EFFECT OF COOLING RATE ON TITANIUM AND STAINLESS STEEL JOINTS PERFORMED WITH COPPER INTERLAYER

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Abstract

Microstructure and mechanical response of joints of titanium and AISI 304 stainless steel performed using copper foil as an interlayer was evaluated in the study. The process was carried out in vacuum in the temperature range of 850 to 900 °C for 60 min. The effect of temperature and cooling rate after processing stage on the microstructure and mechanical properties of the joints were analyzed by means of optical and scanning electron microscopy (SEM), electron probe microanalyses, microhardness measurements and tensile and shear strength tests. The maximum values of tensile and shear strengths were obtained for the joints processed at 900 °C for 60 min. The cooling rate after bonding stage had significant influence on the microstructure and composition of the copper/titanium interface that could contain the eutectoid mixture of α-Ti+$CuTi_2$ or the undercooled β-Ti solid solution. The difference in mechanical properties of joints slowly and rapidly cooled reached up to 25 MPa and increased with lowering joining temperature. Observation of fracture surfaces of the joints slowly and rapidly cooled demonstrated that failure took place through the eutectoid mixture or stainless steel/copper interface, respectively.

Keywords: Titanium, stainless steel, copper interlayer, microstructure, properties

1. INTRODUCTION

Titanium has good corrosion and erosion resistance and very high specific strength. Unfortunately it is a fairly expensive metal. Therefore it led to an interest in joining titanium and its alloys to steel for many applications [1-3]. Since the solubility of iron in alpha titanium at room temperature is very low, welding of titanium and stainless steel is very difficult. During welding there are formed very hard and brittle intermetallic phases FeTi and Fe_2Ti near the interface. Regrettably they prevent the production of technically usable welds [4]. One of the most suited methods to achieve strong joints of titanium to steel is brazing. In order to braze titanium many different pure filler metals can be used and also silver base alloys, titanium base alloys and copper base alloys [5, 6]. Unfortunately, such reactive metal as titanium, reacts easily with liquid filler materials and forms intermetallic phases that are located as continuous layers on braze boundaries [1]. Very useful method of joining different materials is diffusion bonding that produces solid-state coalescence through the application of pressure at a temperature below the melting point of the joined materials [7, 8]. But also joints produced by direct diffusion bonding between titanium and steel show the formation of brittle FeTi, Fe_2Ti and Fe_2Ti_4O phases in the diffusion interface [2, 3]. It seems that the effective way to obtain strong joints of titanium to steel is diffusion bonding with an appropriate filler metal. Copper, nickel and aluminum are good candidates. They have a low price and their melting points are lower with respect to joined metals. Transient liquid phase (TLP) bonding is an attractive alternative for joining and repair of similar and dissimilar materials [9, 10]. It mixes the merits of liquid phase joining and diffusion bonding processes. In the TLP bonding the temperature must be lower than the solidus temperatures of the bonded materials and higher than the liquidus temperature of the filler metal. The interlayer metal melts and rapidly attains equilibrium with the solid materials through the process of melt-back dissolution of the substrates. As a consequence of interdiffusion of alloying elements between the base materials and the liquid, the melting point of the interlayer liquid at the liquid-solid interface increases resulting in solidification. If sufficient time for complete solidification is not allowed it can lead to formation of intermetallics and eutectic mixtures occurring along the joint centerline that may be hurtful for joint's properties [9]. In principle, three solid solutions: α-Ti, β-Ti, the solid solution of titanium in copper, and six intermetallic phases, can be produced during the reaction between Ti and Cu [6]. If temperature is high enough for rapid diffusion, copper atoms can migrate in titanium lattice with hcp or bcc structures below and above β-transus at 882 °C, respectively. The diffusion of copper into the titanium substrate lowers the eutectoid transformation temperature since copper is a strong β-Ti stabilizer element [10]. As a result α-Ti + β-Ti aggregates can form due to the decomposition of β-Ti during cooling. Thus the final microstructure and mechanical properties of titanium alloys containing less than 33 at. % of copper depends strongly on cooling rate [11]. Therefore the another benefit of the diffusion bonding using copper relies on the fact that through proper process optimization it is possible to prevent the formation of the injurious eutectoid microconstituent that is brittle and could degrade properties [12]. The objective of this work was to study the influence of the cooling rate on the microstructure and mechanical behavior of diffusion bonded joints of titanium and stainless steel, produced with the use of copper as an interlayer. The results of the investigations are reported and discussed in this paper.

