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Joining SiC ceramics with silicon resin YR3184

Xiaokun Yuan *, Shu Chen, Xuehong Zhang, Tounan Jin

College of Materials Science and Engineering, Beijing University of Technology, Beijing 100124, China

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Abstract

Joints between reaction-bonded silicon carbide (RBSiC) and joints between pressureless sintered silicon carbide (SSiC) were produced respectively using a polysiloxane silicon resin (YR3184, GE Toshiba Silicones) as joining material. Samples were heat treated in a nitrogen flux at temperatures around 1200 °C. The maximum three point bending strength of the joints between reaction-bonded silicon carbide is 197 MPa. The maximum three point bending strength of the joints between sintered silicon carbide is 163 MPa. The join layers are continuous, homogeneous and densified and have thickness of 2–5 μm. The joining mechanism is that the amorphous silicon oxycarbide ceramic pyrolyzed from YR3184 acts as an inorganic adhesive.

Keywords: Polysiloxane; Silicon resin; YR3184; SiC ceramics; Ceramics joining

1. Introduction

Silicon carbide is an important engineering ceramic because of its high strength and stability at high temperatures. In order to use ceramics as structural materials, a suitable joining technique has to be developed. Ceramics joining is an issue with a view to fabricate ceramic components with complicated shapes and reduce the cost of manufacture. The use of polysiloxane as crosslinking adhesives at low temperatures and ceramic precursors at high temperatures offer an alternative to ceramics joining. Polysiloxane undergo a conversion to amorphous silicon oxycarbide ($Si_xO_yC_z$) ceramic with high productivity when heated at temperatures ranging from 800 °C to 1400 °C, provided a strict control of atmosphere inertia.

Polysiloxane is a promising material for ceramics and ceramic matrix composites (CMCs) joining because it is comparatively cheap and commercially available. Using polysiloxane as joining material, the ceramics joining is possible at relatively low temperatures with simple processing method, and the joint is microstructurally and compositionally stable at high temperatures. The polysiloxane that have been involved in or touched upon SiC ceramics bonding include SR350 [1] and SR355 [2] of GE, PMS(MK) [3], PPS(H62C) [4], H44 [5], MTES [6] and PTES [7] of Wacker Chemie, D_4H, D_4Vi [8] and SR249 of Dow Corning, PHMS [9] of Gelest, M130 [10] of Lukosil, and PR6155 of Fluorochemsilanes.

2. Materials and methods

The SiC ceramics used for joining include the reaction-bonded SiC (RBSiC) with a density of 3.0 g cm^{-3} and an open porosity less than 1.5%, and the sinterd SiC (SSiC) with a density of 2.9 g cm^{-3} and an open porosity less than 2.0%. The SiC ceramics specimens $\varphi 10 \times 20$ mm^3 in size were 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 YR3184 (GE Toshiba Silicones), its molecular weight is about 7 kg/mol and its pyrolysis in inert atmosphere yields amorphous $Si_xO_yC_z$ ceramic, with a weight loss of about 19% [11]. 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 SiC ceramics to be joined using a spatula, and the samples were overlapped to obtain a sandwich structure.

Subsequently, the specimens were loaded with an axial pressure of 20 kPa, and then it is preheated (at 180 °C) and

* Corresponding author. Tel.: +86 10 67396260.
E-mail address: yuanxiaokun@bjut.edu.cn (X. Yuan).

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pyrolyzed (range from 1100 °C to 1300 °C) for 1 h respectively in a nitrogen (99.99%) flux. The heating with a rate of 5 °C/min and the cooling by itself were performed in order to minimize possible residual stresses due to thermal expansion mismatch.

The interface between SiC substrate and join layer was observed by scanning electron microscopy (JEOL JSM-6700F). The three point bending strength of the joint was measured by materials testing machine (Instron 5544). Phase analysis of YR3184 with 2.5 wt.% Silquest A-1100 after pyrolyzed at different temperatures was done by X-ray diffractometer (Rigaku D/max 2200PC). Thermogravimetric analysis of YR3184 and YR3184 with 2.5 wt.% Silquest A-1100 were carried out respectively by thermogravimetric analysis (NETZSCH STA449C), in a nitrogen (99.99%) flux with a heating rate of 5 °C/min up to a maximum temperature of 1300 °C.

