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Vacuum brazing Nb and BN-SiO_2 ceramic using a composite interlayer with network reinforcement architecture

L.X. Zhang*, J.H. Yang, Z. Sun, X.P. Liu, J.C. Feng*

State Key Laboratory of Advanced Welding and Joining, Harbin Institute of Technology, Harbin 150001, China

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ABSTRACT

A novel composite interlayer with a reinforced network was designed using a SiC ceramic with a network structure and Ti-Ni-Nb composite filler foils, to which the Nb and BN-SiO_2 ceramic were successfully brazed under vacuum. For a brazing temperature of 1160 °C and holding time of 10 min, the interfacial microstructure of the Nb/BN-SiO_2 ceramic joint was Nb/(βTi,Nb)-TiNi eutectic structure+$(\beta Ti,Nb)_2Ni$+SiC+TiC/TiN+Ti_2N +TiB+Ti_5Si_3+TiO/BN-SiO_2 ceramic. In addition, the shear strength and nano-hardness were analyzed to evaluate the effect of the composite interlayer with a network reinforcement architecture on the mechanical properties of the joint. During brazing, the Ti-Ni-Nb filler metal infiltrated and reacted with the SiC to form the network reinforcement architecture, resulting in the residual stress being relieved and the mechanical performance of the joint being significantly improved. A maximum shear strength of 102 MPa was achieved, which was 60 MPa (142%) higher than that of the joint brazed without the network reinforcement architecture. A reduction in the residual stress on the BN-SiO_2 ceramic side from 328 MPa to 210 MPa was observed with the network reinforcement architecture, and the fracture path of the joint changed from the surface of the BN-SiO_2 ceramic to the interfacial reaction zone.

1. Introduction

The properties of SiO_2 ceramic can be improved by the addition of hexagonal boron nitride (*h*-BN) into the SiO_2 ceramic, [1]. Therefore, in recent years, BN-SiO_2 ceramics have attracted considerable attention in the aeronautic industry [2] due to their superior physical, chemical and mechanical properties. In particular, its unique low dielectric constant makes BN-SiO_2 ceramic suitable for astronautic applications, such as the electromagnetic radome or window [3–5]. However, the application of BN-SiO_2 ceramic for large and complex components is restricted by its poor formability. Thus, joining BN-SiO_2 ceramic with other materials, especially metals and alloys, can significantly broaden the range of potential applications.

To date, various techniques for joining ceramics and metals have been investigated, including mechanical fastening, brazing, transient liquid phase bonding, and diffusion bonding [6–8]. Among these techniques, active brazing is considered an optimal method for joining ceramics and metals due to its convenience and high reliability. In addition, brazing is one of the most useful techniques for improving the interfacial microstructure [9]. As BN-SiO_2 is a novel ceramic, most previous studies have focused on its fabrication. Only a few studies investigated the brazing of the BN-SiO_2 ceramic to Invar [10] or Ti [11] using the Ag-Cu-Ti filler metal. However, the relatively low melting temperature and poor oxidation resistance of the Ag-Cu-Ti filler metal mean that it is unsuitable for high temperature applications. In addition, previous result [10] showed that BN-SiO_2 ceramic and metal had significantly different in their coefficients of thermal expansion (CTE, for short), which led to high residual stresses and low strength of the BN-SiO_2 ceramic/metal joint. Hence, there is considerable interest in relieving the residual stress of the joint when brazing BN-SiO_2 ceramic to Nb at high temperature.

Up to now, many techniques have been developed to moderate the residual stress of brazed joints, such as adding a low CTE material to brazing filler metals [12] or using soft interlayers [13]. A few studies reported that the addition of fibers [14,15] or particles [16,17] to the filler metals could effectively alleviate the residual stress. However, such additives are generally not distributed homogenously in the brazing seam, which deteriorates the quality of the joints [18]. Thus, the addition of a soft interlayer is the most suitable method for moderating the residual stress of the brazed joint. For example, single Ti foil [19] and multilayer metallic foils (Ti foil+Cu foil+Mo foil) [20] were used as buffer layers to join ceramic with metal, and the average strength of the ceramic-metal joint was improved significantly. Additionally, metallic foams of Ni foam or stainless steel foam have

* Corresponding authors.
E-mail addresses: hitzhanglixia@163.com (L.X. Zhang), fengjc@hit.edu.cn (J.C. Feng).

