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ERADICATING BRITTLENESS OF Ti-6Al-4V/Cu DISSIMILAR BRAZED JOINT USING A Nb DIFFUSION BARRIER

Directly brazing titanium (Ti) alloy and copper (Cu) using silver (Ag) based filler alloy has been recognized to be of limited success owing to the embrittlement of intermetallic compounds (IMCs) presented at joint interface. In the current investigation, strong and reliable brazing of Ti-6Al-4V and Cu was achieved by using a niobium (Nb) diffusion barrier deposited on Ti-6Al-4V base material and Ag-Cu-Ti active braze. The Nb diffusion barrier effectively suppressed the interaction between Ag-Cu-Ti filler and Ti-6Al-4V base material, avoiding formation of brittle Ti-Cu IMCs. Joint consisting of Ti-Nb solid solution, unconsumed Nb interlayer, remnant Ag-based braze and small quantity of tiny Ti-Cu IMC particles was obtained via diffusion bonding at Ti-6Al-4V/Nb interface and active brazing between Nb interlayer and Cu based material. Bonding strength exceeding the Cu base material property was achieved in the resultant joint, ascribed to elimination of continuous bulk brittle interfacial reaction products.

Keywords: Ti-6Al-4V; Cu; Brazing; Interfacial reaction; Intermetallic compounds

1. Introduction

Ti alloys are widely used in various industrial sections such as aerospace, nuclear power plant and chemical industry owing to their excellent corrosion resistance and mechanical properties [1]. In many cases, it is required to combine Ti alloys and other more commonly used metallic materials such as stainless steels, Al alloys, Ni-based super-alloy and Cu, in order to achieve optimized properties and balanced cost [2-14]. For instance, joining of Ti to Cu is anticipated for heat exchanger fabrication specific to nuclear application, where Cu would serve as heat sink material [15]. The joining of Ti alloy and Cu has been long recognized to be challenging because of poor metallurgical compatibility and significant mismatch in physical properties of the Ti-Cu system. There is very limited mutual solubility in Ti-Cu binary system, and a range of IMCs would be expected upon reaction between Ti and Cu. Meanwhile, the great difference in coefficient of thermal expansion of Ti and Cu is prone to result in considerable residual stress in such hybrid components upon welding temperature field. Consequently, conventional fusion welding techniques are infeasible to join Ti and Cu, since the presence of brittle IMCs and notable residual stress tend to result in catastrophic cracking and in turn, unsatisfactory bonding strength of the joint [16].

Brazing is an alternative joining technique by which the base metals are bonded at interface by reacting with molten filler alloy at temperature well below the melting point of base metals. Such a bonding characteristic is highly desirable for Ti/Cu dissimilar joining as the excessive growth of brittle interfacial reaction products can be readily avoided because of the significantly reduced heat input [17]. Previously, attempts have already been made to join Ti and Cu by brazing. Commercially available filler alloys such as Ag-28Cu, Ag-28Cu-2Ti and Ag-5Al were tested to evaluate the feasibility of Ti/Cu dissimilar brazing [18-20]. Indeed, the problems encountered in fusion welding can be circumvented and sound Ti/Cu brazed joints were documented. For instance, defect free Ti/Cu joint with tensile strength of ~70 MPa was obtained by vacuum infrared brazing at 810°C using Ag-28Cu-2Ti filler [20]. However, despite that the problems associated with conventional Ti/Cu welding were alleviated by brazing, the success was limited as brittle Ti-Cu IMCs still dominated the joint. The bonding strength of brazed Ti/Cu joints remains far less than the base metal property. In addition, the bonding strength was highly sensitive with bonding parameters,

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and marginal changes in brazing temperature/soaking time would lead to notable decline in joint strength. As a result, the bonding strength of brazed Ti/Cu joint was generally considered to be of poor reproducibility and reliability [20]. From a practical standpoint, it is of great technological interest to refine this Ti/Cu brazing technique to synergistically improve bonding strength and reliability of Ti/Cu dissimilar joint.

In this investigation, brazing of Ti-6Al-4V to Cu with the aid of a Nb diffusion barrier pre-deposited on faying surface of Ti-6Al-4V was developed. It was demonstrated that the formation of continuous bulk brittle IMCs encountered in conventional Ti/Cu practices was successfully hindered and bonding strength higher than Cu base metal property was obtained. The metallurgical principles guiding the diffusion barrier design and bonding mechanism of resultant joint were elucidated in the experimental results.

