ARTICLE IN PRESS

Ceramics International xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Ceramics International

journal homepage: www.elsevier.com/locate/ceramint

Interfacial microstructure and mechanical properties of porous-Si_3N_4 ceramic and TiAl alloy joints vacuum brazed with AgCu filler

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ARTICLE INFO

Keywords:
Porous Si_3N_4 ceramic
TiAl alloy
Brazing
Interfacial microstructure
Mechanical property

ABSTRACT

Porous Si_3N_4 ceramic was firstly joined to TiAl alloy using an AgCu filler alloy. The effects of brazing temperature and holding time on the interfacial microstructure and mechanical properties of porous-Si_3N_4/AgCu/TiAl joints were studied. The typical interfacial microstructure of joints brazed at 880 °C for 15 min was TiAl/$AlCu_2Ti$/Ag-Cu eutectic/penetration layer (Ti_5Si_3+TiN, Si_3N_4, Ag (s, s), Cu (s, s))/porous-Si_3N_4. The penetration layer was formed firstly in the brazing process. With increasing brazing temperature and time, the thickness of the penetration layer increased. A large amount of element Ti was consumed in the penetration layer which suppressed the formation and growth of other intermetallic compounds. The penetration layer led the fracture to propagate in the porous Si_3N_4 ceramic substrate. The maximum shear strength was ~13.56 MPa.

1. Introduction

Ceramic-metal components have a wide application prospect. Among all the ceramics, silicon nitride (Si_3N_4) is regarded as one of the most promising ceramics due to its superior properties, such as high-temperature strength, good oxidation resistance, thermal-chemical corrosion resistance, and good dielectric property, etc. [1–3], and pore design of Si_3N_4 ceramic is demanded to decrease its density and dielectric constant in practical applications [4–6]. The porous Si_3N_4 ceramic possesses some beneficial properties in comparison to dense Si_3N_4, such as high specific surface area and good thermal shock resistance [7–9]. Therefore, porous-Si_3N_4 ceramics are considered as the next generation of radome materials. Meanwhile, TiAl intermetallics are well-known as promising alternative to titanium alloy in aerospace and automobile industry since they are featured with low density and high specific strength at elevated temperatures [10–13]. Hence, the porous Si_3N_4 ceramic-TiAl alloy component will be applied in different application widely, such as radome and antenna windows.

It is reported that ceramic-metal joints can be fabricated by transient liquid phase bonding, diffusion bonding, and active metal brazing techniques [14–16]. Brazing is one of the most important joining methods in the ceramic-metal manufacturing technologies. Recently, researches focused on the brazing of dense Si_3N_4 to metals such as stainless steel, titanium alloys, and TiAl alloys [17–20]. AgCu based filler [21–25], Cu based filler [26–29] and Ni based filler alloy [30,31] were selected to braze Si_3N_4 ceramic to metals. Of the various active brazing alloys, Ag-Cu based alloys are the most common filler for their good wettability on Si_3N_4 ceramic and high bonding strength of the joints. As an active element, Ti is characterized as having sufficient thermodynamic driving force to destabilize the ionic or covalent bonding of the Si_3N_4 by reacting with it to form reaction layers. The formation of TiN and Ti_5Si_3 layers on the Si_3N_4 interface was reported [22,32]. It was also found that Ti not only obtained from the brazing filler, but also from the dissolution of the Ti-based metal substrate [33]. Furthermore, AgCu based filler alloys were also used in brazing TiAl alloy to non-metallic materials, such as, ZrO_2 ceramic [34,35], Al_2O_3 ceramic [36,37], SiC ceramic [38], C/SiC composites [39] and C/C composite [40]. However, the physicochemical properties of porous Si_3N_4 ceramic are quite different from the dense Si_3N_4 ceramic, such as low flexure strength, different coefficient of thermal expansion, thermal conductivity, etc. [41,42]. Consequently, the interfacial microstructure and joining strength of the joint brazed by porous Si_3N_4 ceramic and TiAl alloy will be definitely distinguishing.

