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Evaluation of Transient Liquid Phase Bonding Between Titanium and Steel

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Titanium's superior corrosion resistance is ideal for many engineering applications. Process industries choose titanium as the material of construction for piping, tanks, pressure vessels, autoclaves, and heat exchangers. When pressures and/or temperatures and size demand very thick plate, the titanium equipment can become considerably more expensive than units constructed from lower cost, lower performance materials.[1] In such cases, there was considerable interest in joining titanium to low cost materials such as steel alloys. The production of such joint is a complicated task since titanium and steel exhibit poor metallurgical compatibility which promotes the formation of brittle intermetallic phases such as Fe-Ti and Ti-C. In addition to the problem rising from joining titanium to steel, a great care should be taken when welding titanium by conventional fusion welding techniques. The application of these techniques to join titanium usually results in interaction of titanium with elements such as oxygen and nitrogen and efficiency of the joints will reduce.

Brazing is a well-established manufacturing technology for joining a variety of dissimilar materials. Unfortunately, most commercially available braze alloys require high process temperatures which can degrade titanium properties.[2] The use of transient liquid phase (TLP) diffusion bonding, however, offers more promise in maintaining the parent alloy microstructure in the joint region. In the TLP process, a filler metal is inserted between the pieces to be joined. Upon heating, a thin liquid interlayer forms because the melting point of the interlayer has been exceeded or because a reaction with the parent metal results in a low melting liquid alloy. The liquid then fills voids formed by the unevenness of the mating surfaces. After the interlayer region has melted, diffusion in the base metal causes isothermal solidification, resulting in the production of a good joint with the same structure as that of the base metal and no heat-affected zone.[3] Other advantages of TLP diffusion bonding compared with the solid-state diffusion bonding are that lower bonding temperature, pressure and less surface roughness may be required.

It has been reported that Ni, Fe, and Cu, either in the form of pure or alloys, were mainly used as interlayer in the TLP process.[4–7] The use of copper alloy interlayer offers a lower bonding temperature than could be employed for alternative TLP bonding strategies. Thus, the present paper examines microstructural development in copper-interlayer TLP bonds between commercially pure (CP) Ti and low carbon steel. The present paper also characterizes the mechanical properties of the joints in terms of room-temperature shear strength and correlates the bond strength observations with the microstructures observed at the bond line.

Experimental Procedures

CP Ti grade 2 and low carbon steel of 2 mm thickness sheet were used in this study. The nominal chemical compositions of the base metals are given in Table 1. The plates were cut into 30×25 mm^2 chips for shear strength testing and 10×10 mm^2 chips for microstructure analysis. These specimens were then subjected to several stages of grinding papers up to 1000 grit and subsequently ultrasonically cleaned by acetone before brazing. The interlayer foil was a 100 µm Cu-12Mn-2Ni (wt %) foil and its melting range was 970 to 990 °C. The foil was cleaned in acetone before TLP process as well and then sandwiched between the overlapped areas of the base metal.

After adjusting the overlap width to 6 mm (three times the thickness of base metal), the joints were fixed with stainless steel clamp, and then carefully placed into the vacuum furnace. TLP experiments were carried out at 870 and 910 °C to study the effect of the temperature on the metallurgical and mechanical properties of the joint. Experiments were carried out for 90 min at a pressure of 2×10^{-5} Pa. The heating and cooling rate was adjusted at 15 °C min^{-1}.

The cross sections of the bonded titanium/steel joints were prepared for metallographic analysis by standard polishing techniques and then etched by 5% HF, 20% HNO_3, and 75% glycerol solution for 60 s for titanium side and by 3% Nital solution for steel side. The microstructures were investigated by an optical and scanning electron microscopes. The hardness measurement was performed with the help of a Vickers hardness testing machine with 25 s impressing time. Tensile shear specimens were machined from brazed lap

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Table 1. Chemical composition of the base metals.

