Original Article

In-Situ synthesized TiC nano-flakes reinforced C/C composite-Nb brazed joint

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A R T I C L E   I N F O

Keywords:
Brazing
C/C composite
Residual stress
Polymer carbonization
In-situ TiC nano-flakes

A B S T R A C T

Brazing C/C composite to Nb is often associated with the problem of high residual stress, resulting in low-strength joints. To overcome these problems, here we carried out a simple polymer carbonization process to acquire uniform carbon-covered Cu composite interlayer, which was subsequently used for soundly brazing C/C composite and Nb with the assembly of C/C composite/Ag-Cu-Ti foil/Cu foam/Ag-Cu-Ti foil/Nb. Microstructure and mechanical properties of the joints were well investigated. The carbonization reacted with Ti elements, forming uniformly distributed in-situ TiC nano-flakes in the joint seam by virtue of the porous Cu foam skeleton. Results present that the in-situ TiC nano-flakes not only greatly reduced the thermal expansion coefficient but also effectively impeded the Cu solid solutions agglomeration. The average shear strength of the joint brazed with 3% C-Cu (wt.%) foam interlayer reached ~52.8 MPa with the brazing temperature of 880 °C for 10 min.

1. Introduction

C/C composites have demonstrated their enormous potential as thermos-structural materials in the aerospace, aviation and industrial applications fields for uses such as heat shields, turbine engines, nozzles and hot press dies due to their low density, excellent oxidation resistance, and high-temperature strength [1,2]. In many applications, the joining of C/C composites to refractory metals, such as niobium, is necessary to fabricate complex structural components [3]. A variety of methods have been developed to materialize the joining of C/C composites to metals such as thermal diffusion bonding, adhesive bonding as well as brazing. Among the current joining technologies for joining C/C composites to metals, brazing has excited extensive attention due to its simplicity, high joint strength, low cost-effectiveness as well as perfect adaptability of joint size and shape [4]. However, C/C composites and metals have significant differences in chemical and physical properties, which make it difficult to acquire a sound brazed joint. In particular, the evident mismatch of their coefficients of thermal expansion (CTE) between C/C composites and metals or brazing alloys always causes high thermal residual stress in brazing joints [5], resulting in the weakening of the brazed joints.

To overcome the problem of high residual stress in brazed joints, many studies suggested that the composite-metal joints could be soundly brazed with low CTE reinforcements or soft interlayers modified composite alloys, such as Al₂O₃ [4], SiC [5], TiB [6], TiC [6–8], Cu foil [9], Ni₇₁Cr₅Si foil [10], Mo foil [11]. Results showed that these reinforcements could indeed reduce the CTE of the joint seam, modestly control the interfacial microstructure and slightly enhance the joint plasticity, all of which contributed to strengthening the joint. However, most of the brazing alloys contain multiple elements, thus the complex mixing process of the brazing alloy powders as well as the thick flat-like foil structure limited the consistency and accuracy of the reinforcements distribution. Therefore, these kinds of composite brazing alloy systems are difficult to achieve for use in industrial applications. For this case, some researchers utilized metal foam as an interlayer to well control the interfacial phases. Lin et al. [12] reported introducing Cu foam as an interlayer could readily acquire homogenously distributed fine-grained Ti-Cu compounds to effectively mitigate the residual stress and consequently strengthen the joint. Zhu et al. [13] used Ni foam as an interlayer to refine the distribution of Ti element in the interface and consequently restrain the formation of brittle intermetallic compounds, which drastically improves the performance of the brazed joint.

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https://doi.org/10.1016/j.jeurceramsoc.2017.11.059
Received 20 July 2017; Received in revised form 28 November 2017; Accepted 29 November 2017
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Please cite this article as: Wang, Z.Y., Journal of the European Ceramic Society (2017), https://doi.org/10.1016/j.jeurceramsoc.2017.11.059
Nevertheless, the foam substrates in their studies collapsed or agglomerated by complicatedly reacting with the brazing alloy, triggering the waste of the good plasticity of the metal foams. By this token, it is undoubtedly conceivable that using a metal foam is a simple way to materialize even dispersion of the reinforcements in the brazing seam. Meanwhile, the homogenous distribution of the foam substrate in the joint seam is also vital for acquiring high-quality brazed joints.

Furthermore, many studies reported uniformly prepared reinforcements on inner-connective porous substrate, which owns highly consistent and repeatable results. Gierszal et al. [14] reported a method of carbonizing phenolic resin film on colloidal silica substrate to obtain uniformly distributed porous carbon shells with full coverage on the surface of the silica balls. Xiong et al. [15] fabricated a nacre-like reduced graphene oxide reinforced Cu matrix composite based on a preform impregnation process, which allowed uniform dispersion and alignment of graphene by virtue of the unique substrate.