2. EXPERIMENTAL PROCEDURE

Cylindrical Grade 2 titanium and AISI 304 stainless steel rods both having 12 mm diameter were cut into 30 mm long specimens. Chemical compositions and room temperature mechanical properties of base materials are given in **Table 1**.

Table 1 Chemical compositions and mechanical properties of the base materials (accordingly to certificates)

Material	**Chemical elements (wt %)**											
	Fe	**Ti**	**C**	**Cr**	**Ni**	**Mn**	**Si**	**O**	**Mo**	**N**	**H**	**P + S**
Titanium	0.171	bal.	0.024	-	-	-	-	0.142	-	0.008	0.001	-
AISI 304	bal.	-	0.025	18.15	8.05	1.46	0.39	-	0.38	0.063	-	0.05
	Yield strength (MPa)				**UTS (MPa)**				**Elongation (%)**			
Titanium	350				420				38			
AISI 304	230				560				42			

The joining surfaces of the cylinders were prepared by conventional techniques using several stages of grinding papers and polished on 1 µm diamond suspension. The copper foil of 0.1 mm thickness (99.99 % Cu) was used as an intermediate metal. Both surfaces of the foil were polished on diamond suspension and then there were cut circular profiles having 12 mm diameter. The stainless steel cylinders and copper foils were etched in an aqueous 5% solution of HNO_3, while the titanium cylinders in an aqueous 2% solution of HF. All specimens were cleaned ultrasonically in acetone and dried rapidly in air. The joined titanium and stainless steel cylinders with inserted copper interlayer were kept in contact in a steel clamp. After that the samples together with the fixture were placed into a vacuum furnace. The compressive stress of 5 MPa along the longitudinal direction was applied at room temperature using a specially constructed piston installed in the vacuum furnace to obtain good initial contact between titanium, stainless steel and copper. The diffusion bonding was carried out at 850, 875 and 900 °C for 60 minutes in 10^{-3} Pa vacuum. After the joining operation, some samples were furnace-cooled at a cooling rate of 3 $°C{\cdot}min^{-1}$ and other were cooled faster in a tight steel vacuumed receptacle in the air at a cooling rate of 60 $°C{\cdot}min^{-1}$ up to 200 °C. The specimens after diffusion bonding were cut longitudinally, mounted in a cold setting resin, mechanically prepared initially with a grade 1000 abrasive paper and finally using Struers polishing machine and 1 µm diamond suspension. Microstructural observations were performed using a JEOL JMS-5400 scanning electron microscope (SEM) and a Carl Zeiss NEOPHOT 2 optical microscope. Before the samples were examined with the optical microscope they had been etched. The titanium side was etched in an aqueous solution of 88 ml H_2O, 8 ml HNO_3 and 4 ml HF. The stainless steel side was etched by a solution containing 3 g $FeCl_3$, 10 ml HCl, and 90 ml C_2H_5OH. A mixture containing 20 g CrO_3, 75 ml H_2O, and 5 ml HNO_3 was used for etching intermetallics comprising copper. The chemical compositions of the phases were determined in atomic percent using an electron probe microanalyser Oxford Instruments ISIS-300. Composition of the phases was ascertain by comparing the results of the microprobe analysis with the data in the ternary Cu-Ti-Fe phase diagram constructed by van Beck et al. (**Figure 1**).

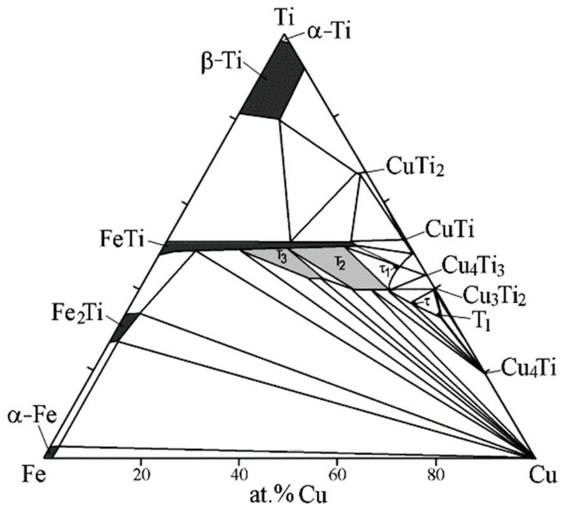

Figure 1 Isothermal cross-section through the ternary phase diagram Cu-Ti-Fe at 850 °C [13]