Alloy	Chemical analyses, at. %									
	C	Fe	Ti	Mn	Si	P	S	N	H	O
CP Ti	0.08	0.03	Bal.	–	–	–	–	0.10	0.11	0.74
Low carbon steel	0.23	Bal.	–	0.17	0.02	0.07	0.02	–	–	–

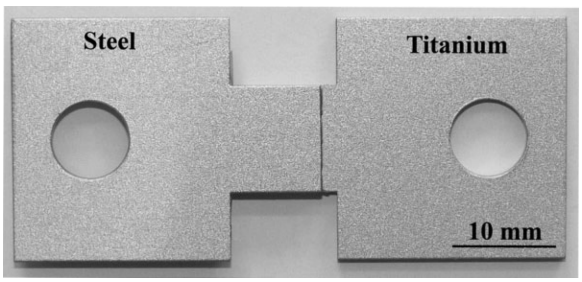
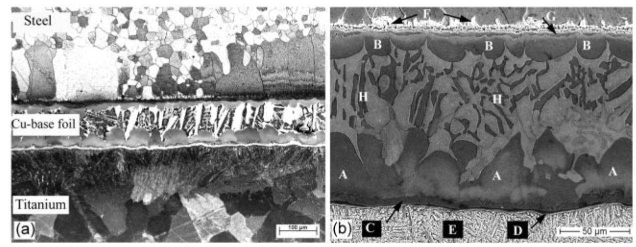


Fig. 1. Tension shear test specimen.

joints, geometry of which is shown in Figure 1.[8] X-ray diffraction study has been done on the fracture surfaces of cp Ti and steel to identify the phases formed on joint fracture surfaces. Cu K_α and Ni were chosen as the X-ray source and filter respectively. The X-ray scan rate was set at 4° min^{-1} in the range between 20° and 150°.



Element at.%	Ti	Fe	Cu	Mn	Ni	Suggested phase
A	37.68	1.55	47.59	10.44	2.74	Ti_3Cu_4
B	34.87	1.79	51.72	9.37	2.25	Ti_3Cu_4
C	48.49	0.20	50.95	-	0.36	CuTi
D	65.25	-	33.94	0.18	0.64	Ti_2Cu
E	90.48	0	7.80	1.20	0.51	α-βTi
F	1.62	87.73	3.38	7.27	-	αFe + (Cu)
G	47.85	31.84	14.05	3.58	2.68	FeTi+Cu
H	20.81	-	70.04	8.11	1.04	Cu_2Ti+Cu_4Ti

Fig. 2. Titanium/steel joint produced at a temperature of 870 °C: (a) optical microstructure, (b) Scanning electron microstructure with point analyses.

Results and Discussion

The characteristic microstructure of the titanium/steel joint produced at a temperature of 870 °C is shown in Figure 2a. It is clear that a sound joint was obtained since a homogeneous microstructure without voids or cracks was observed along the joint. The steel/copper bonding interface is planar in nature and a thin interaction layer was revealed at the interface area. Columnar structure, resulted from diffusion growth accompanied by recrystallization of steel substrate at high temperature, was formed at the steel interface.[9] On the other hand, a thick diffusion layer was observed at the titanium/copper interface.

Observations with SEM revealed that different regions with different chemical analyses were formed in the brazed area, as it is shown in Figure 2b. In the initial stage of the process liquid phase formed between titanium substrate and the Cu-base alloy. According to the Cu-Ti phase diagram shown in Figure 3, the Cu-Ti liquid phase forms within a limited range of composition at slightly lower temperature than 900 °C.[10] Owing to the high percentage of Mn in the Cu-based alloy, it is expected that the Cu-Ti liquid phase forms at quite lower temperatures than suggested by Cu-Ti phase diagram. Titanium continues to diffuse in the liquid

