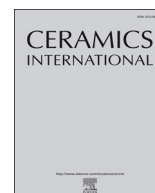




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Interfacial microstructures formation mechanism between SiO₂ substrate and AgCuTi braze alloys

Chenglai Xin^a, Ning Li^b, Junhong Jia^c, Jinsong Du^d, Jiazhen Yan^{b,*}

^a Advanced Research Institute, Chengdu University, Chengdu 610106, PR China

^b School of Manufacturing Science and Engineering, Sichuan University, Chengdu 610065, PR China

^c Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences, Lanzhou 730000, PR China

^d State-owned Assets Supervision and Administration Commission of Sichuan Province, Chengdu 610072, PR China

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ABSTRACT

The wetting characteristics of braze alloys (AgCu eutectic and AgCu eutectic – 2 wt% Ti) on SiO₂ substrate are observed and the results show that small percentage additions of Ti into the AgCu eutectic alloy leads to a dramatic decrease in the steady contact angle. The interfacial microstructures are responsible for the steady contact angle decreasing of different braze alloys on the SiO₂ substrate. The aim of this work is to investigate the interfacial microstructures formation mechanism between SiO₂ substrate and AgCuTi braze alloys. Scanning electron microscopy (SEM) equipped with an energy dispersive X-ray spectroscopy (EDS) and high-resolution transmission electron microscopy (HRTEM) were employed to analyze the interfacial microstructures. The results show that the molten AgCuTi alloy infiltrates into the SiO₂ side, resulting in the formation of SiO₂/Ti₂O/(Ti₄Cu₂O + AgCu)/AgCu/(AgCu + Ti₅Si₃) structures. The Ti atoms in the molten braze alloy firstly accumulated along the surface of SiO₂ and then reacted with SiO₂ to form the Ti₂O layer, at the same time, Si liberated from thermal degradation of SiO₂ dissolved into the molten AgCuTi solution and reacted with Ti to form Ti₅Si₃ compounds. Ti-Cu rich in the molten AgCuTi would react with Ti₂O at the interface to form the Ti₄Cu₂O M6X compounds. Both the experiment results and the theoretical thermodynamics analysis support the proposed viewpoint of interfacial microstructures formation mechanism between the SiO₂ substrate and AgCuTi braze alloys.

1. Introduction

SiO₂ ceramics (SiO₂) present high flexural strength, robust resistance to wear, preferable optical performances, and excellent thermal stability. They have become increasingly more prevalent in aerospace, optics, bioengineering, and electronics industrial fields applications [1,2]. In most cases, SiO₂ ceramics have to be joined to identical materials for practical utilization, however, there are two main issues to resolve during the joining process [3]. The first one is the difference in chemical bonds between ceramics and metals, and the other is the mismatch in the coefficient of thermal expansion (CTE) between ceramics and metals [3]. Advanced welding techniques should be developed to provide an interfacial reaction layer at the ceramic/metal interface, which can create transitional chemical bonds between ceramics and metals, and reduce the interfacial stress as much as possible during the process.

Recently, several welding techniques have been documented [4–11], such as active brazing [4–8], diffusion bonding [9,10], and

partial transient liquid phase (PTLP) brazing [11]. Among these methods, active brazing is a relatively simple and reliable technology for bonding SiO₂ ceramics due to the improved wettability and joining quality by active elements in braze alloys [4]. The work of Liu et al. [2,4] shows that SiO₂ glass ceramics can be brazed to the Ti-6Al-4V alloys with commercially available AgCuTi filler alloys and stronger brazing joints can be realized. The research results of Sun et al. [8] show that SiO₂ can be brazed to Invar alloys by utilizing AgCuTi alloys.

The most commonly used brazing alloys are based on Ag-Cu eutectic with additions of Ti to promote wetting and adhesion. Wetting of AgCuTi alloys on the surface of ceramics is facilitated by the formation of interfacial phases [12,13] such as titanium oxide [13]. It is commonly accepted that wetting and adhesion are achieved by the formation of these interfacial phases [14].