The microhardness along the cross-section of the diffusion bonded joints was performed by a Hanemann microhardness tester under load of 0.981 N with a testing time of 15 s. Tensile strength and shear strength of the bonded joints were evaluated at room temperature using an INSTRON screw machine at a crosshead speed of 0.5 mm·min^{-1}. The reported mechanical properties of the investigated joints are average values of three specimens that were tested at each processing parameter.

3. RESULTS AND DISCUSSION

Microstructural examinations showed that titanium and stainless steel join through the formation of interface layers between stainless steel/copper on one side and copper/titanium on the other side as a result of the diffusion of metallic elements. The example cross-sections of the joints performed at 875 °C and 900 °C are shown in **Figure 2**.

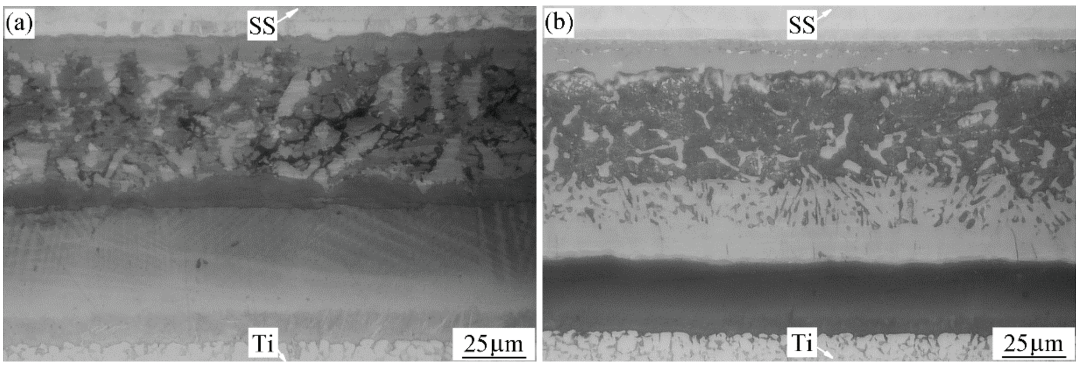

Figure 2 Optical micrographs of the joints prepared at (a) 875 °C and (b) 900 °C for 60 min

According to the chemical analyses and the Cu-Ti-Fe ternary phase diagram [13], it can be assumed that the phases present in the form of layers at the stainless steel/copper interface are FeTi and Fe_2Ti with an amount of Cu, Cr and Ni admixtures. It is worth noting that the Fe_2Ti phase is formed mainly in the joints bonded at temperatures higher than 875 °C. The phases present at the copper/titanium interface are $CuTi_2$, CuTi, Cu_4Ti_3 and Cu_3Ti_2, containing additionally small amounts of Fe, Cr and Ni. The structure of the middle of joints vary significantly depending on bonding temperature (**Figure 1**). Nevertheless, the phases present in the structures are mostly Cu-Ti-based intermetallics and solid solutions based on copper. Microhardness measurements of titanium substrate, interface zones and stainless steel substrate were conducted for all processed samples. The maximum hardness values in the range of 420 to 580 HV were achieved at the stainless steel/copper interface due to the presence of the FeTi and Fe_2Ti intermetallic phases. At the copper/titanium interface and in the middle of the joints hardness values were in the ranges of 280-320 HV and 190-290 HV, respectively. Elrefaey et al. [1], Kundu et al. [12] and Eroglu et al. [14] revealed previously that joints between stainless steel and titanium performed using copper interlayer could not be bonded at temperatures lower than 800 °C, and could be successfully obtained at least at a temperature of 850 °C. However, above mentioned authors did not investigate the effect of cooling rate after bonding stage on the joints microstructure and mechanical properties. Therefore to study the effect of cooling rate, some samples after the bonding stage were cooled at a rate of 3 °C·min^{-1}, while other were cooled 20 times faster at a cooling rate of 60 °C·min^{-1}. The cooling rate did not have any noticeably influence on the microstructure and composition of stainless steel/copper interface in bonded samples. On the other hand, it had significant influence on the microstructure and composition of copper/titanium interfaces. Cross-sections of copper/titanium interface in samples joined at the temperature of 900 °C for 60 min and cooled at two different cooling rates are shown in **Figure 3**.

