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# Wetting of AgCu-Ti filler on porous $Si_3N_4$ ceramic and brazing of the ceramic to TiAl alloy

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ABSTRACT

The effect of Ti content on the wettability of AgCu-Ti filler on porous $Si_3N_4$ ceramic was studied by the sessile drop method. AgCu-2 wt% Ti filler alloy showed a minimum contact angle of 14.6° on porous $Si_3N_4$ ceramic during the isothermal wetting process. The mechanism of AgCu-Ti filler wetting on porous $Si_3N_4$ ceramic is clarified in this paper. Porous $Si_3N_4$ ceramic was brazed to TiAl alloy using AgCu-xTi (x = 0, 2 wt%, 4 wt%, 6 wt %, 8 wt%) filler alloy at 880 °C for 10 min. The effect of Ti content on the interfacial microstructure and mechanical properties of porous-$Si_3N_4$/AgCu-xTi/TiAl joints are studied. The typical interfacial microstructure of p-$Si_3N_4$/AgCu-Ti/TiAl joint is p-$Si_3N_4$/penetration layer (Ag(s,s) + $Si_3N_4$ + TiN + $Ti_5Si_3$)/Ag(s,s) + Cu(s,s) + TiCu/$AlCu_2Ti$/TiAl. The maximum shearing strength of the brazed joint was 14.17 MPa and fracture that occurred during the shearing test propagated in the porous $Si_3N_4$ ceramic substrate for the formation of the penetration layer.

## 1. Introduction

Ceramic-metal joints are used widely in various applications, such as radome, hot gas filter, vacuum tube, electronic packing, etc. The inherent differences in physical and chemical properties between the ceramic and the metal make it difficult to obtain a satisfactory joint. Among the developed methods of joining ceramics to metals, brazing is a relatively simple and versatile technique [1,2], and good wettability of brazing alloy on both substrates is essential to obtain a fine ceramic-metal joint [3,4]. However, most of the brazing alloys cannot wet on the surface of ceramic without the active element, such as Ti, Zr, Cr, etc [5].

$Si_3N_4$ ceramic is a promising material applied to the extreme environment because of its outstanding mechanical properties [6], and brazing $Si_3N_4$ ceramic to metals is a key technology in the application of $Si_3N_4$ ceramic in complex structures [7–10]. In brazing $Si_3N_4$ ceramic to metals, AgCu based brazing alloys have been widely selected due to its excellent effectiveness and high cost-performance [11–14]. For example, AgCu eutectic alloy was used to braze titanium metalized $Si_3N_4$ to 45 steel and the AgCu eutectic alloy could completely wet on the metalized $Si_3N_4$ [15]. Song et al. invested the effect of the brazing temperature on the interfacial microstructure and joining strength of $Si_3N_4$/AgCu/TiAl brazed joints [10]. In addition, the interfacial microstructure around wetting triple lines in AgCu-Ti/$Si_3N_4$ reactive system was reported by Nomura et al. [16]. It was also revealed that TiN and $Ti_5Si_3$, as the products of the reactions between AgCu-Ti and $Si_3N_4$, were formed at the first stage and the second stage respectively.

In practical applications, porous $Si_3N_4$ ceramics with good dielectric properties and mechanical properties are considered as the next generation of wave-transparent materials in antenna radomes. Porous $Si_3N_4$ ceramic needs to be joined firmly to the TiAl alloy holder in the radome assembly process [17,18]. The porosity and the preparation technology of the porous $Si_3N_4$ ceramic have a significant influence on its flexure strength, dielectric constant and other physical properties, which affect the mechanical properties of the brazed joint ultimately [19–21]. The surface of the porous $Si_3N_4$ ceramic can be regarded as a rough surface that effects the wettability of the brazing filler alloy on it [22]. The wettability of active brazes on the dense and porous AlN ceramics were investigated by Taranets et al. [23]. Results of their study revealed that the AgCu-Ti showed good wettability on the dense ceramic while the contact angles of it on porous samples were about 20–30° higher than that of the dense sample. It can be deduced that the wettability of the brazing filler on the porous ceramic would influence the joining properties of porous $Si_3N_4$ ceramic/TiAl alloy brazed joint. In our previous study, porous $Si_3N_4$ ceramic was first joined to TiAl alloy by AgCu eutectic filler alloy, and the effects of brazing temperature and holding time on the interfacial microstructure and mechanical properties of the joint were studied [24]. Liu et al. had designed AgCu-

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Ti/Cu/AgCu multi-layered filler to braze porous $Si_3N_4$ ceramic and Invar alloy [25]. However, the role of element Ti played in brazing porous $Si_3N_4$ ceramic and metal has not been studied yet.

