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Microstructure evolution and mechanical properties of ZrO_2/TiAl joints vacuum brazed by Ag–Cu filler metal

Jicai Feng, Xiangyu Dai, Dong Wang, Rui Li, Jian Cao*

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

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ABSTRACT

Reliable brazing of ZrO_2 ceramic and TiAl alloy was achieved using inactive AgCu filler metal. The effect of brazing temperature on the interfacial microstructure and mechanical properties of ZrO_2/TiAl joints were investigated. The results indicated that the representative microstructure of the joint was TiAl alloy/γ-TiAl/AlCuTi/$AlCu_2Ti$/Ag(s,s)+$AlCu_2Ti$+$AlCu_4$/Cu_3Ti_3O+TiO/ZrO_2 ceramic. With increasing brazing temperature, the thickness of reaction zone adjacent to TiAl substrate and continuous reaction layers next to ZrO_2 ceramic increased obviously. In addition, the $AlCu_2Ti$ particle phase coarsened and aggregated in brazing seam at higher temperature. The Ti and Al dissolved from TiAl substrate have an enormous effect on microstructure evolution of ZrO_2/TiAl joints and the mechanism is discussed. The maximum average shear strength reached 48.4 MPa when brazed at 880 °C for 10 min.

1. Introduction

ZrO_2 ceramic is one of the most important structural and functional advanced ceramic materials due to their high strength and fracture toughness, as well as high ionic conductor and chemical stability at elevated temperature [1,2]. These properties are beneficial in numerous applications, such as gas turbines, oxygen sensors and solid oxide fuel cells [3,4]. Like most advanced ceramics, ZrO_2 ceramic is difficult to manufacture large and complex shaped components [5]. Approaches to join ZrO_2 to other materials, especially to metals and alloys, can overcome the disadvantages. TiAl alloys are considered as potential replacements for superalloys in aircraft turbines because of their low density, high temperature strength retention and oxidation resistance [6–8]. In order to increase the maximum working temperature of the aircraft turbines components, ZrO_2 ceramic are usually joined to TiAl alloys as thermal barrier. Therefore, the reliable joining process is crucial for the success of the engine industrial application.

Numerous techniques for ceramic-metal joining such as sintering metal powder process, diffusion bonding, transient liquid phase bonding and vacuum brazing have been commonly used [9–13]. Among these methods, vacuum brazing is recorded as the main method for joining ceramics to metals due to its convenience, cost-effectiveness and high-quality process [14]. Comparing with other ceramics such as Al_2O_3 or Si_3N_4, the brazing of ZrO_2 ceramic has not been investigated systemically. Recently, some researches have investigated the vacuum brazing of ZrO_2 to TC4 [15–17] and to stainless steel [18–23]. However, up to now, there has been no report on the brazing of ZrO_2 ceramic to TiAl alloy. The effect of Ti and Al dissolved from TiAl substrate on the microstructure evolution of ZrO_2/TiAl joints has seldom been presented. Scanning electron microscopy (SEM), energy dispersive spectroscopy (EDS), and X-ray diffraction (XRD) have been employed to examine the reaction products at the interfaces of ZrO_2–metal or ZrO_2–ZrO_2 joints. However, the preceding analytical techniques were inadequate for revealing finer details of the interface adjacent to ZrO_2 at greater resolution. In addition, the selection of the suitable filler metals is crucial to successfully braze ZrO_2 ceramic to TiAl alloy. It is necessary to release the residual stresses at the joint caused by the mismatch of thermal expansion coefficients. Among those filler metals used for ceramic-metal joining, commercial AgCu eutectic filler metal is often preferred because it is relatively ductile and therefore able to minimize the residual stresses [5].