2. Materials and methods

Commercially pure Cu and Ti-6Al-4V billets with dimension of $20\times20\times35$ mm^3 were machined from as-received plates as base materials. Active braze Ag-28Cu-2Ti (wt.%) with a thickness of ~70 μm was adopted as filler alloy. The solidus and liquidus temperature of the filler alloy are 780°C and 805°C, respectively. Faying surfaces of base materials and filler alloy were grinded using SiC abrasive paper to #1000 and subsequently ultrasonically cleaned in an acetone bath for 5 min. Depositing of Nb interlayer onto the Ti-6Al-4V was achieved in a multi-arc ion sputtering facility using pure Nb target. The sputtering chamber was initially pumped to a vacuum content of 3×10^{-3} Pa and subsequently filled with argon gas. Following heating the substrate to 400°C, the substrate was etched with Ar ion for 1 hour under bias of –600 V. Deposition was subsequently conducted in a pure Ar atmosphere under pressure of 10 Pa, with target current of 100A, sputtering temperature of 400°C and substrate bias voltage of –400 V. Depositing duration of ~4 hours was taken to fabricate a Nb coating with thickness of ~15 μm.

The bonding assembly schematically illustrated as Fig. 1(a) was installed into a vacuum furnace for brazing. Vacuum condition of 1×10^{-2} Pa was maintained all through the bonding procedure. As illustrated in Fig. 1(b), the bonding assembly was initially heated to 760°C at a heating rate of 20°C/min. A soaking duration of 10 min was intended at this temperature in order to promote diffusion bonding between Ti-6Al-4V and Nb interlayer before melting of the filler alloy. Subsequently, the bonding assembly was further heated to the peak brazing temperature 850°C, and held for 2 min and 5 min for brazing.

The as-bonded joints were sectioned by electro discharge cutting, grinded and polished for interfacial microstructure characterization in JSM 7800F scanning electron microscopy (SEM) equipped with energy disperse spectrum (EDS), under the backscattered electron (BSE) mode. Meanwhile, sub-sized specimens with dimensions schematically illustrated in Fig. 1(c) were machined for tensile tests. Room temperature tensile tests were conducted in an Instron 1342 universal test machine under a crosshead speed of 1 mm/min. Three samples were tested for each experimental set to ensure the reliability of the tensile test results.

3. Results and discussion

The interfacial microstructure of brazed Ti-6Al-4V/Cu joints with Nb diffusion barrier is given in Fig. 2. It can be found that the Nb diffusion barrier remained intact under both brazing conditions. The joints are free from brazing defects such as voids, discontinuity and unbonded regions. In the case of brazed at peak temperature for 2 min (Fig. 2(a)), considerable dissolution of Cu base metal took place, leading to evolution of original near eutectic Ag-Cu structure into Ag-rich and Cu-rich duplex structure in the braze. This indicates that good bonding was achieved