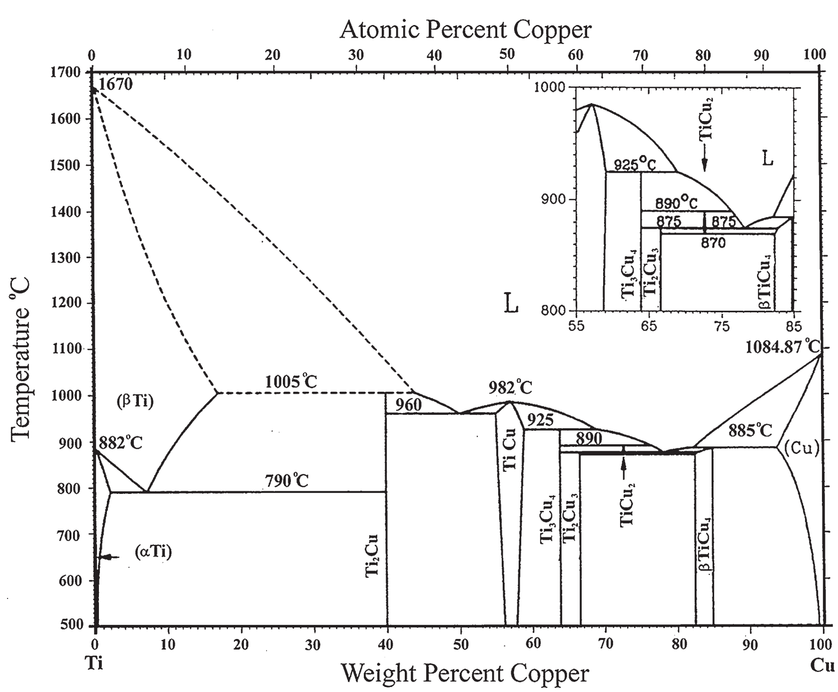


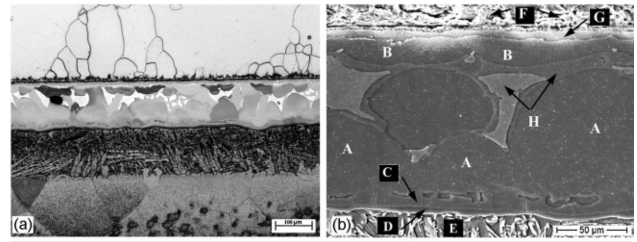
Fig. 3. The Ti-Cu binary phase diagram.

Cu-based alloy and vise versa the low melting point depressant elements diffuse to titanium substrate which causes isothermal solidification areas in the TLP process. These areas are clearly indicated in Figure 2b by symbols A and B. The chemical analyses of these areas showed high percentages of Cu and Ti with significant amount of Mn, Ni, and Fe. The analysis of this phase is closely related to that of Ti_3Cu_4. Area A is in close contact with titanium substrate which reflected to its higher content of Ti in respect to area B.

Three diffusion layers were formed at the titanium/Cu-base alloy. High amount of Ti was diffused from the titanium side to the area close to the titanium substrate as shown from layers C and D. The ratio between Ti and Cu in layers C and D are close to CuTi and Ti_2Cu phases respectively. It is worth noting that the migration of Cu, Fe, and Mn (strong β-stabilizing elements) in the titanium substrate lowers the eutectoid transformation temperature of Ti and α-βTi phase aggregate forms by the decomposition of β-Ti during cooling.[11] The α-βTi phase is shown by region E. On the other hand, the steel/Cu-base alloy showed only two reaction layers. The diffusion of copper into steel substrate produces a solid solution of limited solubility (2.2 wt% at 870 °C) as it is shown from Fe-Cu phase diagram.[10] It is assumed that eutectoid transformation $\gamma \rightarrow \alpha$Fe+(Cu) took place with a high content of αFe in respect to (Cu). The narrow eutectoid structure ($\cong$10 μm in width) is clearly shown in Figure 2b. It is indicated by area F which presented high percentages of Fe in respect to Cu. Additionally, a very thin interaction layer (area G) was formed in between areas B and F. Based on the chemical analysis of this area, it is expected to contain FeTi+Cu intermetallic phase. The Fe-Ti-Cu ternary alloy phase diagram suggests that nearly 38 at % Cu could be dissolved in FeTi.[12] In according to the chemical analysis shown in Figure 2b, area H which is located in the middle of the joint is probably an eutectic mixture of Cu_2Ti and Cu_4Ti with significant percentage of Mn.