Therefore, in this paper, vacuum brazing of ZrO_2 to TiAl using inactive AgCu filler metal was performed. A transmission electron microscope equipped with energy dispersive spectrometer (TEM/EDS) was introduced to accurately analyze the reaction products at the ZrO_2/brazing seam interface. The effect of brazing temperature on the microstructure and mechanical properties of the joints

* Corresponding author. Fax: +86 451 86418146.
E-mail address: cao_jian@hit.edu.cn (J. Cao).

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were investigated in detail. Furthermore, the mechanism of Ti and Al dissolved from TiAl substrate affecting the microstructure evolution and mechanical properties of the joints were discussed.

2. Materials and experimental procedures

Yttria-stabilized ZrO_2 (YSZ) ceramics containing 3 mol% yttria was provided by Shanghai Unite Technology Co. Ltd. Shanghai, China. The porosity, density and three points bending strength of ZrO_2 were approximately 0.45%, 5.95 g/cm^3 and 780 MPa, respectively. A novel TiAl intermetallic with nominal composition of Ti–52Al–1.6V–1.2Cr–0.1Gd (at%) was used in this experiment. The Ag–28Cu (wt%) filler metal foil with the melting temperature of 780 °C was 100 μm in thickness.

The ZrO_2 ceramic was cut into block with the dimension of 5 mm × 5 mm × 5 mm by a diamond cutting machine. The dimensions of TiAl specimens for metallographic observation and strength test were 8 mm × 8 mm × 3 mm and 20 mm × 8 mm × 3 mm, respectively. Prior to joining, all the joined surfaces were polished and then ultrasonically cleaned by acetone. The Ag–Cu foil was placed between ZrO_2 and TiAl specimens to form a sandwich type and a slight pressure was applied to ensure close surface contact by the graphite blocks. The brazing experiments were carried out at 860–940 °C for 5–30 min in a vacuum furnace with the vacuum level up to $(2–5) \times 10^{-3}$ Pa.

The interfacial microstructure of the brazed joints was characterized by scanning electron microscope (SEM, Quanta 200FEG) equipped with energy dispersive spectrometer (EDS). Meanwhile, the microstructure of reaction products at the ZrO_2/brazing seam interface was observed by a transmission electron microscopy (TEM, Tecnai G^2 F30) equipped with EDS. The crystal structures of reaction products were characterized by selected area diffraction pattern (SADP) of TEM and EDS. The thinning process of specimens for TEM was performed by a combination of conventional hand-polishing and focused ion beam (FIB, Helios NanoLab 600i FIB/SEM). Five shear strength samples were carried out by a universal tensile strength testing machine (Instron 1186) at room temperature, as shown in Fig. 1. The crosshead speed was at a constant speed of 0.5 mm/min. The fracture surfaces of the joints were also investigated by SEM and optical microscope (VHX-1000E). In order to identify the interfacial phases accurately, the fracture surfaces were analyzed by using an X-ray diffraction spectrometer equipped with Cu-Kα radiation (XRD, D8-ADVANCE).

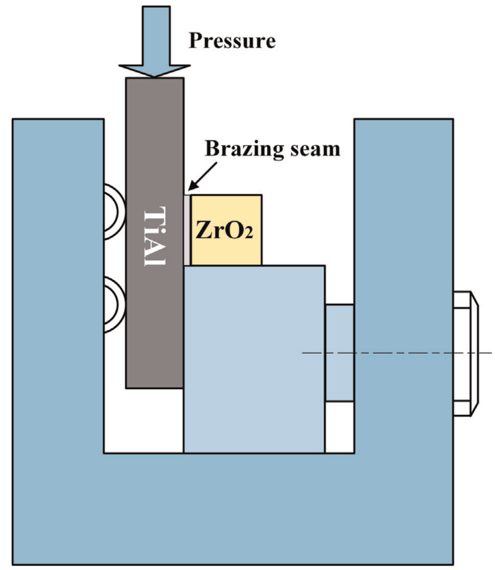

Fig. 1. Schematic description of shear test configuration.