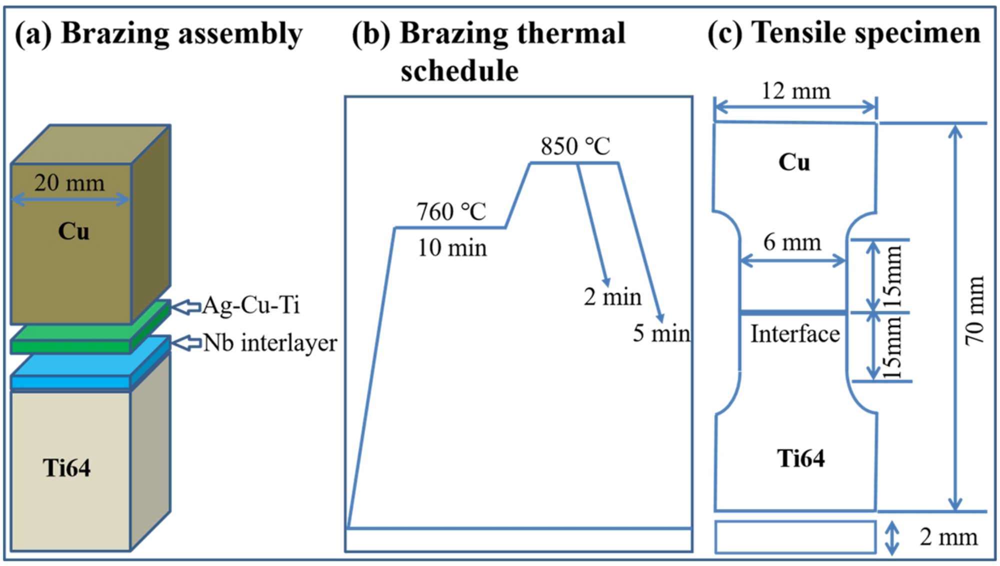

Fig. 1. Schematic illustration of the (a) brazing assembly, (b) brazing thermal schedule and (c) specimen for tensile test

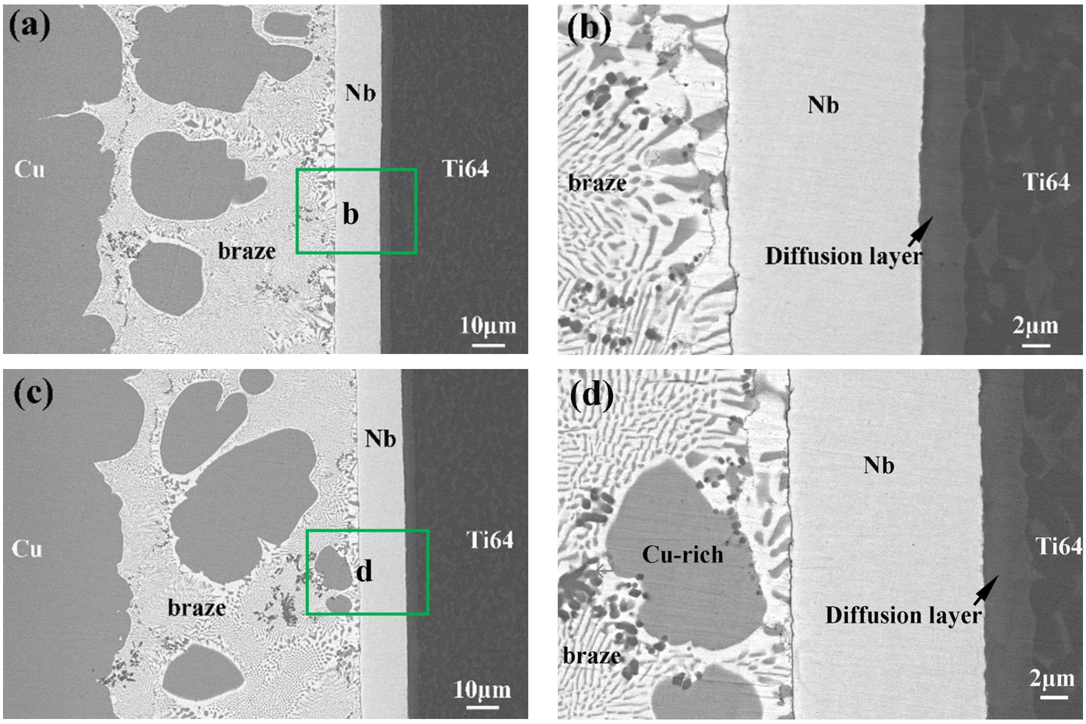

Fig. 2. SEM-BSE images of the Ti64-Nb-Cu joint under brazing duration of (a, b) 2 min and (c, d) 5 min

between the Ag-Cu-Ti filler and Cu base metal. the braze/Nb interlayer/Ti-6Al-4V half was shown in Fig. 2(b). Intimate bonding between the Ag-Cu-Ti braze and Nb interlayer can be observed. Meanwhile, in contrast to the Cu/Ag-Cu-Ti braze interface, the interaction between the braze and Nb interlayer was negligible as the interface is rigid and no additional reaction products can be detected. The majority of the Nb interlayer remains unconsumed and effectively suppressed the interaction between braze and Ti-6Al-4V base material. An additional interdiffusion layer can be observed at the Nb/Ti-6Al-4V interface. In the case of joint brazed for 5 min, the interfacial microstructure, as shown in Fig. 2(c) and Fig. 2(d), is analogous to that of the joint bonded for 2 min to a great extent. The Nb interlayer successfully played the role of diffusion barrier and impeded the interaction between Ti-6Al-4V and braze, leading to a joint consisting of remnant Ag-based filler, unconsumed Nb interlayer and diffusion layer in contact with Ti-6Al-4V base material.