The microstructure feature of titanium/steel joint at 910 °C is shown in Figure 4a. All characteristic feature microstructures or phases present are to great extent similar to the joint brazed at 870 °C. The joint brazed at 910 °C showed coarsening of columnar structure in the steel substrate, coarsening of the intermetalic phases at the steel/Cu-base alloy and titanium/Cu-base alloy, and the extension of isothermal solidification areas to consume most of the eutectic structure in the centre of brazed joint. These features are clearly shown in Figure 4b.

Mapping of Ti, Cu, and Fe in the joint is shown in Figure 5. It is obvious that the joint is enriched by Ti content to the level that titanium could reach to the steel substrate at the interface as it is shown in Figure 5a. Increase in the temperature led to an increase in mutual diffusion between titanium substrate and Cu-based alloy which reflect to the coarsening in the isothermal solidification areas. This coarsening consumes most of the joint area and the rest was left with high concentration of Cu as shown in Figure 5b. On the other hand, with reference to the Fe-Cu phase diagram, the solubility of iron in copper is very limited at a temperature of 910 °C.



Element at.%	Ti	Fe	Cu	Mn	Ni	Suggested phase
A	39.93	1.38	53.24	4.98	0.47	Ti_3Cu_4
B	36.54	2.15	49.21	10.16	1.94	Ti_3Cu_4
C	48.34	-	51.66	-	-	CuTi
D	68.31	-	31.21	-	0.48	Ti_2Cu
E	89.45	-	8.23	1.34	0.98	α-βTi
F	1.92	88.29	3.11	6.68	-	αFe + (Cu)
G	47.80	32.04	14.50	3.44	2.22	FeTi+Cu
H	20.63	0.27	72.37	5.76	0.97	Cu_2Ti+Cu_4Ti

Fig. 4. Titanium/steel joint produced at a temperature of 910 °C: (a) optical microstructure, (b) Scanning electron microstructure with point analyses.

Hence, iron showed negligible diffusion in the Cu-base alloy, as it is shown in Figure 5c, and only reacted with titanium at the interface and forms a thin layer of FeTi phase.

The hardness values in the joint brazed at 910 °C are shown in Figure 6. The interaction layers at the steel/Cu-based brazing alloy presented the highest hardness values owing to the formation of FeTi and Ti_3Cu_4 at the interface. These phases constitute the most brittle and harmful structures in the joint. Hardness values of the copper-rich area, in the middle of the joint, and at α-βTi phase showed the lowest level in the joint.

The fracture shear strength was calculated as the failure load divided by the overlap area. The average shear strength of the joint showed a general tendency to decrease with an increase in temperature since the microstructure contained thick intermetallic compounds. This caused an increase in the brittleness and deteriorated the shear strength of the joint performed at high temperatures compared to the joints brazed

