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Performance and structural evolution of high-temperature organic adhesive for joining Al_2O_3 ceramics

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ABSTRACT

High-temperature organic adhesive was prepared by using preceramic polymer V-PMS as matrix, B_4C powder and low melting point glass powder as additives, and was successfully applied to join Al_2O_3 ceramic. The obtained adhesive exhibited outstanding heat-resistant property and bonding strength. The bonding strength of the adhesive treated at 200 °C, 400 °C and 600 °C were 18.6 MPa, 20.4 MPa and 13.0 MPa, respectively. When treated above 800 °C, the bonding strengths were higher than 40 MPa. Moreover, the structural evolution of the obtained adhesive was studied by FTIR, DSC and XRD analysis. The results indicated that, up to 600 °C, the preceramic polymer and the glass additive play important role in improving the bonding strength. Above 800 °C, the outstanding wetting property and chemical compatibility of B_2O_3 formed by the oxidation of B_4C were mainly responsible for the excellent high-temperature bonding strengths.

1. Introduction

With the development of the aerospace, electronics, nuclear, transportation industry fields and other fields, ceramic materials are being more and more widely used due to their outstanding thermal and physical properties [1,2]. However, the nature of brittleness restricts their application in fabricating structures with large dimensions and complex shapes. Thus, advanced joining techniques have appeared to produce complex ceramic components with large sizes [3–6]. The current main methods for joining ceramics, such as mechanical connection, brazing and diffusion bonding, have significant limitations for further applications, especially in high-temperature areas. For traditional mechanical connections (riveting, bolting and threading), stress concentration is inevitable. While for the diffusion bonding and metal brazing, accurate and complex treatment processes are hard to guarantee. Moreover, the joints achieved by the above techniques still have certain limitations such as the porosity, the mismatch of thermal expansion coefficients, and the low strength at joining sites. Therefore, many studies are still focused on the development of new method for the joining of ceramics.

Recently, adhesive joining technique has drawn special attentions and high temperature adhesives using preceramic polymer as matrix have been proved to be promising and advantageous ceramics joining materials [7–17]. The most important advantages of adhesive joining technique are that joining is possible at relatively low temperatures, processing of the joints is simple and the joining material is generally microstructurally and compositionally stable up to high temperatures. Moreover, it is possible to tailor the composition and property of the joining interlayer by adding suitable inert or active filler to the preceramic polymer. Thus, many works have been done in this field. Wang et al. [7,8] joined graphite and Al_2O_3 ceramic by using phenol-formaldehyde resin as the matrix, and the strength of the joints reached 19.3 MPa after treatment at 1000 °C. Colombo et al. [9,11], Suo et al. [12], and Yuan et al. [13] successfully joined SiC materials by using silicone resin. Zhang et al. [17] prepared a room-temperature cured heat-resistant adhesive based on silicone resin and epoxy resin. However, as far as we are concerned, all of these adhesives cannot be used in the atmosphere or show unsatisfactory bonding strength at high temperatures in air atmosphere. Therefore, novel high-temperature adhesives with outstanding properties in air atmosphere are still urgently needed.

In our previous work [14,15], a novel preceramic polymer (V-PMS) was synthesized by modifying polymethylsilane with D4Vi (1,3,5,7-tetramethyl-1,3,5,7-tetravinylcyclotetrasiloxane, $[CH_3(CH_2{=}CH)SiO]_4$) and used as a high-temperature organic adhesive. However, the bonding strengths of the organic adhesive treated at high temperature were not satisfactory [14]. In this work, B_4C and glass powders were used as fillers to improve the properties of the high temperature adhesive. The performance and structural evolution of the adhesive for joining Al_2O_3 ceramics were investigated.

2. Experimental

2.1. Preparation of V-PMS and adhesives

V-PMS was synthesized as described in our previous report [15,18]. The adhesive HTA was prepared by using V-PMS as matrix

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and B_4C powder (Mudanjiang Qianjin Boron Carbide Co., Ltd., 3–5 μm, 95%), low melting point glass powder (Foushan Glass materials Co., Ltd, 1250 mesh, soften temperature: 400 °C to 800 °C, 99%) as additives. The mass ratio of V-PMS:B4C:(low melting point glass) was 100:40:30.

2.2. Characterization and bonding strength test

Fourier-transform infrared (FTIR) spectra were obtained with a Nicolet Avatar 360 instrument from solid samples embedded in KBr pellets. Thermogravimetric analysis (TGA) and Differential Scanning Calorimetry (DSC) were carried out on a NETZSCH STA449C thermogravimetric analyzer at a heating rate of 10 °C/min with air flow rates of 100 cc/min. Scanning electron microscopy (SEM) images of joint sections were obtained using a JEOL JSM-5600 LV electron microscope. The phase structure of the samples was characterized by X-ray diffraction analysis with CuK_α radiation and a nickel filter.

