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Interfacial reactions and joining characteristics of a Cu–Pd–V system filler alloy with C_f/SiC composite

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Abstract

A novel composition of Cu–Pd–V filler alloy was designed for the joining of C_f/SiC composite. The filler alloy was fabricated into brazing foils with a thickness of 0.15 mm by a rolling process. The alloy′s wettability on the C_f/SiC composite was studied with the sessile drop method. After heating at 1473 K for 10 min the Cu–Pd–V filler alloy showed a low contact angle of 6° on the composite. A $VC_{0.75}$ reaction band was formed at the surface of the C_f/SiC composite under the brazing condition of 1443 K /10 min, and the microstructure in the central part of the joint was composed of (Cu, Pd) solid solution and eutectic-like phase of $Pd_2Si + Cu_3Pd$. The interfacial reaction mechanism is discussed. The room-temperature three-point bend strength of C_f/SiC–C_f/SiC joints brazed with Cu–Pd–V filler alloy at 1443 K for 10 min is 128 MPa, and the joint strengths at temperatures of 873–1073 K are even higher than the room-temperature strength. The presence of refractory Pd_2Si compounds within the joints should contribute to the stable high-temperature joint strengths.

Keywords: A. Joining; Ceramic matrix composites (CMC); Interface diffusion; Reaction

1. Introduction

At present, an interesting carbon fiber-reinforced silicon carbide matrix composite — C_f/SiC composite, has appeared as a new kind of ceramic matrix composites (CMCs). However, the practical use of ceramics or CMCs is often limited by joining techniques. Requirements for these joints include sufficient mechanical strength and the possibility of withstanding high temperatures.

It is well known that brazing is a suitable joining method for ceramic or CMC. The main limitation of brazing for joining ceramics or CMCs is finding proper braze compositions that have desirable properties. However, the progress in the development of high-temperature brazes for C_f/SiC composite joining is rather slow.

Cu–Ti or Ag–Cu–Ti brazing alloys have been the base of C_f/SiC composite joining [1–3]. Furthermore, carbon fiber-reinforced Ag–Cu–Ti brazing material and mixed powders of Cu, Ti and graphite are also used respectively to join C_f/SiC with Ti alloys [4,5]. However, obviously, the service temperature of Cu–Ti and Ag–Cu–Ti system brazing alloys is still a problem.

It was also reported that a kind of Ni-base alloy was used as an interlinker in liquid infiltration joining of 2D C_f/SiC composite [6], but the results showed that at a joining temperature of 1573 K with a holding time of 45 min, the highest three-point flexural strength of the corresponding joints was only 58 MPa. Evidently, the study of high temperature brazes for C_f/SiC joining is far insufficient [7].

To our knowledge, vanadium can be used as an active element for ceramic joining [8–11]. The authors previously investigated the wettabilities of V-active PdCo(Ni)-based alloys on Si_3N_4 and SiC ceramics [12–14]. Recently the authors used a Pd–Co–V powder mixture as the filler metal for C_f/SiC joining, with the brazing parameters of 1523 K/20 min [15]. It was found that the element V played an active role in the joining. Though the corresponding joint strength was only 19 MPa, the feasibility of joining C_f/SiC composite with V-active brazing alloys had been verified.

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In this study we proposed a Cu–Pd–V alloy as a brazing filler metal for the joining of C_f/SiC with C_f/SiC. It is well known that the metal Pd possesses good thermal and oxidation resistance, and Pd-based filler metals have been primarily investigated [16–18]. Therefore Pd should be able to improve the high-temperature property of Cu-based brazing alloys. More importantly, it has been recognized that the element Pd has a large affinity with Si [12,15,19,20]. When Pd with a rather high concentration is added into the brazing alloy, it is likely for Pd atoms to react with Si atoms decomposed from SiC ceramic, resulting in the formation of Pd–Si compounds within the joint during the brazing process. It is believed that the refractory Pd–Si compound formation is favorable towards the joint strength and high-temperature stability.

The objective of the present paper is to study the microstructures and mechanical properties of C_f/SiC–C_f/SiC joints brazed with a Cu–Pd–V system filler metal. This study was intended to address the questions such as what reaction products would result in the composite joint using the Cu–Pd–V brazing alloy, whether the newly-developed brazing alloy can offer a stable high temperature strength for the composite joints, and what would be the relationship between the reaction products within the joint and the high-temperature joint strength.