3. Results and discussion

3.1. Microstructure of the joints

An example of the joint between RBSiC is shown in Fig. 1(a), the microstructure of the interface between RBSiC substrate and amorphous $Si_xO_yC_z$ ceramic pyrolyzed from YR3184 at 1200 °C is shown in Fig. 1(b).

An example of the joint between SSiC is shown in Fig. 2(a), the microstructure of the interface between SSiC substrate and amorphous $Si_xO_yC_z$ ceramic pyrolyzed from YR3184 at 1200 °C is shown in Fig. 2(b).

The amorphous $Si_xO_yC_z$ ceramic interlayer pyrolyzed from YR3184 has a thickness range from 2 μm to 5 μm and shows an uniform and amorphous structure, which appears to be fairly continuous, dense, homogeneous and well adherent to the SiC substrates. The distinct interface between SiC substrate and join layer shows no obvious reaction layer.

EDX spectrum of the amorphous $Si_xO_yC_z$ ceramic interlayer in Fig. 1(b) reveals silicon 54.71 at.%, oxygen 26.47 at.% and carbon 18.82 at.% on average. EDX spectrum of the amorphous $Si_xO_yC_z$ ceramic interlayer in Fig. 2(b) reveals silicon 57.33 at.%, oxygen 23.04 at.% and carbon 19.63 at.% on average.

3.2. The influence of joining temperature to the joint strength

The three point bending strength of the joints between RBSiC substrate as a function of the pyrolysis temperature of YR3184 is shown in Fig. 3(a). The strength ranges from 160 MPa to 197 MPa and achieves the maximum at 1200 °C, which is 52% of the RBSiC substrate strength and thus result in

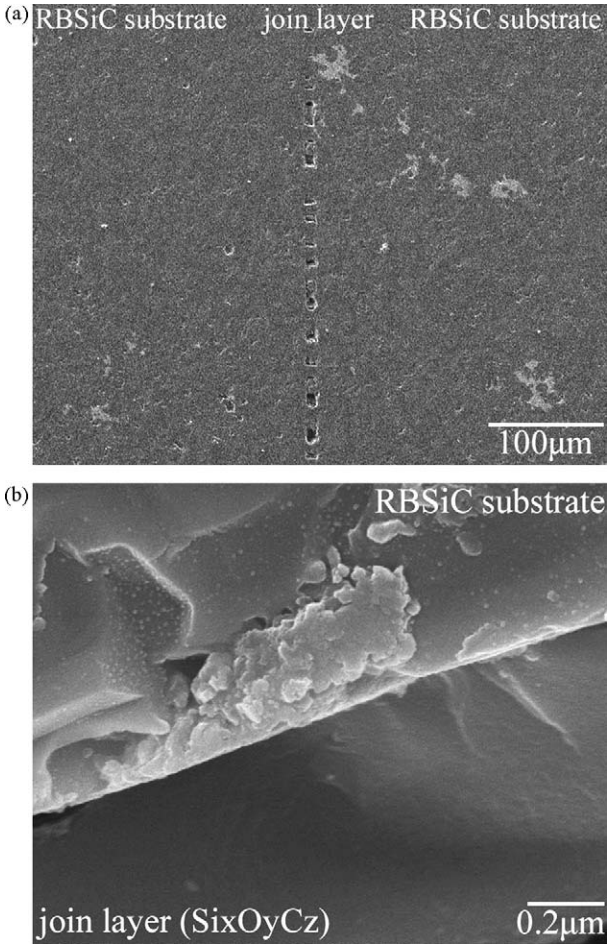

Fig. 1. (a) An example of the joint between RBSiC ceramic substrate. (b) Interface between RBSiC and $Si_xO_yC_z$ pyrolyzed from YR3184.

