International Journal of Adhesion & Adhesives 35 (2012) 17–20

Contents lists available at SciVerse ScienceDirect

International Journal of Adhesion & Adhesives

journal homepage: www.elsevier.com/locate/ijadhadh

Preparation and performance of a heat-resistant organic adhesive obtained via a liquid SiC precursor

Xiao-zhou Wang*, Jun Wang, Hao Wang

Key Laboratory of Advanced Ceramic Fibers and Composites, College of Aerospace and Materials Engineering, National University of Defense Technology, Changsha 410073, PR China

ARTICLE INFO

Article history:
Accepted 30 December 2011
Available online 17 January 2012

Keywords:
High temperature adhesive
Ceramics
Polymethylsilane
Mechanical properties of adhesives

ABSTRACT

A heat-resistant organic adhesive rich in active Si–H bonds and CH═CH_2 bonds has been synthesized by modifying polymethylsilane with D4Vi. The structure and properties of the adhesive have been investigated by FTIR, GPC, TGA, XRD, bonding strength tests, and SEM. The results show that the obtained adhesive exhibits outstanding thermal stability and bonding properties. The ceramic yields of the adhesive treated in Ar or in air at up to 1200 °C were measured as 81% and 90.6%, respectively. The adhesive can maintain an amorphous state even when heat-treated at 1200 °C for 2 h in air. The room temperature shear strength of the adhesive was measured as 14.9 MPa, and this increased to a maximum value of 31.7 MPa after heat-treatment at 1000 °C for 2 h.

1. Introduction

Due to its outstanding mechanical properties and stability at high temperature, silicon carbide is an attractive material that has been widely applied in the aerospace, electronics, nuclear, and transportation industry fields [1]. However, the brittle nature and high rigidity of SiC makes it very difficult to fabricate structures of large dimensions and complex shapes. Thus, many methods have been used for joining SiC structures, of which the adhesive bonding technique is one of the most convenient and promising [2–4].

There are two kinds of high-temperature adhesives. Inorganic adhesives possess outstanding heat resistance, but they are brittle with low bonding strength and require special joints (muff-coupling, groove joint, etc.), which has definitely limited their application in the bonding of structural components. Organic adhesives have attracted particular attention due to their excellent bonding strengths. Although organic resins begin to decompose above 300 °C, the residues derived from them may be converted into carbonaceous char at high temperatures (> 1000 °C), which possess excellent thermophysical properties. Therefore, heat-resistant organic adhesives prepared from polysiloxane and phenol-formaldehyde resin were investigated and used to join ceramic materials, and were found to show satisfactory bonding strength [3–9]. However, as far as we are aware, such adhesives cannot be used in the atmosphere and show unsatisfactory bonding strengths at high temperatures in air [4–6,8,9]. Considering the potential applications of heat-resistant organic adhesives in joining SiC ceramic materials, it was deemed of value to prepare an organic adhesive capable of providing outstanding bonding strength at high temperatures in air.

As a liquid precursor of silicon carbide, polymethylsilane (PMS) has attracted substantial interest in the past years because of its stoichiometric composition (-($SiH(CH_3)$)$_n$-) and high chemical activity [10–13]. By virtue of its many Si–H bonds, PMS can be modified by organoborate additives [11], polyborazine [12], or metal chlorides [13–15] to improve its ceramic yield. In the present study, a polymer of high ceramic yield has been synthesized by modifying PMS with D4Vi (1,3,5,7-tetramethyl-1,3,5,7-tetravinylcyclotetrasiloxane, [CH_3(CH_2═CH)SiO]$_4$). The as-synthesized polymer has been used as an adhesive to join SiC ceramic and its properties have been investigated.

2. Experimental

PMS was synthesized by a condensation reaction according to our previous report [13]. PMS and D4Vi (98%, Shanghai Duolin Chemical Inc., purified by distillation) were first mixed, and then the mixture was heated at 120 °C for 3 h under an N_2 flow, whereupon a highly viscous polymer was obtained. The resulting polymer was used directly as an adhesive.

Fourier-transform infrared (FTIR) spectra were obtained with a Nicolet Avatar 360 instrument. Gel-permeation chromatography (GPC) analysis was performed on a Waters 1515 instrument using THF as eluent. Thermogravimetric analysis (TGA) was carried out on a NETZSCH STA449C thermogravimetric analyzer at a heating

* Corresponding author. Tel.: +86 731 84574789; fax: +86 731 84576578.
E-mail address: wangler2002@163.com (X.-z. Wang).