3. Results and discussion

3.1. Microstructure of ZrO_2/AgCu/TiAl joint

Fig. 2 shows the microstructure and corresponding elemental distribution of ZrO_2/TiAl joint at 880 °C for 10 min. It could be seen that the joint exhibited a sound bonding and was devoid of any defects such as cracks and voids. The joint was mainly composed of three characteristic zones: zone I (reaction zone adjacent to TiAl substrate), zone II (brazing seam) and zone III (continuous reaction layers next to ZrO_2 ceramic). Fig. 2(b) illustrated that the Ag was the less active element and mainly distributed at the center of brazing seam. The dissolution of TiAl substrate resulted in the enhancement of Ti and Al in brazing seam. As shown in Fig. 2(c)–(e), the gray granular phase mainly consisted of Cu, Ti and Al. That was because Ti and Al had a strong tendency to extremely react with Cu to form intermetallic compound. Besides, continuous Ti layer and Cu layer adjacent to the ZrO_2 ceramic were shown in Fig. 2(c) and (d), revealing that the Ti and Cu segregated at the interface of brazing seam/ZrO_2 ceramic. It could be indicated that the active Ti dissolved from TiAl substrate played an important role in brazing of ZrO_2/TiAl joints.

In order to investigate the microstructure characteristic of the joints in detail, the magnified microstructures of zone I–III are shown in Fig. 3. The EDS chemical compositions of each spot in Fig. 3 are listed in Table 1. Fig. 3(a) shows the interface of TiAl substrate and brazing seam. Intensive interaction including dissolution, reaction and inter-diffusion occurred at the interface during brazing, which resulted in the formation of three reaction layers. According to the EDS results and Refs. [24,25], the three reaction layers were γ-TiAl phase (spot A), AlCuTi intermetallic (spot B) and $AlCu_2Ti$ intermetallic (spot C), respectively. The Ti and Al dissolved from TiAl substrate diffused into the molten filler metal, and then reacted with Cu to form $AlCu_2Ti$ phase. With the further inter-diffusion at the interface, the AlCuTi phase appeared between $AlCu_2Ti$ and TiAl substrate [25].

Fig. 3(b) presents the magnified micrograph of gray granular phase in brazing seam and the corresponding line scans is shown in Fig. 4. The white phase across the scanning line enriched in Ag, which was determined to be Ag(s, s) phase. The gray granular phase mainly consisted of Al, Cu and Ti. Based on element stoichiometric ratio and related Refs. [8,26], the gray granular phase was $AlCu_2Ti$ phase. The formation of $AlCu_2Ti$ resulted in the consumption of Cu, and then Ag–Cu eutectic microstructure was transformed into Ag(s, s) phase. The light gray phase in the center of the scanning line, which contained Cu, Al and without Ti, could be determined to be $AlCu_4$ phase. The $AlCu_4$ phase formed in the vicinity of $AlCu_2Ti$ phase and mainly followed the reaction path U_1: L+(Cu)→$AlCu_2Ti$+$AlCu_4$ in terms of Al–Cu–Ti ternary alloy phase diagram [27]. Fig. 5 presents the micro-focused XRD pattern taken from the fracture surface of ZrO_2 side and TiAl side. It could be indicated that the phases including TiO, Cu_3Ti_3O, $AlCu_2Ti$ and Ag(s,s) were detected, which confirmed the correctness of above analysis on the interfacial microstructure and phases.

Two continuous reaction layers with a total thickness of 3–4 μm were formed at the interface of ZrO_2 ceramic and brazing seam, as shown in Fig. 3(c). For layer 1 adjacent to ZrO_2 ceramic, a black band with 0.5–1 μm thickness was mainly composed of Ti and O. While for layer 2 next to brazing seam, the gray layer mainly consisted of Cu, Ti, O and Al. In order to further accurately analyze the reaction mechanism between active Ti and ZrO_2 ceramic, TEM observations was illustrated in Fig. 6. It could be seen that two fine-grain reaction layers were formed adjacent to