To further reveal microstructural details of the diffusion layer between Nb interlayer and Ti-6Al-4V base material, SEM-BSE images at higher magnification of this region is given in Fig. 3. Corresponding EDS analysis results were plotted in Fig. 3(c) and given in TABLE 1. According to EDS point analysis results collected in TABLE 1, it can be found that in both cases of joints bonded at peak temperature for 2 min and 5 min, the interdiffusion layers at the Nb/Ti-6Al-4V interfaces are primarily highly alloyed with Ti and Nb. EDS linear scanning spectrum across the diffusion layer in joint bonded for 2 min is given in Fig. 3(c). No plateaus were detected in the concentration profiles of all elements, indicating that this diffusion layer was continuous solid solution. Referring to the Ti-Nb-Al ternary alloy phase diagram, no intermediate phase can be read [21]. In addition, it is well known that Nb is a strong beta Ti stabilizing element [22]. It is thus deduced that the diffusion layers are β-(Ti, Nb) solid solution. The above results suggest that solid-state diffusion

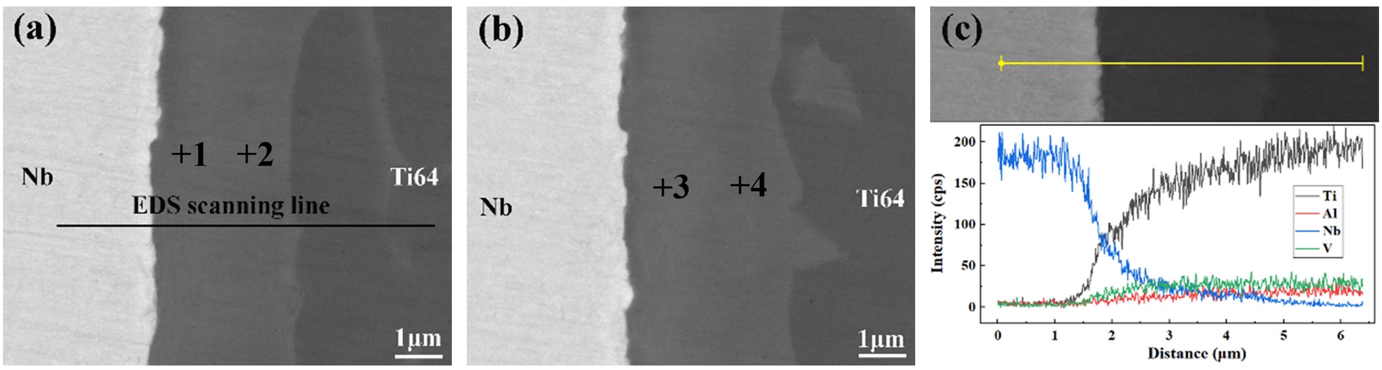

Fig. 3. SEM-BSE images showing the interfacial microstructure of Nb interlayer/Ti64 interface of joint brazed for (a) 2 min, (b) 5 min and (c) EDS linear scanning profile across the 2 min joint interface

bonding occurred between Nb interlayer and Ti-6Al-4V upon the brazing thermal cycle, leading to formation of β-(Ti, Nb) solid solution. Prolonging bonding duration at peak brazing temperature resulted in only marginally increased thickness in β-(Ti, Nb) layer from ~3 μm to ~4 μm.