In this work, porous Si_3N_4 was firstly joined to TiAl alloy using the traditional AgCu eutectic brazing alloy. The effect of porous structure and brazing parameters were studied in comparison with our previous

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http://dx.doi.org/10.1016/j.ceramint.2017.04.149
Received 20 March 2017; Received in revised form 20 April 2017; Accepted 26 April 2017

Please cite this article as: Zhao, Y., Ceramics International (2017), http://dx.doi.org/10.1016/j.ceramint.2017.04.149

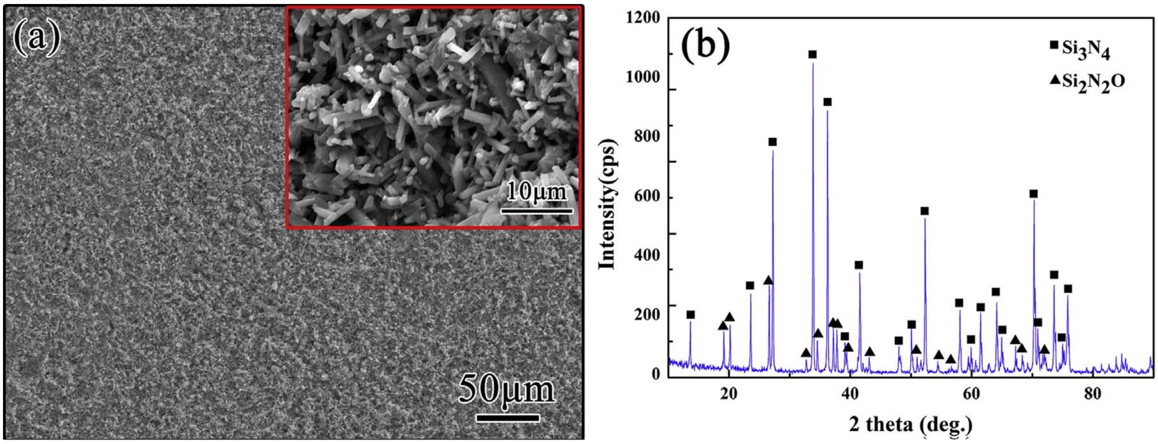

Fig. 1. (a) SEM micrograph of the ceramic; (b) XRD patterns of the ceramic.

work. Especially, the particular penetration layer was formed due to the diffusion and reaction of molten filler into porous ceramic. The influences of brazing parameters on the interfacial microstructure and mechanical properties were analyzed in details.

2. Materials and methods

The porous-Si_3N_4 ceramics with a porosity of 43% used in this work were hot-pressed by β-Si_3N_4 powder. Fig. 1 shows the morphology and the XRD pattern of the porous-Si_3N_4 ceramic substrate. The raw ceramics were cut into blocks with the dimension of 5 mm×5 mm×5 mm using a diamond cutting machine. The dimension of TiAl (Ti-46Al-2Cr-2Nbat%) specimens was 10 mm×5 mm×3 mm. The brazing filler alloy adopted in this study was commercial AgCu eutectic filler alloy (Ag-28Cu) powder within 50 μm.

Brazing procedures were performed in a vacuum furnace with a vacuum of $1.3–2.0\times10^{-3}$ Pa. The brazing surfaces of the porous-Si_3N_4 ceramic and TiAl samples were ground on SiC grit papers prior to joining and then polished by diamond pastes. All of the polished samples were ultrasonically cleaned in acetone and dried by air blowing. The filler alloy powder for 100 μm thickness was placed between the brazing couples. The schematic illustration of the brazed joint is shown in Fig. 2. For vacuum brazing experiments with brazing temperature between 840 °C and 920 °C, holding time ranged from 0 min to 20 min. A 10 min dwell period was incorporated into the brazing cycle on heating to 750 °C, to help reduce any temperature gradients across the components. The joining process was as the following: first heated to 750 °C at a rate of 20 °C/min, and then, the temperature continued to increase to the brazing temperature at a rate of 10 °C/min. Subsequently, the brazing specimens were cooled down to 400 °C at a rate of 5 °C/min. During the brazing process, a pressure of about 0.05 MPa was applied to ensure the surface of couples contact closely.