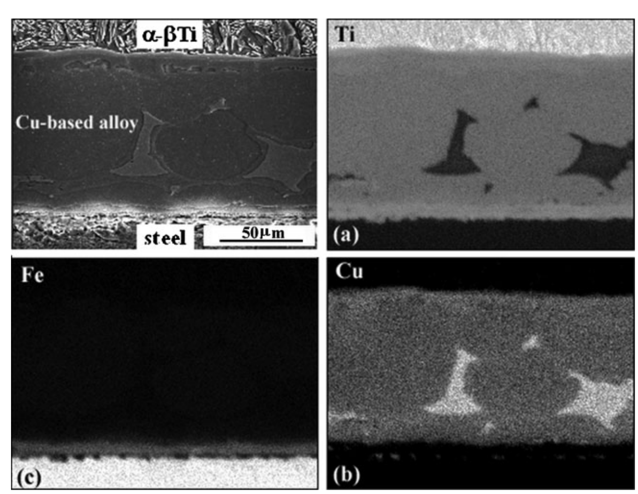


Fig. 5. Mapping of Ti, Cu, and Fe in the joint produced at a temperature of 910 °C.

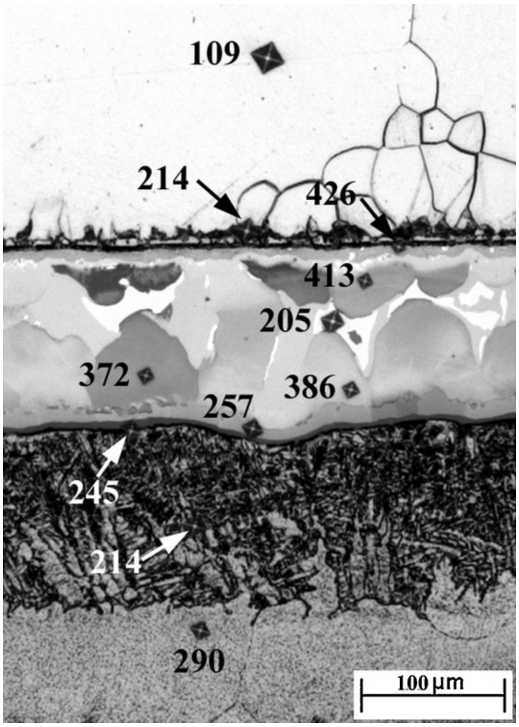


Fig. 6. Hardness distribution at the joint brazed at a temperature of 910 °C.

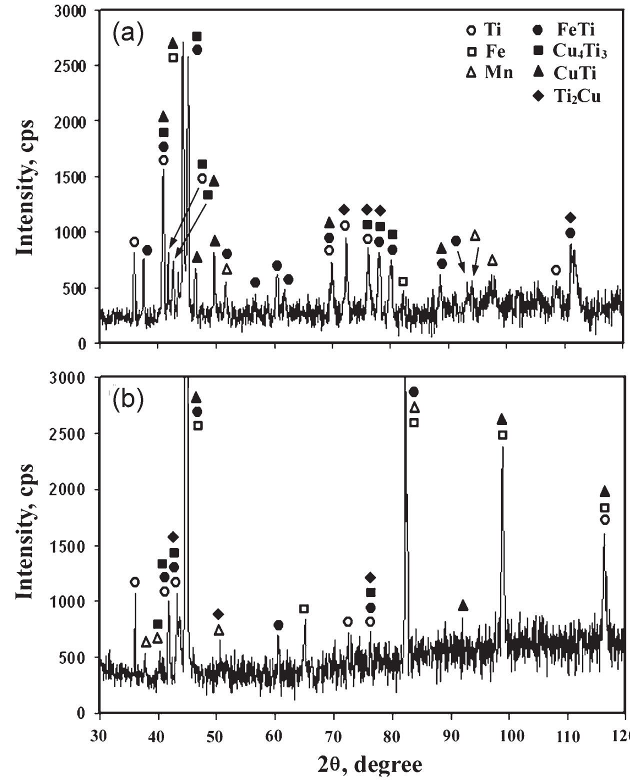


Fig. 8. XRD patterns of the joint brazed at a temperature of 910 °C from the titanium (a) and steel (b) sides.

at low temperatures. The joints bonded at temperature of 910 °C achieved average shear strength of 75 MPa. Meanwhile, the joints bonded at temperature of 870 °C achieved average shear strength of 92 MPa.