TABLE 1

Chemical composition (at.%) of marked locations in Fig. 3

	1	2	3	4
Ti	55.6	73.2	50.4	68.0
Al	5.1	6.3	5.7	5.8
Nb	41.3	20.5	43.9	26.2
Possible phase	β-(Ti, Nb)	β-(Ti, Nb)	β-(Ti, Nb)	β-(Ti, Nb)

The SEM-BSE images showing interfacial microstructure of the Ag-Cu-Ti/Nb interface at higher magnification are given in Fig. 4. It can be found that the Ag-Cu-Ti braze exhibited excellent wettability on Nb interlayer in joints bonded under both brazing conditions, as intimate bonding without any defects was achieved. Dispersed particles denoted as "1" and "4" in Fig. 4, can be observed adjacent to the bonding interface. Associated EDS analysis as given in TABLE 2 revealed that these particles were rich in Ti and Cu. It should be mentioned that the analysis results in TABLE 2 is not so precise as the size of the particles are in the range of sub-micro scale, which is beyond the spatial resolution limit of EDS. Accordingly, the possible phase constituent of these particles is only a speculation. Accurately identifying the composition and phase constituent might be accomplished using FIB/TEM techniques. Since the particles are so small in size and their quantity is limited, it is reasonable to expect that they would exhibit negligible influence on bonding strength. EDS mapping of the joint bonded for 2 min are given in Fig. 4 as well. It is evident that no detectable interdiffusion took place between Nb and Ag-Cu filler. However, segregation of Ti at the bonding interface can be observed, indicating that Ti played a crucial role in promoting interfacial metallurgical bonding between Nb interlayer and Ag-Cu-Ti braze. According to related phase diagrams, Cu-Nb and Ag-Nb systems are immiscible in nature. That is the reason why conventional Ag-28Cu (wt.%) can hardly wet the Nb surface and interdiffusion can hardly take place between the Ag-Cu braze and Nb interlayer.