From the researches above we realize that by introducing in-situ reinforcements coated metal foam may exert their synergistic enhancement. Namely, the in-situ prepared reinforcements could be evenly dispersed in the joint seam with the help of the unique foam substrate. In the meantime, the agglomeration of the metal foam substrate could also be well impeded by the evenly dispersed reinforcements as obstructions. In this work, we developed a highly consistent strategy of fabricating an in-situ prepared carbonized phenolic aldehyde reinforced Cu foam composite (C-Cu foam) for brazing C/C composite and Nb. In particular, the carbonization shell possessed controllable and uniform thickness by controlling the polymer content and using CVD method. Moreover, the in-situ prepared carbonization and Cu foam substrate in the C-Cu foam was expected to exert synergistic reinforcing effect to effectively alleviate the residual stress and consequently significantly strengthen the brazed joint.

2. Experimental

The overall experimental procedure for the fabrication of the C-Cu foam is outlined in Fig. 1a. At first, the phenolic aldehyde resin (guaranteed reagent, obtained from Tianjin Fuyu Fine Chemical Co., Ltd.) was mixed with acetone (analytical reagent, obtained from the Sinopharm Chemical Reagent Co., Ltd.). Specifically, 100 ml phenolic aldehyde resin acetone solutions with different mass fraction (5%, 15%, 25%, 50%) was obtained. Then, a pure Cu foam (98% porosity, 50 PPI, thickness of 0.3 mm, supplied by Suzhou Jiashide Metal Foam Co., Ltd.), which owns porous structure with interconnected 3D scaffold, was dipped in this solution, which was stirred for 2 h for better dispersion and adhesion. The soaked Cu foam was then dried in vacuum at 40 °C for 3 h to obtain the pure phenolic aldehyde resin coated Cu foam. The As-prepared Cu foam sample was inserted into a 2-inch quartz tube and the quartz tube was subsequently loaded inside a vacuum chamber. The As-prepared Cu foam sample was weighed before and after the carbonization process to calculate the mass difference. After that, the sample was subjected to carbonization at a heating rate of 10 °C min⁻¹ up to the temperature of 800 °C and maintained for 3 h, under purging Ar gas at a flow rate of 100 sccm. After that, the system was naturally cooled down to room temperature with a cooling rate of 5 °C per min to ensure a slow cooling with Ar environment maintained. In this way, a C-Cu foam was successfully obtained.

The C/C composite used in this study was semi-3D C/C composite, fabricated by needled felt and carbon fiber cloth in our laboratory. It was cut into 5 mm × 5 mm × 5 mm. The Nb brazing specimens (> 99.9% pure, obtained from the Northeast Nonferrous Metals Co., Ltd.) were 10 mm × 10 mm × 3 mm slices (for metallographic analysis) and 25 mm × 10 mm × 3 mm (for shear strength testing). The filler metal were Ag-26.7Cu-4.6Ti (wt.%) foils with the size of 5 mm × 5 mm (thickness 100 μm, weighed ~0.0476 g), obtained from the Lukesi Brazing Materials Co., Ltd. (thickness of 100 μm, weighed ~0.0476 g) to braze C/C composites and Nb. Before brazing, the bonding surfaces of both C/C composite and Nb samples were ground by 800 grit silicon carbide grit paper and then ultrasonically cleaned in acetone.

The brazing assembly structure was Nb/Ag-Cu-Ti foil/C-Cu foam interlayer/Ag-Cu-Ti foil/C/C composite, as shown in Fig. 1b and c. For brazing process, the comparison experiments were performed with two Ag-Cu-Ti foils sandwiched between the parent materials. C-Cu foam and pure Cu foam were both used as interlayer for brazing. In order to keep the specimens in close contact, the system was held by a graphite jig with slight pressure of ~5 MPa to fix all the parts. The assembly was then heated to 880 °C with a rate of 10 °C min⁻¹ in a vacuum furnace, isothermally held for 10 min, and then cooled down to room temperature at the rate of 5 °C min⁻¹.

The morphology and microstructures of the obtained samples were characterized using optical microscope, scanning electron microscope (SEM) coupled with energy dispersive spectroscopy (EDS), Raman spectroscopy, transmission electron microscope (TEM), high resolution TEM (HRTEM) and X-ray diffraction (XRD). Mechanical tests were performed to evaluate the shear strength of the brazed joints.

3. Results and discussion

The optical micrographs of pure Cu foam and the C-Cu foams were given in Fig. 2a. It could be clearly seen that the external features of Cu foams, coated with different weights of carbonized phenolic resin, vary from each other. Along with the mass increasing of the carbonized phenolic resin, the surface color of the obtained C-Cu foams is getting darker, suggesting the Cu foam is gradually covered by thicker and thicker carbon shell. As shown in Fig. 2a, the corresponding mass fraction of the carbon in each C-Cu foam was calculated to be 1.0% (1% C-Cu foam), 3.0% (3% C-Cu foam), 5.0% (5% C-Cu foam) and 10%...
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