In this work, active AgCu-Ti brazing alloys with different Ti content were used to join porous $Si_3N_4$ ceramic and TiAl alloy. The effect of Ti content on the wettability of AgCu-Ti filler alloys on porous $Si_3N_4$ ceramic is investigated, and the wetting and spreading processes of AgCuTi filler alloy on the porous $Si_3N_4$ ceramic are analyzed. Based on the research of wettability of AgCu-Ti on porous $Si_3N_4$ ceramic, brazing of porous $Si_3N_4$ ceramic to TiAl alloy was carried out, and the effect of active Ti on the interfacial microstructure and mechanical properties of the joints are also discussed in detail in this paper.

## 2. Materials and experimental procedure

Porous silicon nitride (p-$Si_3N_4$) ceramic with a porosity of 43% was supplied by the Institute of Special Ceramics, Harbin Institute of Technology. TiAl intermetallic with the nominal composition of Ti-46Al-2Cr-2 Nb (at%) was used in this study. The p-$Si_3N_4$ ceramic mainly consist of needle-like β-$Si_3N_4$, and the TiAl alloy mainly consist of lamellar ($\alpha_2+\gamma$) (Fig. S1, supplementary material). All the samples were grounded by SiC grit paper and washed in the ethanol with ultrasonic and finally dry by air blow before the experiments.

The dimension of p-$Si_3N_4$ ceramic used for the wetting experiments was 15 mm × 15 mm × 5 mm. The brazing filler AgCu-xTi (x = 2, 4, 6, 8 wt%) were prepared by ball milling Ag-28Cu (wt%) powder with Ti powder for 8–10 h. The corresponding atoms percent of Ti contents are 3.40, 7.28, 10.73, 14.08 at%, respectively. The mixed brazing filler powders were compacted into a cylinder to complete wetting experiments. The diameter of the brazing filler powder blocks was 3 mm which was squeezed under a cold-press of about 400 MPa. The sessile drop technique was applied to determine the contact angle between p-$Si_3N_4$ and the brazing filler alloy. The p-$Si_3N_4$ ceramic was horizontally placed in a strainless-stress chamber. The Ag-Cu-Ti block was placed on the top of the p-$Si_3N_4$ substrate before wetting experiment. The heating processes of the wetting experiments were same as the brazing process, which maintained a vacuum of 1.3–2.0 × $10^{-3}$ Pa. The samples were firstly heated to 750 °C at a rate of 20 °C/min. A 10 min dwell period was held at 750 °C to help reduce any temperature gradients across the components. And then, the temperature was raised to the 880 °C at a rate of 10 °C/min and held at 880 °C for 3600 s. Finally, the specimens were cooled down to 400 °C at a rate of 5 °C/min. Real time images were photographed by a camera and the contact angle changing was followed as a function of time. The contact angles were calculated by drop-analysis software after the wetting experiment.

The raw p-$Si_3N_4$ ceramic and TiAl alloy used for the brazing experiments were cut into blocks with the size of 5 mm × 5 mm × 5 mm and 10 mm × 5 mm × 3 mm respectively. For the brazing experiment, the filler alloy powder with 100 μm thickness was placed between the brazing couples, and a pressure of about 0.05 MPa was applied to ensure the surface of couples contact closely (Fig. S2, supplementary material). The brazed joints with different brazing fillers were brazed at 880 °C for 15 min.

The cross-section of the wetting samples and the brazed joint were characterized by a field emission scanning electron microscope (FESEM, MERLIN Compact, ZEISS) that equipped with an energy dispersive spectrometer (EDS, OCTANE PLUS, EDAX). Samples for SEM analysis were polished by standard metallographic techniques and coated with a thin layer of Au before observation. The X-ray diffraction (XRD, DX-2700) spectrometer equipped with Cu Kα radiation was adopted to further identify the reaction phases at the interface. The joining strength was measured by a room temperature shearing method which performed by a universal testing machine (Instron 5967) at a constant speed of 0.5 mm/min. At least five samples for each set of experimental data were used to calculate the average joining strength. The schematic of the shearing test was the same as the study of Liu et al. [26].