The bonding properties of the adhesive were tested by joining pieces of Al_2O_3 (Yixing Fengrui Ceramics Co., Ltd.) with a size of $20 \times 10 \times 5$ mm^3. Specimens were prepared and tested as described in the literature [15]. Before joining, the Al_2O_3 pieces with an average roughness of less than 3.2 μm were cleaned with alcohol in an ultrasonic bath for 30 min and then dried. The adhesive was uniformly brushed onto the surfaces of the SiC pieces and the SiC specimens with a thickness of the bonding layer about 30–50 μm were bonded together at room temperature in air atmosphere. After joining, all of the bonded samples were cured at 200 °C for 2 h in air. To obtain satisfactory bonding properties, 0.1 MPa pressure was applied on the joints until the specimens were cured. The cured samples were then placed in a muffle furnace and heat-treated at different temperatures ranging from 400 to 1200 °C. Each bonded sample was heat-treated at a given temperature for 2 h. The test method was performed as shown in Fig. 1. Five specimens were tested under the identical conditions. The average value and standard deviation values of the join strength were calculated for each case.

3. Results and discussion

3.1. Thermal properties of HTA

An organic adhesive for high-temperature applications should display high ceramic yield and excellent heat-resistance properties [10,19]. The ceramic yield of the high-temperature adhesive is crucial parameter to determine the strength of the joints. The amount of residual porosity, presence of cracks, joint thickness and shrinkage stresses are related to the ceramic yield of high-temperature adhesive [10]. Therefore, the thermal properties of HTA were first investigated by TGA and DSC (Fig. 2). HTA exhibited a maximum mass loss of about 9% as the temperature was increased to 710 °C. The lower mass loss always leads to less volatiles and smaller volume shrinkage, which is beneficial for high bonding strength. The specimens then showed a mass increment due to oxidation reaction between B_4C and oxygen or oxygenic volatiles derived from V-PMS. The lower the variation in mass, the higher the bonding strength can be expected. Therefore, the bonding strengths of HTA should be larger than that of V-PMS.

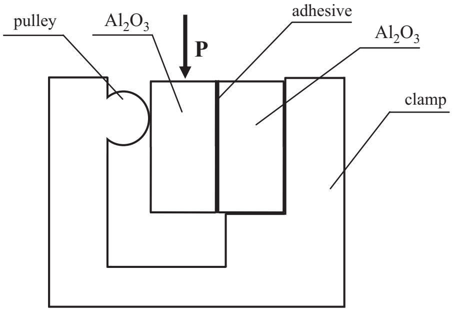

Fig. 1. Schematic show of the test of shear strength.

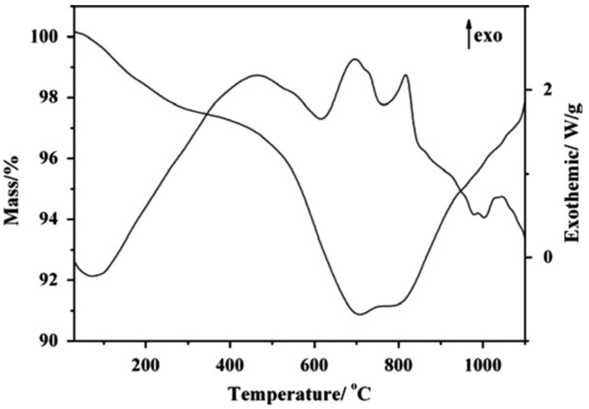

Fig. 2. TGA and DSC curves of HTA.

Table 1
Bonding strengths of Al_2O_3 joints treated at different temperatures.

	Bonding strength treated at different temperatures/MPa					
Adhesive	200 °C	400 °C	600 °C	800 °C	1000 °C	1200 °C
HTA	18.6 ± 1.1	20.4 ± 1.3	13.0 ± 1.4	43.1 ± 5.4	66.9 ± 6.8	50.1 ± 6.1

3.2. Bonding strengths of HTA

Table 1 listed the bonding strengths of the Al_2O_3 joints treated at different temperatures. It is shown that the adhesive exhibits excellent adhesive strengths. After cured at 200 °C for 2 h, the shear strength of the joints was 18.6 MPa. When the treated temperature was increased to 600 °C, the shear strength of the joints decreased to the minimum value (13.0 MPa). SEM micrograph (Fig. 3 (a)) shows that, in the joints treated at 600 °C, the broken sites of the joints are located in the interior of bonding cement, indicating a cohesive failure mode of the joints. The sample treated at 800, 1000, 1200 °C exhibits shear strength of 43.1, 66.9, 50.1 MPa, respectively. Thus, due to the satisfactory bonding force, the broken sites of the joints were observed both in the interior of cement and on Al_2O_3 substrate (Fig. 3 (b)), which indicates that the failures are mixture mode in the samples treated above 800 °C.

The SEM micrographs of the interfacial area of the joints bonded by HTA are presented in Fig. 4. It can be seen that the bonding layers of the HTA were uniform, dense and intact with no obvious cracks or pores when the treated temperatures were below 400 °C. As the temperature was increased, some cracks or pores appeared in the bonding layer due to the decomposition of the organic structures and release of volatiles. The interfaces between Al_2O_3 substrate and the bonding layer were indistinct and almost disappeared when the temperature was increased above 800 °C, indicating chemical reactions occurred at the interface. Consequently, excellent bonding strength of Al_2O_3 joints was obtained.