The cross-sections of joints were characterized by a field emission scanning electron microscope (FESEM, MERLIN Compact, ZEISS) equipped with an energy dispersive spectrometer (EDS, OCTANE PLUS, EDAX) to monitor the homogeneity of the interfacial morphology and chemistry across the joint. Samples for SEM analysis were mounted in an acrylic polymer at room temperature, polished using standard metallographic techniques and finally coated with a thin layer of Au. The reaction phases at the interface were identified using an X-ray diffraction (XRD, DX-2700) spectrometer equipped with Cu Kα radiation. The room temperature shear tests were performed at a constant speed of 0.5 mm/min using a universal testing machine (Instron 5967). For each set of experimental data, at least five samples were used to average to joint strength. The optical digital microscope (DSX 510, OLMPUS) was used to observe the fracture surface of the joint after shear test.

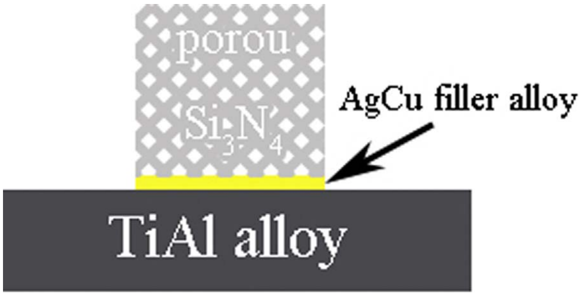

Fig. 2. Schematic illustration of the porous-Si_3N_4/AgCu/TiAl brazed joint.

3. Results and discussions

3.1. Microstructures characterization of the porous-Si_3N_4/AgCu/TiAl joints

Fig. 3 shows the typical microstructure and elemental distribution of the porous-Si_3N_4/AgCu/TiAl joint brazed at 880 °C/15 min. As shown in Fig. 3(a), the brazed joint can be divided into three zones: Zone I (reaction zone adjacent to TiAl substrate), Zone II (brazing seam) and Zone III (penetration layer). According to the element distribution results shown in Fig. 3(b-f), Zone I was a reaction layer adjacent to TiAl substrate which mainly contained of elements Ti, Al, Cu; and element Ag, Cu distributed in the Zone II in form of typical Ag-Cu eutectic structure; in addition to the porous Si_3N_4 ceramic substrate Zone III mainly composed of element Ag, Cu, and Ti. It should be noted that element Ti in Zone III was gradient distribution in the direction perpendicular to the porous Si_3N_4 ceramic.

In order to investigate the microstructure of the joints in details, high magnified interfacial microstructures of Zone I-III are shown in Fig. 4, and the EDS chemical compositions of each spot in Fig. 4 are listed in Table 1. The dark gray phase in Zone I was a strip, distributing with the directionality, and primarily consisted of Al, Cu, and Ti, which could be considered as $AlCu_2Ti$ intermetallics [43]. Occasionally, small quantities of AlCuTi were found between TiAl and $AlCu_2Ti$ layer which was marked as A in Fig. 4(a). According to the Al-Cu-Ti ternary alloy phase diagram and the reaction schemes shown in Fig. 5 [44], the peritectic reaction P_2:L+TiAl+Ti_3Al→AlCuTi happened for the low content of Cu, and the $AlCu_2Ti$ layer was formed by the reaction e_2:L→$AlCu_2Ti$+(Cu). The white phase marked as C and light gray phase marked as D in Fig. 4(b) were Ag based solid solution and Cu based solid solution respectively. Meanwhile, the reaction between Ti dissolved from the TiAl substrate and porous Si_3N_4 ceramic was not only carried out on the ceramic surface but also in the porous structures.