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In situ consume excessive Ti element and form fine Ti based compounds as reinforcements for strengthening C/C-TC4 joints

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ABSTRACT

Brazing C/C composites to TC4 is often associated with the problems of excessive Ti and high residual stress, which results in low-strength joints. To overcome these problems, AgCu brazing alloy incorporated with Si_3N_4 or SiO_2 particles was used to join the C/C composites to TC4 alloy. The effect of Si_3N_4 or SiO_2 particles on the microstructure and mechanical properties of brazed joints was investigated in detail. Results show that Si_3N_4 or SiO_2 particles can react with Ti element from TC4, which is beneficial to inhibit too many brittle compounds at the C/C side. Meanwhile, it can in situ form the homogeneous distribution of fine-grained Ti based compounds during the brazing process. Due to their fine size, uniform distribution, and favorable cohesion with the matrix, the in situ Ti based compounds as reinforcements have excellent reinforcing effects in the brazing seam. Finite element analyze was adopted to investigate the residual stress state of the joins, and the result shows that the addition of silicide particles can reduce the residual stress obviously. The highest average shear strength reached about 45 MPa for Si_3N_4 and 41 MPa for SiO_2 particles additions, which was higher than that of joints brazed using pure AgCu.

1. Introduction

Due to the low density, very high thermo-elastic stability, moisture insensitivity and excellent oxidation resistance, carbon/carbon (C/C) composites have exhibited great potential for the thermal structure in the frontier of aerospace [1,2]. TC4 (Ti-6Al-4V) alloy has been widely applied to the field of aerospace owing to its superplasticity, low weight and high mechanical resistance [3,4]. Therefore, it is vital to join C/C composites with TC4 alloy for further practical applications.

Among various joining techniques, brazing is the most common method for joining C/C to TC4 due to its simplicity, low cost, and mass production capability [5,6]. However, brazing C/C composites still suffers from two following problems when the metallic part is TC4. One of the major issues about brazing ceramics, including C/C composites, to metals has been an appearance of an excessive residual stress in the joint, as a result of large differences in the coefficients of thermal expansion (CTEs) between C/C and TC4 [7,8]. Additionally, brazing ceramics with TC4 alloy often suffers from the problem of excessive Ti from the dissolution of TC4, which results in forming too thick reaction layer at the ceramic side and consequently deteriorates the joint strength [9,10]. Therefore, it is vital to alleviate the residual stress and consume or control the excessive dissolution of Ti element during brazing C/C composites with TC4 alloy.

As for reducing the residual stress in the joints, typical solution would be adding tiny ceramic particles or fibers with low CTE into traditional brazing alloys as reinforcements to improve the mechanical properties [11,12]. However, the problems of uneven distribution and nonwetting of the reinforcements in the brazing seam should be further improved [13,14]. In order to efficiently consume too much Ti from TC4 side, Liu et al. demonstrated that Ni foil as interlayer can consume Ti element and form Ti-Cu-Ni compounds, which improved the mechanical properties of brazed joints [9]. Previous research has reported that Cu foil as interlayer can consume dissolved Fe element and inhibit forming brittle compounds at the ceramic side [15]. However, it may still form

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continuous brittle compounds to harden the original ductile brazes and preclude effective stress relief, which would lead to weaken the mechanical properties of joints [7]. To overcome the above inherent problems associated with conventionally performed joints, in situ synthesized reinforcements originated from the brazing process have been developed. Q. Qiu et al. [16] reported that added B element into AgCu alloy to braze Al_2O_3 ceramic to TC4 alloy. The reaction of B and Ti improved the microstructure in brazing seam and reduced the mismatch of CTE between ceramic and alloy so as to reduce the residual stress. And the shear strength of Al_2O_3-TC4 joints achieved 111 MPa, which increased 36% strength of samples brazed with pure AgCu alloy. T. Lin et al. [17] utilized the reaction of TiB_2 and Ti to form TiB whiskers. The addition of TiB_2 into CuNi brazing alloy can bond C/C composites and TC4 reliably. The formation of TiB whiskers can strengthen the seam and reduce the crack caused by residual stress. And the shear strength of C/C-TC4 joints was 18.5 MPa, which was 56% higher than that of joints brazed with CuNi alloy. W. Yang et al. [18] introduced Ti/Cu lamination to braze ZrB_2-SiC and TC4. B element diffusing from ZrB_2-SiC can react with Ti in liquid brazing alloy rapidly. The TiB as reinforcement improved the properties of joints and reduced the CTE of brazing alloy to decrease the residual stress, and the shear strength reached 90.7 MPa. The in situ synthesized reinforcements with uniform distribution can significantly alleviate the residual stress and improve the joint strength. X.Y. Zhao et al. [1] and X.G. Song et al. [19] introduced nano-Si_3N_4 into AgCuTi alloy to braze ceramic and Ti alloy. Si_3N_4 can consume Ti by the reaction during brazing process and acted as the nucleation site of Ti-Cu phase. The mechanical properties were enhanced by Si_3N_4 reinforcement.