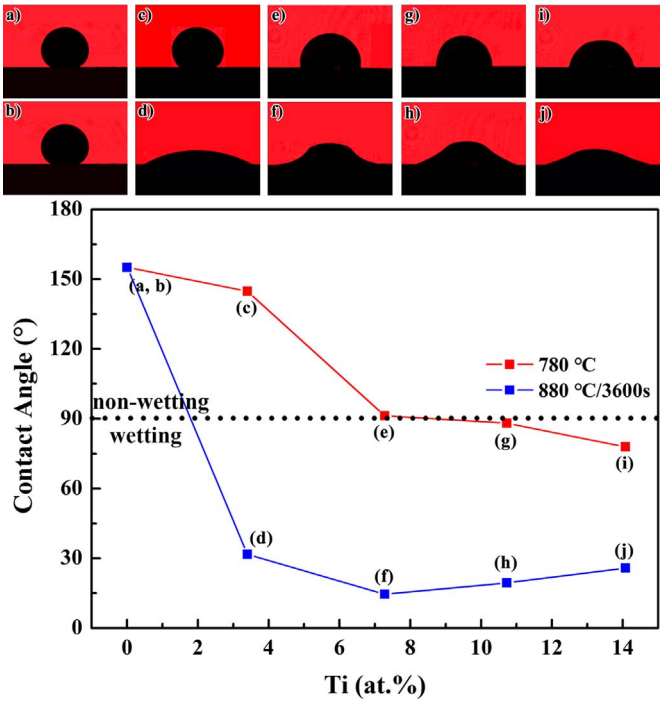


**Fig. 1.** Projective images obtained by camera at beginning (780 °C) and ending (880 °C) of the wetting process. (a-b) 0 at% Ti; (c-d) 3.40 at% Ti; (e-f) 7.28 at% Ti; (g-h) 10.73 at% Ti; (i-j) 14.08 at% Ti.

## 3. Results and discussion

### 3.1. Effect of Ti content on the wetting behavior of AgCu-Ti/p-$Si_3N_4$ system

It is accepted that the wetting and spreading properties of brazing alloy on the ceramic substrate have a critical influence on the brazing process. In order to analyze the effect of Ti on brazing process accurately, the wettability of AgCu-x at% Ti on porous $Si_3N_4$ (p-$Si_3N_4$) ceramic was studied at first. It can be observed that all the filler alloys melted at AgCu eutectic point (780 °C). Thus the moments of temperature arrived at 780 °C and the 3600 s held at 880 °C are considered as the beginning and the ending of the wetting process respectively, and the projective images and the angles of them are shown in Fig. 1. As shown in Fig. 1(a, b), the contact angle between the AgCu eutectic liquid drop and p-$Si_3N_4$ ceramic steadied at 155.1° from the beginning to the end of the wetting process during the isothermal experiment, which indicated that AgCu eutectic filler without Ti element cannot wet the p-$Si_3N_4$ ceramic. Furthermore, it is clear from the line graph in Fig. 1 that the contact angles of beginning and ending of the wetting process decreased with the Ti content of filler alloy increasing. Additionally, it is noteworthy that the curvatures of the liquid drop center are apparently different from that of the liquid drop edges in Fig. 1(f), (h).

The variations of contact angles between the molten filler alloys and p-$Si_3N_4$ during the isothermal wetting process are shown in Fig. 2, which illustrate that the content of active Ti has a great influence on the contact angle. The contact angles of molten droplet with 3.40, 7.28, and 10.73 at% Ti experienced a dramatically decrease during the first 100 s holding at 880 °C. Then the angles gradually declined to certain values and steady around those points in the following time, while the contact angle of molten droplet with 14.08 at% Ti consistently decreased from 86.8° (880 °C/0 s) to 23.3°. Among four kinds of brazing alloys, the droplet with 7.28 at% Ti shows the best wettability for its minimum contact angle 14.6°.

It can be speculated from the changing tendency of four curves that