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been applied as the interlayer to form metal-ceramic bonds and the joints exhibited good mechanical properties and thermal shock resistance [21,22]. Jarvis et al. [23] proposed that using metallic foam as an interlayer could divide the brazing seam into multiple zones, decreasing the strain energy by redistributing the stress and plastic strain in the ductile interlayer. However, the propagation of the wave was obstructed and the electrical transparency deteriorated when using the metallic interlayer [24].

To satisfy the requirements for the application of an electromagnetic radome, a composite interlayer with network reinforcement architecture (for convenience, it is named as CINRA in this research) was designed with a SiC ceramic with a network structure (for convenience, we name it as SNS) and Ti/Nb/Ni composite filler foils, which we propose could reduce the residual stress on the BN-SiO_2 ceramic side without destroying the electrical transparency property of the joint.

In this study, the Nb and BN-SiO_2 ceramic were brazed under vacuum with the CINRA. The effects of the CINRA and brazing temperature on the microstructure and mechanical properties of the Nb/BN-SiO_2 ceramic joint were investigated in detail. In addition, the residual stress on the BN-SiO_2 ceramic side was estimated to analyze the effect of the CINRA on the mechanical properties of the joint.

2. Experimental procedure

The BN-SiO_2 ceramic was processed by hot pressing a mixture of BN and SiO_2 powders with a volume fraction of BN of 60%. The relative density of the BN-SiO_2 ceramic was 95% after hot pressing. Commercially available Nb with a purity of 99.96% was used for brazing. The SNS with 80% porosity was produced by Shanghai Yubei Precision Ceramic Company. The morphologies of the BN-SiO_2 ceramic and the SNS are shown in Fig. 1, where it can be seen that the SiO_2-BN ceramic was composed of amorphous fused silica and hexagonal boron nitride (*h*-BN). The average diameter of the uniformly distributed pores in the SNS was about 0.2 mm.

The CINRA used to braze the BN-SiO_2 ceramic and Nb was composed of Ti foil (100 μm thick), Ni foil (50 μm thick), Nb foil (100 μm thick) and the SNS (2 mm thick). The CINRA in the form Ti +Nb+Ni+SNS+Ti+Ni were sandwiched between the BN-SiO_2 ceramic and Nb, as shown in Fig. 2.

Before brazing, the BN-SiO_2 ceramic and metal foils were cut into pieces with dimensions of 6×3×2 mm and 10×10×3 mm, respectively. The bonding surfaces of the samples were coarsely polished using 1000#SiC grit paper. Then, all the samples were ultrasonically cleaned in acetone for 10 min prior to brazing and dried at room temperature. The brazing was performed at 1140–1200 °C for 10 min in a vacuum furnace (Centorr-3520) under the vacuum of 2.5×10^{-3} Pa. The heating rate was 10 °C/min and the cooling rate was 2 °C/min to 1000 °C and then 5 °C/min to 200 °C. A small pressure of 0.5–0.6 MPa was applied

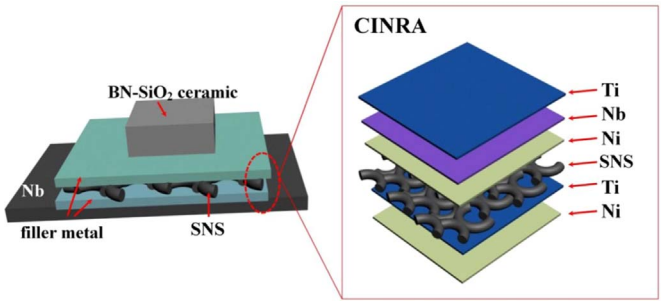

Fig. 2. Schematic diagram of the brazed assembly.

to prevent the specimens from moving during brazing.