0143-7496/$ - see front matter © 2012 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ijadhadh.2012.01.002

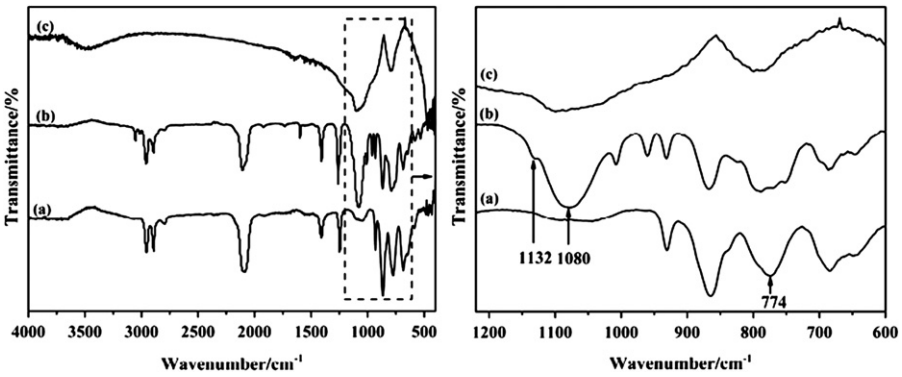
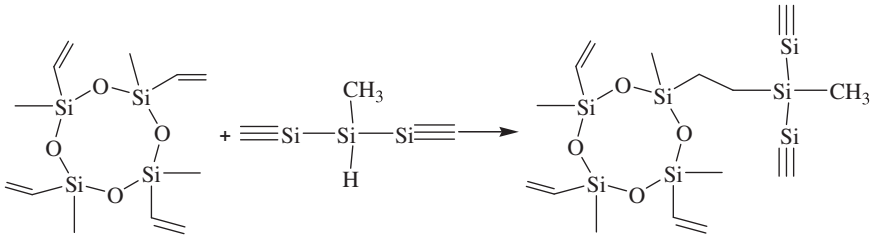


Fig. 1. FTIR spectra of (a) original PMS, (b) adhesive, (c) adhesive heat-treated in air at 1200 °C for 2 h.



Scheme 1

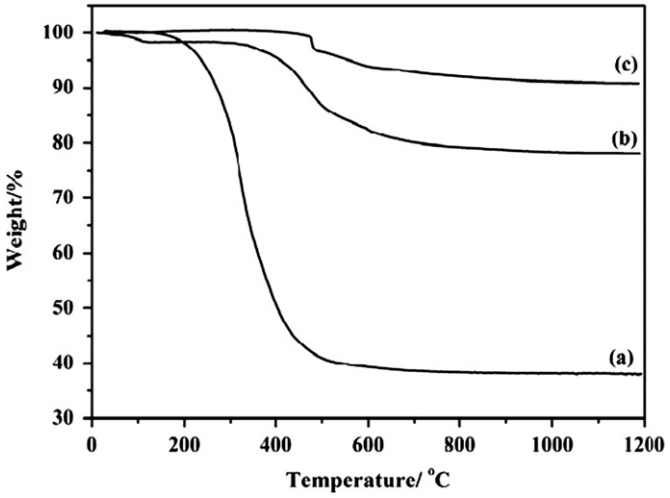


Fig. 2. TGA curves of (a) PMS under Ar atmosphere, (b) adhesive under Ar atmosphere, and (c) adhesive under air.

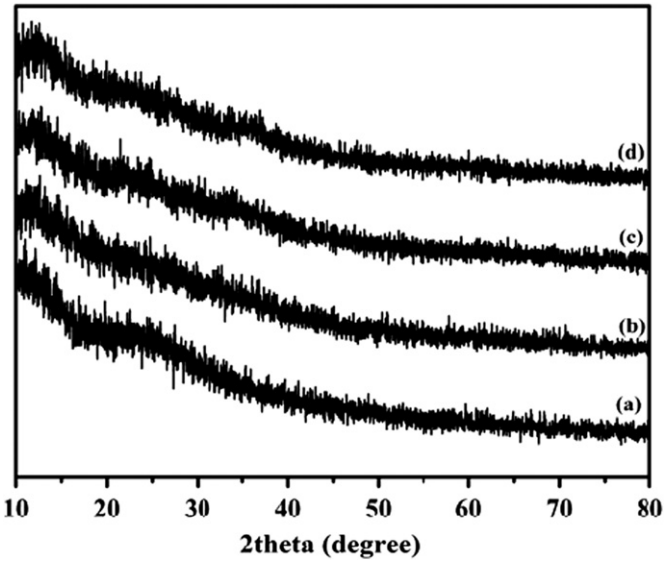


Fig. 3. X-ray diffraction patterns of adhesive heat-treated for 2 h in air at different temperatures ((a) 600; (b) 800; (c) 1000; (d) 1200 °C).

rate of 10 °C/min and Ar or 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 sintered SiC (Weifang Huamei Fine Ceramic Company) of size $20 \times 10 \times 5\ mm^3$. The adhesive was uniformly brushed onto the surfaces and the SiC specimens were bonded together at room temperature. The bonding area was $10 \times 10\ mm^2$. All of the bonded samples were cured at 200 °C for 2 h. 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 bonding strengths of the SiC joints treated at different temperatures were measured with a universal mechanical testing machine with a load rate of 0.5 mm/min at room temperature. The method for testing the bonding strengths of the SiC joints were performed as the literature published [3].

3. Results and discussion

After heat-treatment at 120 °C for 3 h, the molecular weight of the polymer increased significantly and the numerical values of Mn and Mw were augmented from 293 and 707 to 550 and 3309, respectively. The FTIR spectra presented in Fig. 1 indicate that PMS reacts with D4Vi by hydrosilylation, as evidenced by the new