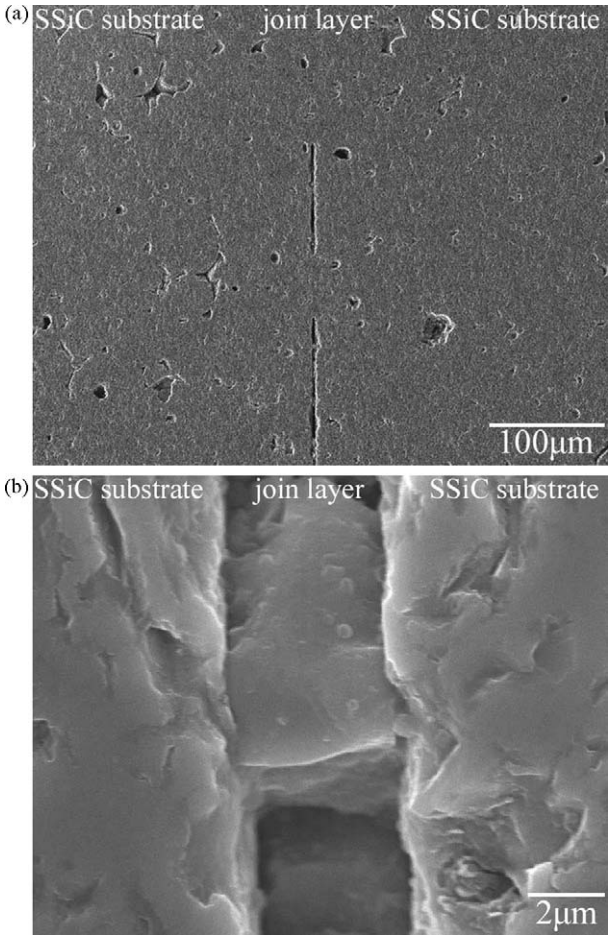

Fig. 2. (a) An example of the joint between SSiC ceramic substrate. (b) Interface between SSiC and $Si_xO_yC_z$ pyrolyzed from YR3184.

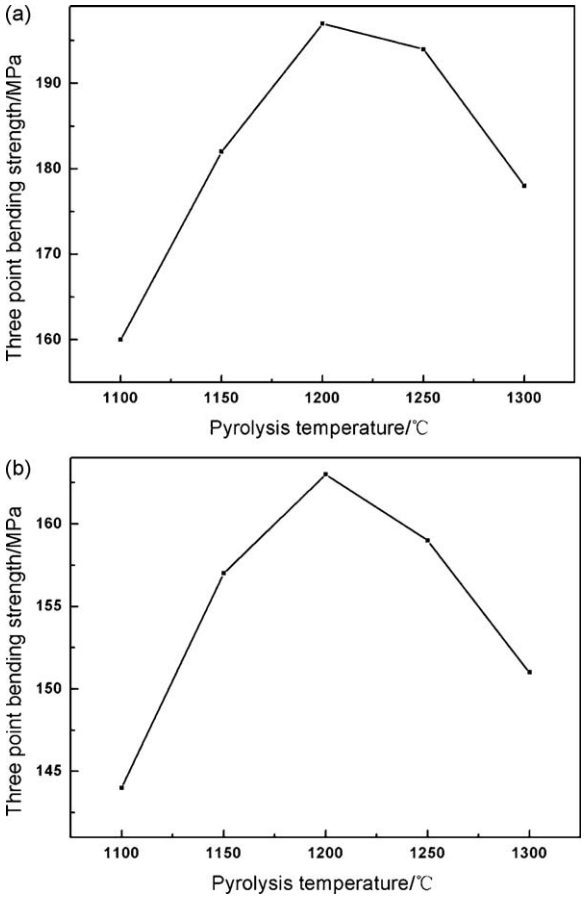

Fig. 3. (a) Three point bending strength of the RBSiC joints. (b) Three point bending strength of the SSiC joints.

the formation of mixed fracture when the joint undergo the three point bending strength test, as is shown in Fig. 4.

