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Carbon nanotubes-reinforced Ni foam interlayer for brazing SiO_2-BN with Ti6Al4V alloy using TiZrNiCu brazing alloy

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ABSTRACT

Brazing SiO_2-BN with Ti6Al4V is often associated with the problems of high residual stress and excessive Ti-based compounds formed. To overcome these problems, we report a new type interlayer of carbon nanotube (CNT) reinforced Ni foam fabricated by plasma enhanced chemical vapor deposition. The in-situ grown CNT are homogenously dispersed on 3D structure Ni foam, which could effectively avoid damage and agglomeration in the brazing seam. Result shows that Ni foam could consume excessive dissolved Ti, and CNT was beneficial for restricting the growth of phase, improving joining strength and releasing residual stress rapidly. The average shear strength of the joint brazed with CNT-Ni foam is about 50 MPa, and this value is about 5 times higher than that of joints brazed with pure TiZrNiCu. Further, a new simulated experiment was carried out innovatively to solving the difficulty of investigating interfacial behavior of CNT in brazing seam. Then the results clarify that integral structure of CNT can prevent the reaction with Ti. In situ growth of CNT on Ni foam could provide a way for introducing CNT into brazing seam without damaging structure of CNT.

1. Introduction

SiO_2-BN ceramic reinforced by BN particles is a newly silicide matrix composite promisingly utilized in the field of aerospace due to its low density, low coefficient of thermal expansion (CTE), good mechanical strength and high thermal shock resistance [1,2]. To avoid the fabrication difficulty of monolithic bulks, the application of SiO_2-BN ceramic always needs to be joined to aerospace metal such as Ti6Al4V, which possesses high mechanical strength, light weight and good thermal properties [3,4]. Among various ceramic-metal joining technique developed currently, brazing exhibits prominent advantages of high joining quality, convenience and low cost-effectiveness [5,6]. However, there are two main problems for brazing SiO_2-BN to Ti6Al4V. Firstly, the CTE mismatch between two base materials inevitably induces high residual stress, which impairs the integrity of joint severely [7,8]. Secondly, Ti element dissolves from Ti6Al4V into brazing seam excessively during brazing process. This process results into a large amount of brittle and continuous compounds among brazing seam which are detrimental to integrity of the joints [9]. Therefore, it is crucial to release residual stress and control the formation of brittle Ti-based compounds to improve mechanical properties of SiO_2-BN/Ti6Al4V joints.

In order to alleviate residual stress, introducing low-CTE reinforcement (such as ceramic particles, fibers, and carbon nanotube, etc.) into brazing alloy is adopted [10–12]. Compared with traditional reinforcements, carbon nanotube (CNT) possesses out-standing mechanical properties, extraordinarily low CTE and low density, which shows the great advantages in the field of brazing [13–15]. Many researches [16–18] found out that the addition of CNT into Sn matrix solder would dramatically improve the mechanical properties. Kuzumaki et al. [19] and Xue et al. [20] investigated Ti matrix composites reinforced by CNT and found out that CNT can modify mechanical strength and Young's modulus of Ti composites and greatly reduce the CTE of Ti composites. It is feasible to fabricate CNT-reinforced Ti-based brazing alloy according to Ti matrix composites above. But there are still two main problems impairing the effects of CNT in the alloy. On one hand, when the content of CNT is increased, it is difficult to disperse CNT among brazing seam homogenously. Agglomeration of CNT may weaken the reinforcing effect and act as defect point instead [21]. On the other hand, traditional ball-milling mixture can disperse the

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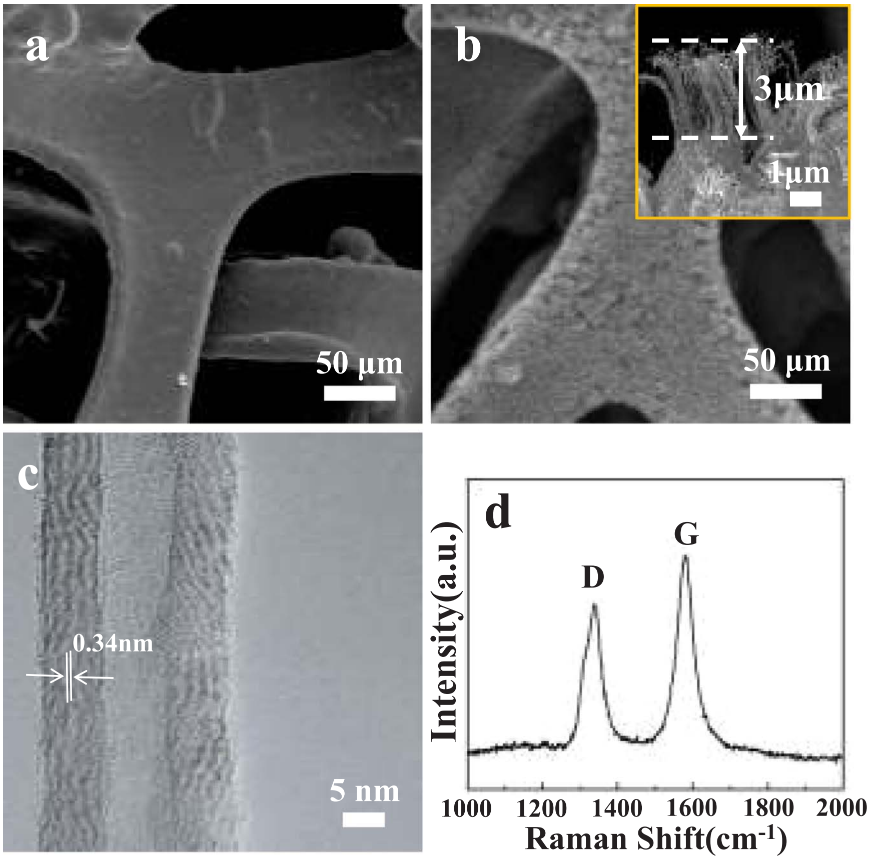