In this paper, to overcome the formation of excessive brittle intermetallic at the C/C side and to reduce high residual stress of C/C-TC4 joint, micro Si_3N_4 or SiO_2 particles reinforced AgCu brazing alloy was developed to braze C/C composite with TC4 alloy. In addition, the effect of SiO_2 or Si_3N_4 particles on the joining properties and the fracture modes was also investigated. The decrease of residual stress by ceramic particles was investigated with finite element (FE) methods, and the residual stress in the joints can be measured and visualized to highlight the effect of adding ceramic particles through FE methods. Results show that introducing Si_3N_4 or SiO_2 particles can not only consume too much Ti dissolved from TC4 side, but also in situ form the homogeneous distribution of fine-grained Ti based compounds in the brazing seam, which shows a synergistic effect on reducing the residual stress and enhancing the mechanical properties of brazed joint.

2. Experimental procedure

The parent materials used in the experiment were 3D C/C composite and TC4 (TC4 is a grade of titanium alloy defined in China, and its nominal compositions is Ti-6Al-4V (wt.%)). Before brazing, the C/C composite and TC4 were cut into blocks with a dimension of 5 mm × 5 mm × 5 mm and 16 mm × 12 mm × 3 mm respectively. Micro-Si_3N_4 particles (<5 μm) with purity of 99.9% or SiO_2 (<5 μm) with purity of 99.9% were added into Ag-28Cu (wt%) alloy powder (<45 μm) with purity of 99.9% as the particles to powder ratio was 1:24 to make the content of ceramic particles reach 4 wt%. Then the composite filler was fabricated by a planetary ball mill at room temperature. The ball to powder weight ratio was 15:1, the rotation speed of vial was 300 rpm and the milling time was 2 h. Fig. 1 shows the method to press powder into foil by tablet machine. 0.15 g pure AgCu powder is added into fixture and then pressed to attain 300 μm foil. The mixed composite filler (containing 4 wt% Si_3N_4 or SiO_2) can get the almost equal thickness foil through the above process. And the fixture above can limit the size of brazing foil as 7 mm × 7 mm. So the weight of pure AgCu alloy is

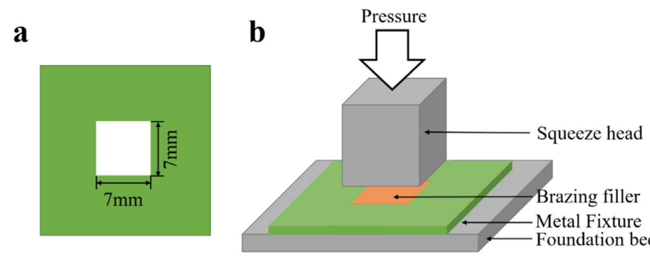

Fig. 1. Schematic diagrams of (a) metal fixture and (b) tablet machine.

almost same in each group, which benefits to the research on the effect of ceramic particles. All of the joined surfaces on the samples were polished by SiC paper up to 1000 grit and were ultrasonically cleaned in acetone for 20 min. The as-milled composite filler (AgCu + Si_3N_4, AgCu + SiO_2 or AgCu) was sandwiched between the parent materials, and 0.7 MPa pressure was applied to maintain the close contact of brazing surface by carbon fixture. The brazing was carried out at 850 °C for 5min in a vacuum furnace. The pressure of furnace was less than 5×10^{-3} Pa during brazing process. Then the specimen was heated to 700° at 10 °C/min, and next heated to brazing temperature at 3 °C/min. When holding time was over, the furnace was cooled down to room temperature at 5 °C/min.

The obtained samples were characterized by scanning electron microscope (SEM) coupled with an energy dispersive spectrometer (EDS) for the morphology and microstructure observations. The phase constituents of obtained samples were identified by micro-XRD (JDX-3530 M). Samples brazed with pure AgCu were grinded to the interface of composite/braze to confirm the composition of reaction layer. While samples brazed with AgCu + Si_3N_4 or SiO_2 particles were grinded to the brazing seam to ascertain the resultant of alloy and ceramic. Mechanical strength of brazed joints was evaluated by shear tests using a universal tensile strength testing machine (Instron 1186) at a rate of 1 mm/min, and the schematic diagram of shear strength test is shown in Fig. 2. The contact surface of the steel clamping apparatus was grounded by 600 grit SiC paper and then cleaned by acetone to avert the influence of friction force between the brazed joint sample and the fixture. Five samples of each group were tested to get the average of the shear strength. The fracture surfaces of the joints were also investigated by optical microscope (VHX-1000E).

3. Results and discussions

Fig. 3a–c shows the morphologies of obtained AgCu, AgCu + Si_3N_4 and AgCu + SiO_2 fillers. Compared with pure AgCu,

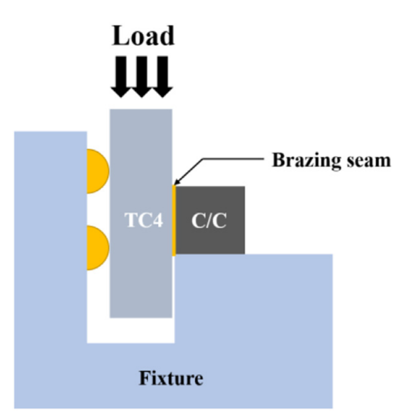

Fig. 2. Schematic diagrams of shear test experiment.