The three point bending strength of the joints between SSiC substrate as a function of the pyrolysis temperature of YR3184 is shown in Fig. 3(b). The strength ranges from 144 MPa to 163 MPa and achieves the maximum at 1200 °C, which is 58% of the SSiC substrate strength. The relatively lower bending strength of SSiC joints compared with RBSiC joints may be due to the higher porosity of SSiC, which can influence the bonding between SiC substrate and amorphous $Si_xO_yC_z$ ceramic.

Fig. 4. Optical micrograph of the SiC joint samples (joined at 1200 °C).

Fig. 5 shows X-ray diffraction patterns of silicon resin YR3184 with 2.5 wt.% Silquest A-1100 after pyrolyzed at different temperatures. Ranging from 1100 °C to 1200 °C, the XRD patterns show continuous broad diffract peaks, which indicates the pyrolyzate of YR3184 is amorphous and acts as an inorganic adhesive. When the temperature is above 1300 °C, the distinct diffraction peaks of SiC and SiO_2 can be seen in the XRD pattern, which indicate the formation of SiC and SiO_2 microcrystals at this temperature. The microcrystals are brittle and can result in the decrease of the SiC joint strength, which can be seen in Fig. 3(a) and (b) respectively.

3.3. Pyrolysis of YR3184

The TG-DTG curve of the pure silicon resin YR3184 and YR3184 with 2.5 wt.% Silquest A-1100 are shown in Fig. 6(a) and (b) respectively. With silane crosslinking agent filled in, the silicon resin reveals higher pyrolyzate output capacity due to the different pyrolysis mechanism.

3.3.1. Pyrolysis of YR3184

Fig. 6(a) shows the TG-DTG curve of silicon resin YR3184 and reveals three chemically distinct weight loss events.

Below 180 °C, some remnant solvents and micromolecules are volatilized.

In the range of 180–600 °C, weight loss occurs probably due to the release of some substances in gas phase including hydrogen, water and ethanol:

$$\equiv Si-OH + H-Si\equiv \rightarrow \equiv Si-O-Si\equiv + H_2(g) \quad (1a)$$

$$\equiv Si-OH + HO-Si\equiv \rightarrow \equiv Si-O-Si\equiv + H_2O(g) \quad (1b)$$

$$\equiv Si-OC_2H_5 + HO-Si\equiv \rightarrow \equiv Si-O-Si\equiv + C_2H_5OH(g) \quad (1c)$$

The decomposition of the silicon resin results in the higher weight loss at around 800 °C, probably due to the organic-inorganic conversion, with the formation of amorphous $Si_xO_yC_z$ ceramic that occurs between 600 °C and 1100 °C. Gas phase can play a major role for material transport as well as filler reaction during this course. The dominant gaseous species are SiO and CO which may form in $Si_xO_yC_z$ at about 1000 °C according to [3]:

$$Si(s) + SiO_2(s) \rightarrow 2SiO(g) \quad (2a)$$

$$C(s) + SiO_2(s) \rightarrow SiO(g) + CO(g) \quad (2b)$$

The two gaseous species can in turn promote the formation of SiC, which can be the key step in the formation of the tight bonding between SiC substrate and $Si_xO_yC_z$ [8,11]:

$$2Si(s) + CO(g) \rightarrow SiC(s) + SiO(g) \quad (3a)$$

$$SiO(g) + 2C(s) \rightarrow SiC(s) + CO(g) \quad (3b)$$

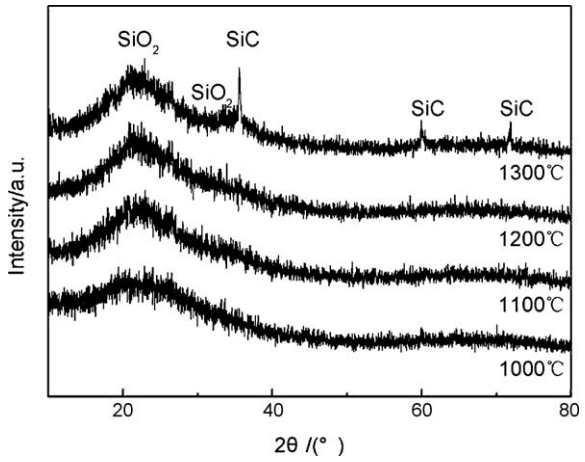

Fig. 5. XRD patterns of YR3184 after pyrolyzed at different temperatures.