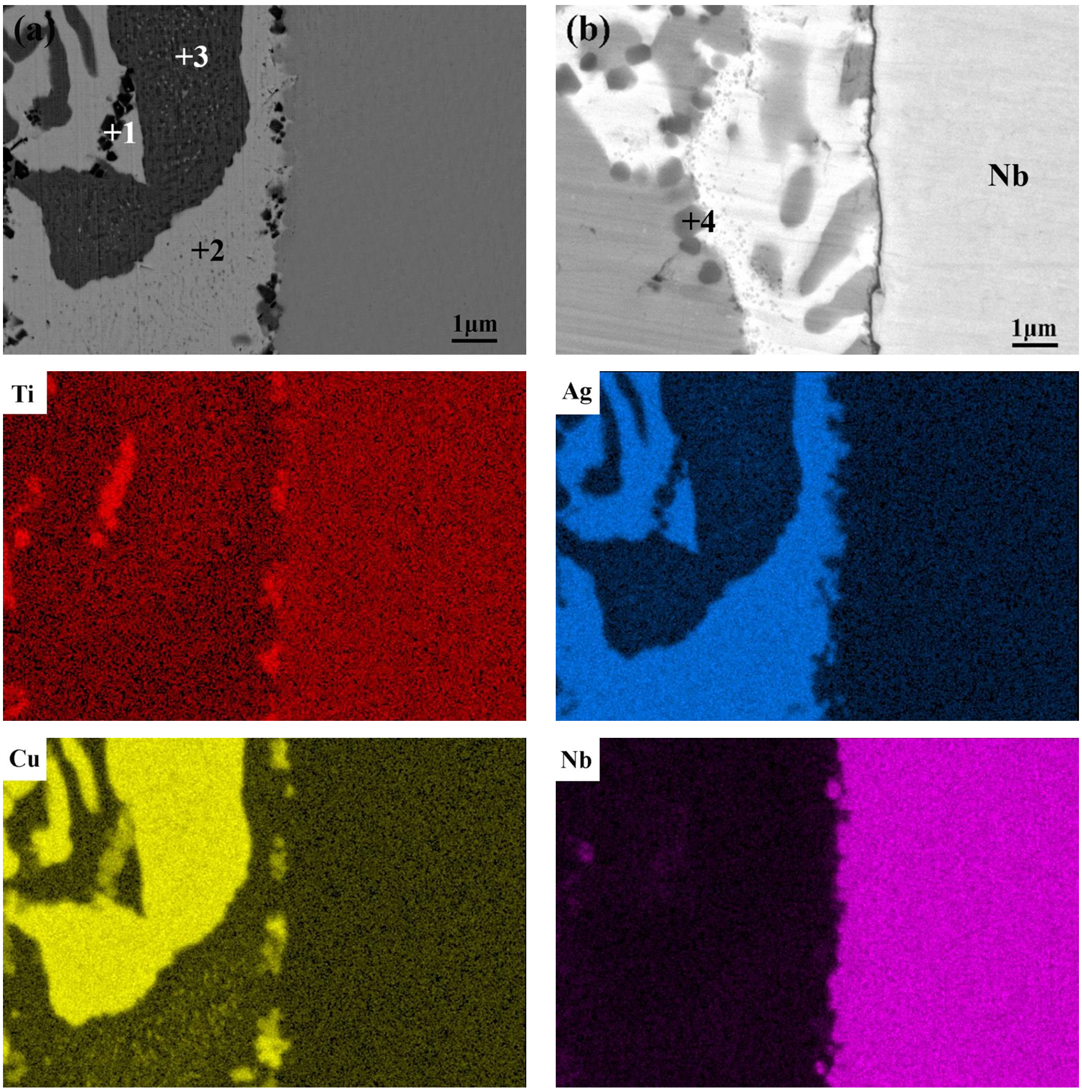

Fig. 4. SEM-BSE images of Ag-Cu-Ti/Nb interface of joints bonded for (a) 2 min and (b) 5 min along with EDS mapping of the 2 min joint

The aggregation of Ti at bonding interface is quite analogous to reactive brazing inert ceramics such as Al_2O_3 using Ag-Cu-Ti active filler alloy. In those cases, bonding was achieved by forming a Ti-Cu-O layer via chemical reaction [23]. In the present case, a Nb-Ti solid solution might be expected as Ti is soluble in Nb.

TABLE 2

Chemical composition (at.%) of marked locations in Fig. 4

	1	2	3	4
Ti	43.7	0.5	2.2	34.3
Cu	52.3	4.4	95.9	54.9
Ag	4.0	95.1	1.9	10.8
Possible phase	Ti_3Cu_4	Ag	Cu	Ti_3Cu_4

The bonding strength of resultant joints were evaluated by room temperature tensile test and the results are given in TABLE 3 and Fig. 5. Both of the joints bonded at peak temperature for 2 min and 5 min exhibited excellent bonding strength up to 220 MPa. Such a bonding strength is notably improved compared with previous investigations where direct brazing was adopted to join Ti and Cu. It was reported that tensile strength of only ~70 MPa can be obtained when direct brazing Ti and Cu using Ag-28Cu-2Ti filler alloy. More importantly, all the joints fractured in the Cu base material, indicating that the bonding strength of the joints were higher than the base material property. Representative tensile curves and photographs of the fractured specimens were shown in Fig. 5. It is apparent that the joints and Ti-6Al-4V base material remained stable during tensile test, whilst the Cu base material accommodated the whole plastic strain till fracture, leading to fully developed stress-strain curves.

TABLE 3

Tensile test results of joint brazed under different conditions

	2 min	5 min
Bonding strength (MPa)	221±4	219±3
Fracture location	Cu base material	Cu base material

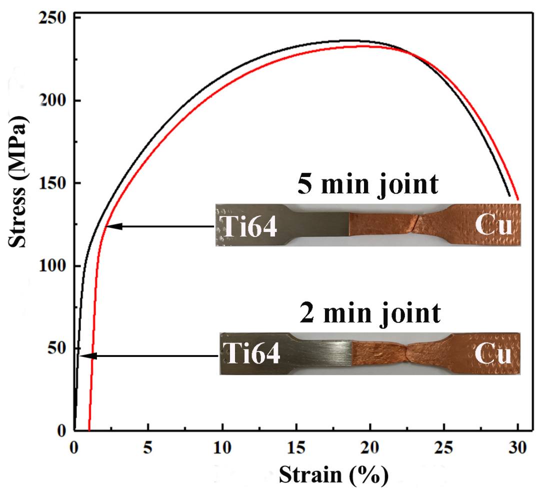

Fig. 5. Representative tensile curves of the joints with fractured specimen inserted