The fracture path after performing the shear test is shown in Figures 7a and 7b. It is notable that, at both bonding temperatures, the fracture took place at the interfacial region between the steel substrate and the Cu-based foil. This implies that the FeTi intermetallic compound is the weakest structure. The corresponding microscope fractography of these fracture surfaces, from steel side, are presented in Figures 7c and 7d. The fracture pattern produced is cleavage topography surface in both cases. XRD analysis for the fracture area revealed nearly equal atomic percentages which support the notion that the fracture path follows the FeTi intermetallic layer formed at the interface. This phase is detrimental to the strength of the joint, particularly at high temperature, since thicker and variable types of intermetallics were formed.

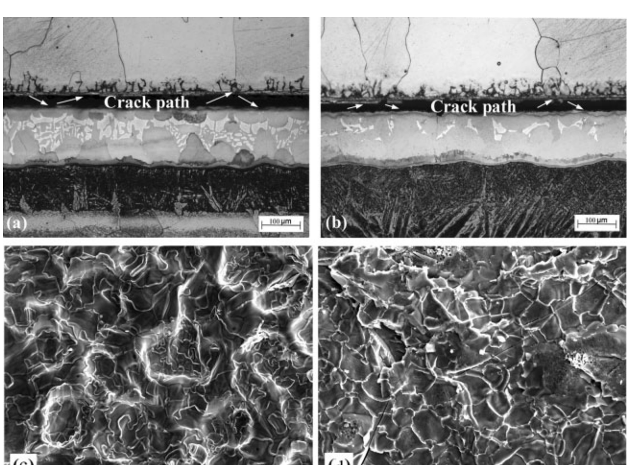


Fig. 7. Cross section of fracture surface and fracture morphology: (a) and (b) Fracture surfaces for the joint brazed at a temperature of 870 and 910 °C respectively; (c) and (d) Fracture morphologies for the joint brazed at a temperature of 870 and 910 °C, respectively.

The intermetallic compounds in the fracture area from titanium and steel sides are identified by X-ray diffraction technique and are given in Figure 8. It has been found that the fracture area of the joints brazed at 910 °C contains FeTi, CuTi, Cu_4Ti_3, and Ti_2Cu. The presence of Fe, Ti, and Mn has also been found in the reaction zone. Meanwhile, Cu has not reflected many peeks and not identified clearly. It can be seen that FeTi and Cu_4Ti_3 are widely observed in titanium and steel fracture surfaces in contrast to CuTi and Ti_2Cu since the fracture originated and propagated at the steel/Cu-based alloy where the FeTi and Cu_4Ti_3 are located.

Conclusion

A copper-based foil containing the melting point depressant manganese has been employed successfully as a TLP brazing interlayer for commercially pure titanium and steel. The following conclusions were drawn from the present study:

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1. The microstructure of the joint consisted of isothermal solidification area at both sides of the joint which coarsened by an increase in TLP bonding temperature. The coarsening structure, identified as Ti_3Cu_4, consumes most of the joint area and the rest was left with an eutectic mixture of Cu_2Ti and Cu_4Ti. FeTi phase was formed at the steel/Cu-based interlayer and constitutes the most harmful structure in the joint. At the same time, Ti_2Cu and CuTi, and α-βTi phases were formed at the titanium/Cu-based interlayer.
2. The average shear strength of the joint showed a general tendency to decrease with an increase in temperature since the microstructure contained thick intermetallic compounds.
3. Microhardness measurements revealed that FeTi presented the highest hardness microstructure in the joints. Meanwhile, the copper-rich area in the middle of the joint produced the lowest one.
4. At all brazing temperatures, the fracture took place at the interfacial region between the steel substrate and the copper-based alloy as a result of the formation of FeTi intermetallic compounds. FeTi and Ti_3Cu_4, which formed at the interfacial region where the fracture took place, were clearly identified by several peeks using X-ray diffraction technique.

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