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Microstructure, residual stress and mechanical properties of Al_2O_3/Nb joints vacuum-brazed with two Ag-based active fillers

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ABSTRACT

Vacuum brazing was applied to join Al_2O_3 ceramic and pure Nb with two active Ag-based fillers, Ag-Cu-Ti and Ag-Cu-Ti + Mo. The influences of the brazing parameter and the content of Mo particles on the microstructure and mechanical properties of the brazed joint were studied. The typical interfacial structure of Al_2O_3/Nb joint brazed with Ag-Cu-Ti filler metal at 900 °C for 10 min was $Al_2O_3/Ti_3(Cu,Al)_3O/TiCu + Ti_2Cu_3 + TiCu_4 + Ag(s,s) + Cu(s,s)$/Nb. In the joints brazed with Mo particle-modified composite filler, the large blocky Ti-Cu compounds disappeared, and a large number of fine phases formed in the brazing seam. The dissolved Mo played a better role in refining the microstructure compared with residual Mo particles. In addition, the finite element method (FEM) calculated results showed that the residual stress peak value of Al_2O_3/Nb brazed joint decreased from 296 MPa to 253 MPa when 8 wt. % Mo particles were added in the composite filler. The shear strength of Al_2O_3/Nb brazed joints could reach up to 203 MPa at 900 °C for 10 min. The joining properties of the Al_2O_3/Nb brazed joint were primarily dependent on the dispersion degree of fine Ti-Cu compounds and the thickness of $Ti_3(Cu,Al)_3O$ reaction layer.

1. Introduction

Al_2O_3 ceramic, characterized as having superior mechanical properties as well as excellent resistance to corrosion and wear, is widely applied in electronic, aerospace and automotive industries [1–3]. However, like most other ceramic substrate, the inherent rigidity and brittleness associated with the Al_2O_3 ceramic limits its structural application, especially in the manufacturing of complex structures [4,5]. Therefore, the joining of Al_2O_3 ceramic to itself or to metals is commonly used to fabricate components with large size and complex shape for engineering applications. As a refractory metal, niobium has good plasticity, corrosion resistance and weldability [6,7]. Components with both ceramic and metal properties can be obtained by the successful joining of Al_2O_3 and Nb. For instance, the joining of Al_2O_3 ceramic and Nb is the key technology in micro nuclear reactor engineering [15]. This fully illustrates that it is necessary to join Al_2O_3 ceramic and Nb.

At present, ceramic-metal joints could be fabricated by different bonding methods, such as diffusion bonding, adhesive bonding and brazing [8–10]. Among the various joining technologies, vacuum brazing is a versatile and effective technique to realize the bonding of dissimilar materials [1–3,8]. There are many kinds of elements in the joint, which are easily formed oxide. In vacuum condition, the oxidation film can be restrained effectively and the joining properties can be improved. In terms of the special chemical characteristics related to the Al_2O_3 ceramic, the design of the active brazing alloy is essential to realize the joining of two materials. Ti element added in the filler metal can act as a kind of active component to wet the Al_2O_3 ceramic surface by chemical reactions, resulting in a good metallurgical bonding [11,12]. Near-eutectic Ag-Cu alloy with a certain proportion of Ti is the most common filler metal for brazing Al_2O_3 ceramic due to its excellent performance in plastic deformation [3,6,10].

Another important consideration is the mismatch of mechanical properties between Al_2O_3 ceramic and Nb. Detrimental thermal stresses are inevitably produced in the brazed joint as a result of the different mechanical responses of the Al_2O_3 ceramic and metal in the brazing process. If the residual stresses are high enough in the brazed joints, failure can occur even though the external force is comparatively small. Therefore, it is essential to decrease the harmful residual stresses as much as possible to increase the joint strength. Extensive efforts have been made to relieve the residual stresses by the design of the brazing filler [13–16]. The most frequently used solution is to add particles or whiskers with a low coefficient of thermal expansion (CTE) into the brazing filler to fabricate the composite filler metal. He et al. [17] reported that the bending strength of the Si_3N_4/Si_3N_4 joints brazed with Ag-Cu-Ti + 5 vol % SiC filler was 35.7% higher than that of the joints brazed with the filler without SiC. Yang et al. [18] reported that when