The structure transformation of $Si_xO_yC_z$ and the formation of microcrystal occur above 1100 °C and show little weight loss.

3.3.2. Pyrolysis of YR3184 with silane crosslinking agent

Fig. 6(b) shows the TG-DTG curve of the silicon resin YR3184 with 2.5 wt.% silane crosslinking agent Silquest A-1100 added in. Comparing Fig. 6(b) with Fig. 6 (a), it can be seen that below 150 °C, weight loss of YR3184 shows a slower trend; hydrogen, water and ethanol release in the range of 150–400 °C; the decomposition of the silicon resin and the organic–inorganic conversion show two distinct DTG peaks between 400 °C and 1200 °C; the structure transformation of $Si_xO_yC_z$ and the formation of microcrystal occurs above 1200 °C. This is due to the crosslinking effect of Silquest A-1100 in silicon resin YR3184 which can coalesce the molecular chains, micromolecules and volatiles and thus form a more compact structure at lower temperatures, the pyrolysis of silicon resin in turn shows a different course high temperatures. With silane crosslinking agent, the pyrolyzate ratio of YR3184 increases from 48% to 71%, the weight loss procedure become slower at the same time, which can reduce silicon resin shrinkage and the formation of more continuous and compact join layer.

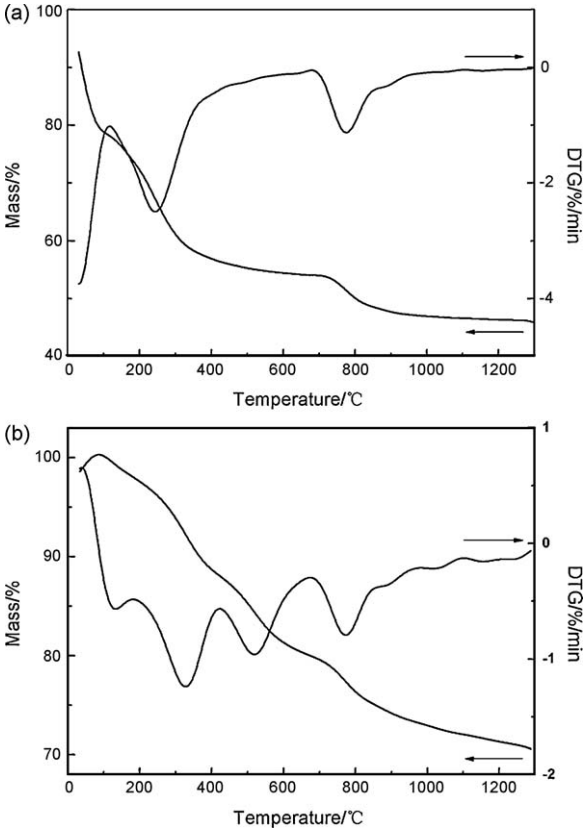

Fig. 6. (a) TG-DTG curve of YR3184. (b) TG-DTG curve of YR3184 with 2.5 wt.% Silquest A-1100.

4. Conclusion

By using a polysiloxane silicon resin (YR3184, GE Toshiba Silicones) as the joining material, it can produce ceramic joints between SiC parts, with the amorphous $Si_xO_yC_z$ ceramic pyrolyzed from YR3184 acting as an inorganic adhesive. The three point bend strength values of the SiC joints can achieve maximal at 1200 °C pyrolysis temperature of YR3184. With silane crosslinking agent Silquest A-1100 added in, the silicon resin YR3184 shows slower weight loss and higher pyrolyzed ceramic yield, which can promote the formation of continuous and dense join layer.

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