2. Experimental procedure

We designed a novel composition of filler alloy, Cu–(35.0–42.0)Pd–(6.0–10.0)V (in wt%). The room-temperature plasticity of the filler alloy is very good so that it can be fabricated into brazing foils by an alternating room-temperature rolling process and vacuum annealing treatment. The final thickness of the foils is 0.15 mm.

3-D C_f/SiC ceramic matrix composites were used respectively as the wetting substrate and the joined material. The C_f/SiC composite consists of carbon fibers embedded in a silicon carbide matrix, with about 10.0 vol% porosity. The production process of this composite is based on the Polymer Infiltration Pyrolysis (PIP) process [21].

The wettability of the Cu–Pd–V brazing alloy on C_f/SiC composite was studied with the sessile drop method. The size of the used C_f/SiC substrate is $10 \times 10 \times 2$ mm^3. The chamber of the furnace was first evacuated to 1.5×10^{-3} Pa at room temperature. Then, the vacuum system was turned off, and high-purity (99.999%) argon was introduced into the chamber with a pressure of 0.1 MPa. Subsequently the chamber was evacuated to 1.5×10^{-3} Pa again, the vacuum system was turned off, and argon was introduced into the chamber for the second time. The brazing alloy sample was heated to 1473 K from ambient temperature at a heating rate of 8–10 K/min, and was held at this temperature for 10 min. During the heating process, the morphologies of the molten droplet were recorded dynamically by taking sideways photos at intervals when heated and held at the temperature of 1473 K. The contact angles of the brazing alloys on the composite were thus calculated from these droplet images using a computer method based on the Laplace equations. The accuracy of the contact angle measurements should be within $\pm 1^\circ$.

The samples to be brazed were vertically fixed in a specially designed graphite jig to form a butt joint of C_f/SiC–C_f/SiC. The size of the C_f/SiC composite bars to be joined is $3 \times 4 \times 20$ mm^3. The heating rate of the brazing experiment was 15 K/min. During the brazing experiment the vacuum was kept between 3.0×10^{-3} Pa and 7.0×10^{-3} Pa. After brazing the cooling rate was 15 K/min down to 673 K, then about 3 K/min to room temperature. The joint strength was determined by a three-point bend test in air at room temperature and at high temperatures. Five specimens were tested for each experimental condition, and the joint strength is given in the form of error bars, in which the value of the upper line has the maximum strength among the five specimens, the value of the lower line corresponds to the minimum strength, and the point in the middle of the error bar represents the average strength.

A JEOL-5600LV (Akishima City, Japan) type scanning electron microscope (SEM) was used for the microstructure observation of the brazed joint. Furthermore, an Oxford Inca type X-ray energy-dispersive spectrometer (XEDS) was used for semi-quantitative composition analysis of some microzones. The sample used in this analysis was coated with a thin layer of gold. So 0.20–0.60 at% Au was also detected in those microzones. The listed data of the XEDS analysis in the present paper are the renormalized results by removing the element Au.

An X-ray diffraction (XRD) analysis was also performed on a simulated specimen of the C_f/SiC–C_f/SiC joint brazed at 1443 K/10 min. The simulated specimen was prepared by the following process: a 0.15-mm-thickness brazing alloy plate was placed on a C_f/SiC sample with a size of $8 \times 8 \times 2$ mm^3; then, it was put in a vacuum furnace, heated to 1443 K and held there for 10 min, followed by slow cooling. Subsequently, the unreacted brazing alloy at the outermost surface of the sample was removed and gradual polishing was conducted through the reaction zone at the sample surface, and XRD analysis was carried out successively on the surface step by step.

3. Results and discussions

3.1. Wettability of Cu–Pd–V alloy on C_f/SiC composite

According to a differential thermal analysis (DTA) result, the solidus and liquidus temperatures of the Cu–Pd–V alloy are 1402 K and 1407 K, respectively. It was found that, once the Cu–Pd–V alloy melted on the surface of the C_f/SiC composite, it showed a contact angle of 87°, indicating wetting with the composite (Fig. 1(a)). With the increase of the temperature, the contact angle became smaller gradually, and when the alloy sample was heated to 1455 K, its contact angle decreased to 65°. Further increasing the temperature caused rapid spreading of the molten droplet, that is, when the temperature was raised to 1473 K the contact angle remarkably decreased to 18°. After the holding time of 10 min at 1473 K the contact angle reached the equilibrium value of 6° (Fig. 1(b)). Fig. 2 presents the typical morphologies of Cu–Pd–V alloy molten droplet on C_f/SiC composite at different heating stages.