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40 vol % TiB whiskers were generated in the TC4/Al_2O_3 joint, the shear strength of brazed joints increased by 50%. These improvements in the joint strength were greatly attributed to the reduction of residual stress. However, only a few works have investigated to enhance the Al_2O_3/Nb brazed joints. Zhu et al. [19] have reported that the addition of Mo net enhanced the thermal shock resistance of Al_2O_3/Nb brazed joint. Wang et al. have reported that the addition of Mo in Ag-Cu-Ti filler could refine the blocky brittle Ti-Cu compounds to improve the shear strength of ZrO_2/Nb brazed joint [6]. At the same time, Mo particles with CTE of 5.1×10^{-6}/°C can reduce the CTE of the composite filler, which can notably decrease the degree of mismatch between the base materials and filler alloys and then relieve the residual stress of the brazed joint. For these two reasons, the composite filler containing Mo particles was adopted to braze Al_2O_3 ceramic and Nb. However, the microstructure of Al_2O_3/Nb joint brazed with Ag-Cu-Ti + Mo composite filler was not provided and the relationship between microstructure, residual stress and joint strength was also not clear.

In this study, Ag-Cu-Ti and Ag-Cu-Ti + Mo composite filler were used to braze Al_2O_3 ceramic and Nb. The influences of the brazing parameter and the content of Mo particles on the microstructure and shear strength of the brazed joint were studied. Additionally, FEM was used to calculate the magnitude and distribution of residual stresses of the brazed joints to reveal the reinforcing mechanism of Mo additive.

2. Experiments and modelling

2.1. Experimental procedures

Al_2O_3 ceramic with a purity > 95% and pure niobium plate (purity > 99%) were selected as the parent materials. The Al_2O_3 ceramic and Nb in this work were produced from Suzhou Maiang New Materials Co., Ltd. and Baoji Titanium Industry Co., Ltd., respectively. The mechanical property of Al_2O_3 ceramic was measured via three point bending test, and average value of the flexural strength was 241 MPa. The size of the Al_2O_3 ceramic sample for brazing was 5 × 5 × 5 mm. Nb sample was sawed into 10 × 15 × 2 mm slices. The surfaces of the sample were hand abraded to 1200 # using SiC sand paper, and then, ultrasonic cleaning with acetone was performed for 10 min before the brazing experiment. Two types of brazing filler, commercial Ag-21Cu-4.5Ti (wt. %) powder and Mo particle-modified composite filler, were used for brazing the Al_2O_3 ceramic and Nb. The composite filler was a mixture of Ag-Cu-Ti powder and Mo particles by milling in the planetary ball mill at a 220 rpm rotation speed for 120 min under argon atmosphere. The average particles diameters of Ag-Cu-Ti and Mo were 10 μm and 1.5 μm, respectively. They were produced from General research institute for nonferrous metals and Beijing Xing Rong Yuan Technology Co., Ltd., respectively.

Before the brazing experiment, the filler paste was smeared onto the brazed surfaces of Al_2O_3 ceramic with a thickness of 100–200 μm. Then, the assembled specimens with the structure of Al_2O_3/filler paste/Nb were put into a vacuum furnace to do the brazing experiments. The vacuum level of the furnace was higher than 7×10^{-4} Pa during the whole brazing process. The suitable temperature range was decided by the melting point of brazing filler. The melting point of Ag-Cu-Ti was about 800 °C and the melting point of composite filler would not change significantly when adding Mo particles [3]. In addition, brazing temperature must be higher than the melting point of brazing filler to ensure well joining. In our previous experiments, the best brazing parameter for ZrO_2/Nb joint was 900 °C for 10 min and the optimal condition for Al_2O_3/Ti_6Al_4V was 880 °C for 10 min [3,6]. So the selection of temperature range was 860–920 °C. Holding time depended on the size of the sample and the reaction between base materials and brazing filler, so the holding time was set to be 5–30 min.