The interfacial phases and microstructures between SiO₂ and AgCuTi alloys have been investigated during the brazing joint of SiO₂ to metals. Liu et al. [4] studied the interfacial microstructures and reaction products during the brazing joint of SiO₂ to Ti-6Al-4V alloys using

* Corresponding author.

E-mail address: yanjiazhen@scu.edu.cn (J. Yan).

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AgCuTi braze alloys and the results showed that the interfacial microstructures are related to the chemical reaction products. Moreover, Liu et al. [2] proved that the $\text{Ti}_5\text{Si}_3 + \text{TiO}_2$ layer and $\text{Cu}_x\text{Ti}_{6-x}\text{O}$ layer formed at $\text{SiO}_2/\text{AgCuTi}$ interface. Previous studies showed that the interfacial phases and microstructures between SiO_2 and AgCuTi play a crucial role in wetting and joining quality. However, little work was done in the past to understand the interfacial microstructures formation mechanism between AgCuTi and SiO_2 substrate. In this reason, the interfacial microstructure formation mechanism between SiO_2 substrate and AgCuTi braze alloys should be comprehensively studied.

In order to eliminate metal dissolution into the AgCuTi alloys, the wetting experiments on SiO_2 ceramics surface with AgCuTi and AgCu eutectic alloys are performed. In present work, the wetting characteristics were observed, and the interfacial interaction between the AgCuTi and SiO_2 was also studied. The interfacial reaction phases were identified by HRTEM to confirm the formation mechanism.

2. Experimental materials and procedure

The base materials are wafer SiO_2 ceramics (diameter 10 mm, thickness 1 mm) with purity of 99%. The braze alloys are commercial Ag-28 wt% Cu eutectic alloy and with an addition of 2 wt% Ti. Prior to wetting experiments, the SiO_2 substrates were cleaned in acetone and placed in an ultrasonic bath for up to ~ 15 min.

Wetting experiments were performed in self-made induction heating furnace. The furnace with stainless steel chamber can reach a ultra vacuum degree of 1×10^{-3} Pa under the effect of the diffusion pump. The furnace chamber with an observation window can allow us to observe the melting of the braze alloys on the SiO_2 surface. Schematic diagram of self-made in-situ observation device is shown in Fig. 1.

Characterization and analysis of the interfacial microstructures formation were made using scanning electron microscopy (SEM, HITACHI S-4800) equipped with an energy dispersive X-ray spectroscopy (EDS) analyzer, and high-resolution transmission electron microscopy (HRTEM, G2 F20 S-TWIN) with selected-area electron diffraction (SAED). The specimen for TEM observation were fabricated by focused ion beam technique (FIB).

3. Results and discussion

3.1. Wetting characteristics

The variation of contact angle θ as a function of time for the wetting of SiO_2 ceramics by braze alloys (AgCu eutectic and AgCu eutectic + 2 wt% Ti) are shown in Fig. 2. The time origin was taken to be the room temperature, as shown in Fig. 2(a). The braze alloys began to melt after a short heating period of 30 s due to a faster heating speed of

induction heating. After melting of the braze alloys, the contact angle θ decreased. The AgCuTi braze alloy can decrease the contact angle θ from 110° to 12° while the AgCu eutectic hardly decrease the contact angle. Careful observation reveals that the decreasing rate of the contact angle θ was high as the melting time up to 75 s, and then became very slow up to 120 s. Finally the contact angle θ kept a steady angle. The wetting process can be divided into two stages. During the first stage, labeled 1, the contact angle θ decreases dramatically; the second stage, labeled 2, the contact angle θ decreases slowly. Finally, the contact angle θ is constant. The variation of contact angle θ as a function of time is similar to other reactive systems such as AgCuTi/ Al_2O_3 system [15–17]. The additions of Ti in the braze alloys is responsible for a lower contact θ on the SiO_2 ceramics surface. More precisely, small percentage additions of Ti leads to a dramatic decrease in the steady contact angle in the CuAgTi/ SiO_2 wetting system, from values close to 110° to $10\text{--}20^\circ$.