The microstructures and chemical compositions of the joints were studied by scanning electron microscopy (SEM; Hitachi S-3400) equipped with an energy dispersive spectroscopy (EDS) system. Transmission electron microscopy (TEM; Tecnai G2 F30) was used to further characterize and identify the structure of the reaction products. Thinning of specimens for TEM was performed using a focused ion beam (FIB; Helios NanoLab 600i). An X-ray diffraction spectrometer (XRD; D8-Advance) system equipped with Cu-Kα radiation was employed at a voltage of 40 kV to identify the reaction phases of the joint. The strength of the joint was measured by shear tests using an Instron5569 mechanical testing machine with a constant speed of 0.5 mm/min at room temperature. The average strength was determined by measuring at least five samples under the same conditions and the sketch of the shear test was shown in Fig. 3. In addition, the hardness and elastic modulus across the joint were characterized using a nanoindenter to evaluate the performance of the reaction products. The fracture surfaces were observed using an optical microscope (OlympusSZX12).

3. Results and discussion

3.1. Microstructure of the Nb/CINRA/BN-SiO_2 ceramic joint

The SEM micrographs presented in Fig. 4 show a typical microstructure of the Nb/CINRA/BN-SiO_2 ceramic joint brazed at 1160 °C for 10 min. It can be seen that a defect-free joint was successfully obtained. It is evident from Fig. 4a and d that the brazed joint contained two distinct zones (for the sake of convenience, the zones are marked I and II, respectively). Zone I was an infiltration zone including the Ti-Ni-Nb filler metal and SNS (as shown in Fig. 4a), and zone II was a continuous thin reaction zone adjacent to the BN-SiO_2 ceramic (as shown in Fig. 4d).

In order to further investigate the microstructure of zone I, higher magnification images of the characteristic microstructures are shown in Fig. 4b and c, respectively. In addition, the results of the elemental

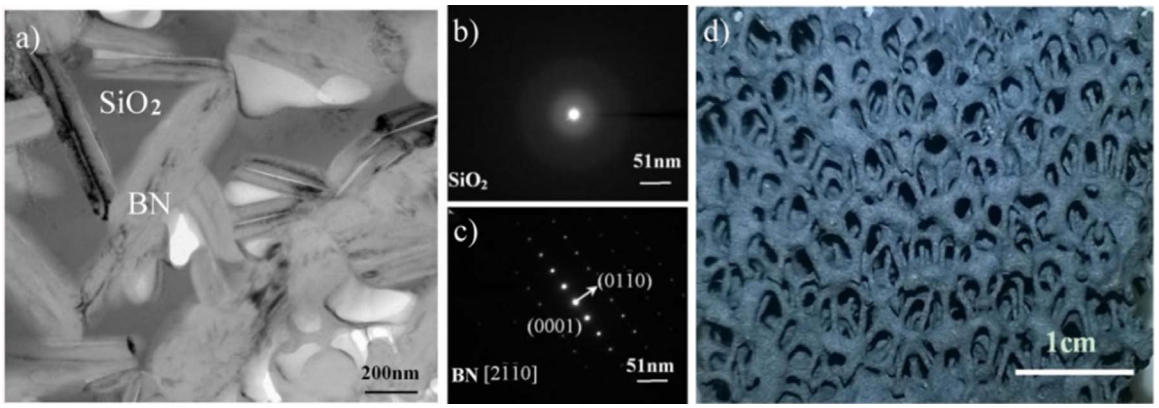

Fig. 1. Morphology of the base substrates: a) TEM observation of the BN-SiO_2 ceramic; b)-c) electron diffraction pattern of the phases; d) macrostructure of the SNS.