After brazing, the cross-section of the Al_2O_3/Nb brazed joint was observed by a scanning electron microscope (SEM, JSM-7800) equipped with an energy dispersive spectrometer (EDS). The phase analysis was carried out by an X-ray diffractometer (XRD, D8-ADVANCE). The position of X-ray detection originated from the layer by layer stripping of the Al_2O_3/Nb joint until the reaction layer near the Al_2O_3 ceramic was observed. The reaction phase structure was examined by transmission electron microscopy (TEM, JEM-2100F). The foils used in the TEM experiment were derived from the Al_2O_3/Nb joints using Ar ion milling (Gatan 695 PIPS II) to electron transparency. The shear strength of the brazed joints was tested by a universal testing machine (MTS E45). Moreover, the fracture path after shear test was observed with a digital microscope (VHX-2000).

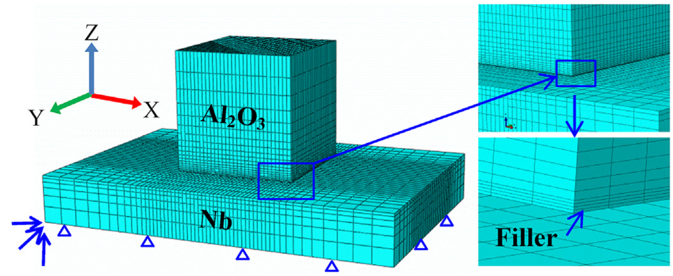

Fig. 1. Typical finite element mesh and boundary conditions of Al_2O_3/Nb brazed joint.

2.2. Model description

The residual stresses distributed in the Al_2O_3/Nb brazed joints were calculated by FEM using Abaqus 6.14 software. As seen in Fig. 1, a three-dimensional model was created, whose dimensions were consistent with the brazing assembly. The model was meshed with hexahedral reduced brick elements with eight nodes (C3D8R). A non-uniform meshing method was used, and the mesh size close to the interlayer was refined, as can be seen in the local magnified figure. The displacements of the nodes on the bottom of the Nb substrate were rigidly constrained in the Z direction, and one of the corners at the bottom was fixed in three directions.

The property parameters [15,20,21] of the materials used in the computation are listed in Table 1. In order to improve the computational accuracy, the variations of these parameters caused by temperatures were considered during the FE stress calculation. The linear elastic model was selected for the Al_2O_3 ceramic, and the elastoplastic model was used for the Nb substrate as well as the brazing fillers. For the joints brazed with the Ag-Cu-Ti + Mo composite filler, the elastic modulus and CTE of the composite filler were calculated by formulas (1) and (2), respectively [22,23]. The results are listed in Table 1.

$$E_c = E_m \left(1 - \frac{3\,V_r(E_r - E_m)}{(E_r - E_m)(1 + 2\,V_r) + 3\,E_m}\,V_r\right)^{-1} \tag{1}$$

$$\alpha_c = \alpha_m(1 - \sum V_r) + \sum \alpha_r V_r \tag{2}$$

Where E and α represent the elastic modulus and the CTE of the brazing seam, respectively; Subscripts c, r and m represent the composite filler, reinforced particle and matrix, respectively; V_r is the volume fraction of reinforced particle.

3. Results and discussion

3.1. Microstructure of Al_2O_3/Ag-Cu-Ti/Nb brazed joints

Fig. 2 shows the typical interfacial microstructure of Al_2O_3/Nb joints brazed with the Ag-Cu-Ti filler at 900 °C for 10 min. A sound Al_2O_3/Nb brazed joint was obtained without any defects or discontinuity, such as micro-voids and cracks. The brazing seam with a thickness of 40 μm formed between the Al_2O_3 ceramic and Nb. As can be seen from Fig. 2(a), two characteristic regions could be distinguished