The wetting results of braze alloys on SiO_2 surface are shown in Fig. 3. It can be seen that the AgCuTi droplets were found to have broken off SiO_2 substrates during cool-down owing to differential thermal contraction, implying a stronger adhesive strength between AgCuTi and SiO_2 [18] as shown in Fig. 3(a). In the case of AgCu eutectic/ SiO_2 , the bonding is weak [18] as shown in Fig. 3(b). Analyses by profilometry on the wetting system show that the interfacial reactions consist of a slight dissolution of SiO_2 into the AgCuTi alloys other than AgCu eutectic alloys. In other words, the dissolution of SiO_2 was more pronounced in the presence of Ti, and dramatic chemical reactions occurred at the AgCuTi/ SiO_2 interface with an addition of Ti.

The addition of 2 wt% Ti to AgCu eutectic has great influence on wetting and mechanical behaviour. The steady contact angle θ can be greatly decreased and the interfacial bonding strength can be improved. It is significant that the properties of braze alloys on the SiO_2 surface are affected by the addition of titanium. It is greatly related to the interfacial microstructures produced by interfacial chemical reaction. Thus, the interfacial microstructures of AgCuTi/ SiO_2 wetting system should be investigated.

3.2. Interfacial characteristics

Fig. 4 shows the SEM cross-sectional micrographs of interface between AgCuTi and SiO_2 . Also the EDS results from the TEM cross-sectional micrographs of interface are presented in Fig. 5. It can be seen that the interface consists of three reaction layers: an interfacial reaction layer adjacent to SiO_2 ceramic marked as layer A, an interfacial reaction layer adjacent to braze alloy marked as layer B, and a transition layer between layer A and layer B marked as layer C. The EDS results indicate that layer A is composed of Ti, Cu, O and Ag; layer B is composed of Ti, and Si; layer C is composed of Ag, and Cu. These structures indicate that the AgCuTi braze alloys have reacted with the SiO_2 , resulting in the formation of these three reaction layers. Upon further analysis, it is considered that the molten AgCu alloy penetrated into the interface reaction layer and evolved into a transition layer during the solidification process. Moreover, higher quantity of Si was detected in layer B, indicating the diffusion of Si atoms from SiO_2 substrates. Since large amounts of Si were found in reaction layer B, it can be surmised that Ti dissolves O from SiO_2 rather than from low levels of oxygen impurities in furnace vacuum.

In order to clarify the mechanism of interface reactions, Fig. 5 further presents the cross-sectional TEM micrographs of the interface between AgCuTi and SiO_2 . The results show that the thickness of layer A is uneven and has curved edge, which suggests a severe interfacial reaction between SiO_2 and AgCuTi. Nanoparticles identified at ~ 100 nm are also observed in reaction layer A, which are scattered throughout the matrix. These nanoparticles contain Ag and Cu, and are named as such AgCu nanoparticles. Results acquired by electron diffraction patterns (EDP) and Energy Dispersive Spectrometer (EDS) in zone '1' show that the matrix comprises Ti-Cu-O compounds ($\text{Ti}_4\text{Cu}_2\text{O}$, $\text{Ti}_3\text{Cu}_3\text{O}$),

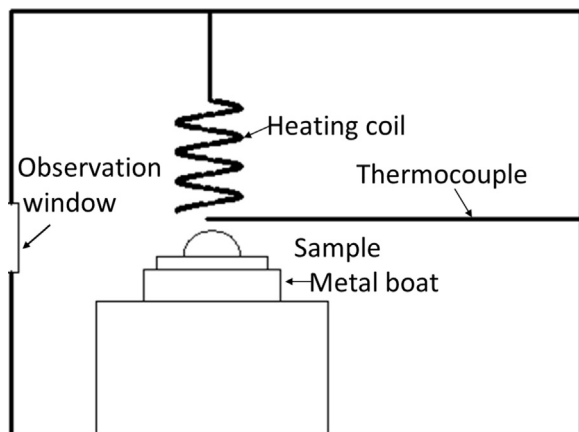


Fig. 1. Schematic diagram of self-made in-situ observation device.

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