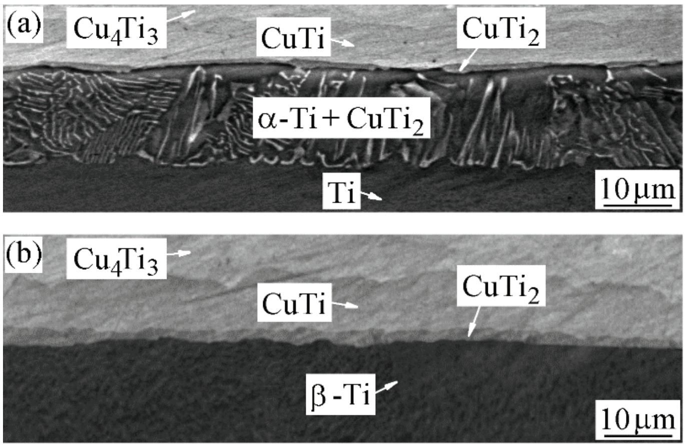

Figure 3 Microstructure of the layers adjacent to titanium cooled at rates: (a) 3 and (b) 60 °C·min^{-1}

The layers between titanium and intermetallics (**Figure 2**) contain between 87.85 and 91.03 at. % Ti and between 6.97 and 9.24 at. % Cu. According to the Cu-Ti phase diagram, Cu-Ti alloys containing between 1.6 and 33 at. % Cu undergo an eutectoid transformation at 790 °C. Therefore the samples furnace-cooled at a cooling rate of 3 °C/min contain the two-phase layer of the eutectoid mixture of α-Ti and $CuTi_2$ (**Figure 3(a)**). Samples cooled at a cooling rate of 60 °C/min contain the undercooled and unbalanced solid solution of copper in beta titanium (**Figure 3(b)**) that is adjacent from one side to the $CuTi_2$ layer and from the other side to α-Ti + β-Ti aggregates formed due to the decomposition of β-Ti. The microstructure and composition of the copper/titanium interfaces influence the mechanical properties of joints processed at temperatures below 900 °C. The difference in mechanical properties is the larger, the lower is the joining temperature. The tensile and shear strengths are lower about 25 and 15 MPa at 850 and 875 °C, respectively, for joints cooled slowly at a cooling rate of 3 °C·min^{-1} in comparison with joints cooled rapidly at a cooling rate of 60 °C·min^{-1} (**Figure 4**). The difference is caused by distinct failure mechanisms.

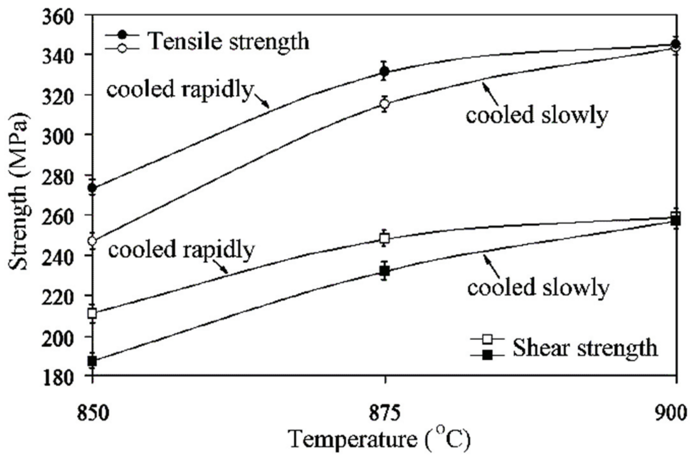

Figure 4 Relation between average tensile and shear strengths of the joints processed for 60 min, type of cooling, and bonding temperature

The fracture paths after performing shear testing for slowly and rapidly cooled samples bonded at 875 °C are shown in **Figure 5**.

