit result from dynamic fatigue test shows $N=11.89 \pm 0.27$, and best fit result from cyclic fatigue test shows $N=11.61 \pm 0.62$. The similar results can prove the feasibility of using the critical time of the joint after the cyclic loading test as the life prediction method of the ceramic joint. On the other hand, the difference between the two results may result from the diverse porosity in joining layer and the diverse combined area between amorphous Si$_x$O$_y$C$_z$ joining layer and SSiC substrate in different joints.
4 Conclusions

Joints between sintered silicon carbide were produced using a polysiloxane silicon resin YR3370 as joining material. The three point bending strength of the joint can reach the maximum of 179 MPa. The joining mechanism is that the amorphous silicon oxycarbide ceramic pyrolyzed from silicon resin YR3370 acts as an inorganic adhesive to silicon carbide substrate.

The characteristic parameters of amorphous silicon oxycarbide ceramic fitted respectively from dynamic fatigue test and cyclic fatigue test are nearly same, which proves the feasibility of using the measurement of the critical time of the joint after the cyclic loading test as the method of life prediction of the ceramic joint.

References


Biography

YUAN Xiao-kun He received his Ph.D. degree from Beijing University of Aeronautics and Astronautics in 2007. His current researches focus on the design, fabrication and application of ultra-high temperature structural ceramics and ceramic matrix composites.

E-mail: prince-of-egypt@163.com