The current investigation aimed at improving bonding strength of Ti alloy/Cu dissimilar joint via eradicating brittle interfacial IMC phases. Previous investigations concerning Ti/Cu dissimilar brazing have already revealed that the formation of detrimental Ti-Cu IMCs was attributed to preferential reaction between Ti and Cu from the filler [18,19]. In this regard, it is essential to suppress the interaction between Ti-based base material and brazing filler. Designing a diffusion barrier which is concurrently compatible with Ti base material and brazing filler is a feasible route for this intention. For instance, Lee et al. developed an Ag diffusion barrier on Ti base material in the case of Ti/Cu dissimilar brazing [20]. Upon infrared brazing under sophisticated thermal conditions, interaction between Ti and brazed was avoided and joint free from Ti-Cu IMCs was obtained. The bonding strength of thus formed joint was superior to strength of Cu substrate. In spite of the success in eliminating brittle IMCs, the main downside of this method is that the Ag diffusion barrier can be readily eroded by molten Ag-Cu filler. Consequently, very stringently controlled brazing parameters are required to keep the Ag diffusion barrier effective in isolating Ti base metal and brazing filler. It is thus of great technological interest to develop an alternative diffusion barrier which is more resistant against erosion from molten braze.

Refractory metals including W, Mo, Nb and Ta can be considered as an appealing candidate for this purpose since they are highly soluble with Ti, meanwhile they would not form IMCs when combined with Ag and Cu, in this way the formation of brittle IMC can be circumvented [21]. Moreover, they exhibit melting point exceeding 2000°C, and negligible solubility in Ag and Cu. It is thus expected that they would meet the requirement of being resistant to erosion by Ag-Cu filler alloy as well. Among these metals, W and Mo exhibit poor fracture toughness at ambient temperature, thus Nb is preferred considering its plasticity and relatively lower cost. However, it should be noted that Nb can be hardly wetted by commonly used Ag-28Cu (wt.%) filler alloy, because of the immiscibility [23]. To resolve this problem, an active Ag-28Cu-2Ti was employed to facilitate interfacial wetting and metallurgical bonding between braze and Nb interlayer, since excellent bonding strength was documented in brazing Nb, Mo and W using Ag-Cu-Ti active brazing filler alloy [23,24]. It is expected that the combination of Nb diffusion barrier and Ag-Cu-Ti active filler would yield IMC free Ti-6Al-4V/Cu joint with high bonding strength.

The experimental results are in good agreement with the above predictions. Nb interlayer successfully impeded the interaction between Ti-6Al-4V base material and Ag-Cu-Ti filler, avoiding the formation of undesirable IMCs. Upon brazing thermal cycle, joint primarily consisting of β-(Ti, Nb) solid solution, unconsumed Nb interlayer, remaining Ag-rich braze and small quantity of tiny Ti-Cu IMC particles was obtained. The bonding mechanisms can be summarized as solid-state diffusion bonding in the Ti-6Al-4V/Nb half and active brazing in the Nb/Ag-Cu-Ti/Cu half. Owing to the absence of continuous bulk IMC layers at joint interface and excellent interfacial bonding, the brittleness of the joint was successfully eradicated and bonding strength exceeding the Cu base material properties was achieved. Additionally, as the solid-state diffusion between the Nb interlayer and Ti-6Al-4V is rather slow, and the Nb interlayer is highly resistant to erosion from molten braze because of negligible mutual solubility of Nb-(Ag, Cu) system, the Nb interlayer would remain effective as a diffusion barrier over a wide range of brazing parameters. Thus, desirable joint microstructure can be obtained without stringent brazing thermal condition control required. This is highly appealing in comparison with previous practices of direct brazing Ti to Cu and Ti/Cu dissimilar brazing using Ag diffusion barrier, as strong and reliable bonds can be obtained under flexible brazing conditions.

4. Conclusion

(1) Ti-6Al-4V/Cu dissimilar brazed joint free from bulk intermetallic compounds continuously distributing along joint interface can be obtained by employing a Nb diffusion barrier and Ag-28Cu-2Ti active filler. The resultant joint was composed of β-(Ti, Nb) solid solution, unconsumed Nb interlayer, remaining Ag-rich braze and discrete tiny Ti-Cu IMC particles .

(2) Excellent bonding strength exceeding the Cu base material property can be obtained in the Ti-6Al-4V/Cu brazed joint with Nb interlayer, ascribed to the absence of continuous bulk brittle IMC phases and excellent interfacial bonding.

(3) The bonding mechanisms of the joining method developed here can be summarized as brazing in the Cu/Ag-28Cu-2Ti/Nb interlayer half and solid-state diffusion bonding between Ti-6Al-4V and Nb interlayer.

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