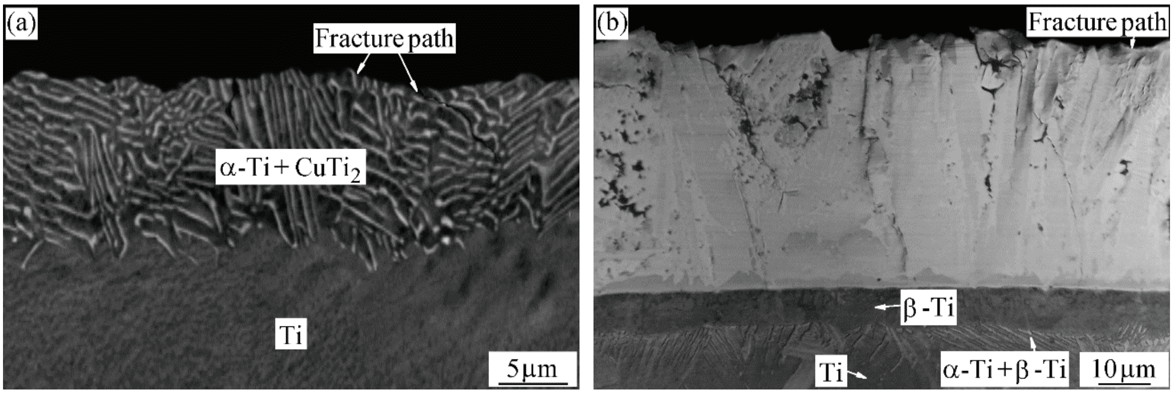

Figure 5 Cross-section of the fracture surface from titanium side of the joints processed at 875 °C for 60 min cooled with two different cooling rates: (a) 3 and (b) 60 °C·min^{-1}

For slowly cooled samples (**Figure 5(a)**), the fracture occurred inside the two-phase eutectoid mixture at the copper/titanium interface. It can be seen that the crack propagated along and crosswise the $CuTi_2$ phase. The brittleness of the eutectoid mixture and the resulting harmful effect on the shear strength of the joints are clearly confirmed by the initiation of several cracks during the shear test. On the other hand, for rapidly cooled samples (**Figure 5(b)**), the fracture took place along the stainless steel/copper interface containing thin layers of the FeTi phase. For all samples joined at the temperature of 900 °C, the fracture occurred at the stainless/copper interface due to the presence of wide layers of brittle FeTi and Fe_2Ti intermetallic phases.

4. CONCLUSION

The main conclusions are as follows:

1) Diffusion bonding of titanium to AISI 304 stainless steel using copper foil as an interlayer can be properly carried out in the temperature range from 850 to 900 °C resulting in joints with good repeatable quality.
2) The microstructure of the joints and thickness of reaction products change significantly with increasing in the processing temperature and time.

3) With a rise in the processing temperature from 850 to 900 °C the tensile and shear strengths of the joints grow. The maximum values of tensile and shear strengths, 343 and 258 MPa, respectively, have been obtained for the joints processed at 900 °C for 60 min.
4) The maximum hardness values in the range of 420 to 580 HV were achieved at the stainless steel/copper interface due to the presence of the FeTi and Fe_2Ti intermetallic phases.
5) The cooling rate after bonding stage has not any noticeably influence on the microstructure and composition of the stainless steel/copper interface, but has significant influence on the microstructure and composition of the copper/titanium interface. When joints are cooled slowly at a rate of 3 $°C \cdot min^{-1}$, they contain the two-phase layer of eutectoid mixture of α-Ti and $CuTi_2$. Joints cooled at a rate of 60 $°C \cdot min^{-1}$ contain the undercooled β-Ti that is adjacent from one side to the $CuTi_2$ layer and from the other side to α-T i+ β-Ti aggregates formed due to the decomposition of β-Ti.
6) The difference in mechanical properties of the joints slowly and rapidly cooled reaches up to 25 MPa and increases with lowering joining temperature. The slowly cooled joints processed at 850 and 875 °C fracture inside the two-phase eutectoid mixture layer at the copper/titanium interface. The rapidly cooled joints containing the β-Ti layer, fracture at the stainless steel/copper interface.

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