Fig. 1. SEM images of (a) pure Ni foam and (b) CNT-Ni foam. The inset of (b) is SEM image of obtained CNT. (c) HRTEM image of the microstructure of CNT. (d) Raman spectra of pure Ni foam and CNT-Ni foam.

CNT in some degree, but cannot avoid destroying the microstructure of CNT, which results in the reaction between active elements and CNT [19]. Therefore, it is significantly to overcome the problems above to ensure the reinforcing effect of CNT in brazing seam.

Moreover, to control the formation of brittle and continuous Ti-based compounds, soft interlayer is introduced to prevent Ti from diffusing to the ceramic side. Brochu et al. [22] pointed out that Cu interlayer can consume Ti in the brazing alloy, inducing the decrease of brittle compounds. Liu et al. [8] investigated the SiO_2/Ti-6Al-4V joint brazed with AgCu, and found that introducing Ni foil could hinder Ti atoms diffusing to the ceramic side by reaction effectively, which improved the mechanical strength of joints remarkably. It is obvious that metal foil interlayer can control the size of compounds at ceramic side and maintain good joining strength between ceramic and brazing seam. But the reaction between Ti and interlayer would induce the formation of continuous compounds along brazing seam, which is a potential threat to the integrity and mechanical properties of the joints. Therefore, it needs to find a way to control or dilute the content of Ti without the formation of continuous compounds.

In this paper, CNT-Ni foam interlayer was fabricated by plasma enhanced chemical vapor deposition (PECVD) and then sandwiched between TiZrNiCu foils for brazing SiO_2-BN ceramic and Ti6Al4V alloy. Ni foam could in-situ consume the dissolution of Ti from Ti6Al4V and disperse compounds uniformly in the brazing seam because of its unique 3D structure. Meanwhile, the great specific surface and 3D scaffold of Ni foam can provide CNT with in-situ grown matrix, and promote the uniform distribution of CNT. And CNT can control the growth of phases and decrease the CTE mismatch to release residual stress, which would improve mechanical properties of joints effectively. Moreover, the interface behavior of CNT with brazing alloy was investigated in detail.

2. Experimental

The Ni foam (more than 95% porosity, 50 PPI) with the thickness of 0.5 mm acted as the interlayer. The CNT-Ni foam interlayer was synthesized by PECVD [23]. Prior to PECVD, the Ni-foam with nickel particles catalyst was fabricated via the method of a homogeneous deposition-precipitation process [24]. The Ni foam samples were dipped in 0.1 M $Ni(NO_3)_2{\cdot}6H_2O$ (99.9% purity) ethanol solution with constant stirring for 1 h, which covers the surface of Ni foam with the Ni-based particles uniformly. The dry $Ni(NO_3)_2$-Ni foam was placed at the same horizontal level in a PECVD reaction chamber. The PECVD chamber was evacuated to a pressure below 5 Pa, and then the chamber was maintained at 200 Pa with H_2 at a rate of 25 sccm. To grow the CNT, the chamber was heated to 700 °C for 15 min plasma treatment with CH_4/H_2 mixture at the rate of 40/10 sccm. Final, a rapid cooling process can obtain high quality CNT.

The SiO_2-BN ceramic used is fabricated by hot pressing sintering, which is composed of SiO_2 + 60 wt% BN powder. The ceramic and Ti alloy were cut at the dimension of 5 mm × 5 mm × 5 mm and 12 mm × 16 mm × 3 mm respectively. The brazing alloy used in this paper is Ti-35%Zr-15%Ni-15%Cu (wt%) foil at 30 μm thickness. All the surface to be jointed was grounded by SiC papers up to 1500 grit. And then all the materials were ultrasonically cleaned in ethanol to remove impurity. The SiO_2-BN and Ti6Al4V were assembled as two TiZrNiCu foil, TiZrNiCu/Ni foam/TiZrNiCu and TiZrNiCu/CNT-Ni foam/TiZrNiCu three group respectively, and graphite jip were applied to maintain the close contact of specimens. Then the assemblies were placed in the vacuum brazing furnace with the pressure less than 3×10^{-3} Pa during brazing process. The furnace was heated to 800 °C at 10 °C/min, and next heated to 970 °C at 3 °C/min. The brazing temperature was held for 10 min, and then cooled down to room temperature at a rate of 5 °C/min.

The morphology and structure of the samples were characterized by