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THE EVOLUTION CHARACTERISTICS AND NUMERICAL ANALYSIS OF DIFFUSION BONDING INTERFACE STRUCTURE OF TITANIUM ALLOY/Cu/STAINLESS STEEL

Shuying Liu[1,2], Kuan Xu[3], Guangbao Liu[4], Congcong Cao[1] and Zhonghao Heng[1]

[1]Henan University of Science and Technology, School of Materials Science and Engineering, China
[2]Henan Province Key Laboratory of Advanced Non-Ferrous Metals, Henan, Luoyang 471003, China
[3]Suzhou Hailu Heavy Industry Co., Ltd, Jiangsu, Zhangjiagang 215600, China
[4]Henan Technician College of Economics and Business, Department of Electrical and Electronic, Henan, Xinxiang 453700, China

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Abstract. Carries on the investigation to the titanium alloy/Cu/stainless steel intermetallic compound of bonding interface in the meantime, to make a thermodynamic model of the interface element diffusion to have a numerical simulation of the diffusion distance and diffusion temperature, time. Using analysis methods of stretching test, microhardness test, SEM and EDS, to investigate and research the mechanical properties, the interface structure characteristic, the principal element atomic diffusion mechanism of joints thermal simulation and the vacuum diffusion bonding of Ti-6Al-4V/Cu/304, the reacting phases are produced and the distribution range. The results show that when bonding pressure is 5.0 MPa, the joint's tensile strength first increase and then decreases, with bonding temperature and time rising, When bonding temperature is 1223K, bonding time is 3.6 ks, there is a maximum tensile strength that is 162.73 MPa. However, it will is disadvantageous to performance of the joints, when bonding temperature and time extended overly. It formed multi-phase transition organizations by solid solution, intermetallic compounds in the bonding interface, such as Ti_2Cu, Ti_xCu_y, Ti_2Fe, $TiFe_2$ and TiFe. Effect of Ti_xFe_y on strength of the joints is slightly inferior the Ti_xCu_y compound. The fracture is mainly by the titanium alloy side region III for the source dehiscence, developing in the weak diffusion layer.

1. INTRODUCTION

The key for the condition that dissimilar metals structure of titanium/steel extension from aerospace and national defense to the other field of industry is Connect manufacturing technology [1]. Titanium alloy is very expensive. And as for a kind of structural materials of important and special material, it is not mature for the other materials to bond at present. At present, on the study of bonding methods of titanium alloy to stainless steeldiffusion bonding is applied widely in our country and abroad [2-5].

So far, for the study of titanium diffusion bonding with Steel it major study on the analysis of diffusion bonding technology and study of joints performance [6] in recent years. However, the diffusion bonding of titanium alloy/stainless steel is an important core and difficulty on the study and theoretical analysis, as for the affect of joints performance, evolution

Corresponding author: Shuying Liu, e-mail: liusy168@yahoo.com.cn

feature of bonding interface structure and the design and improvement of interlayer [7].

Thermodynamic model that is adapted to diffusion bonding of dissimilar metal materials and reflects diffusion of bonding interface element is established in theoretical analysis. And theoretical analysis is take place between bonding distance and bonding temperature, bonding time. The prediction of element bonding, selecting appropriate diffusion parameters, it can reduce a lot of cumbersome experimental to guide practice.

In fact, titanium alloy and stainless steel inevitable form hard and brittle Ti-Fe intermetallic compounds by diffusion bonding. To avoid base metal element excess bonding and to prevent bonding interface forming the intermetallic compounds that make joints brittle, the results of thermal simulation compare with the results of vacuum diffusion bonding for titanium alloy and stainless steel with the interlayer of Cu. By the study of structure of bonding interface and compound distribution and the way of controlling intermetallic compounds growing, the purpose of issue make the joints develop to toughness and strength. On the other hand, the effect of thermal simulation diffusion bonding is studied to improve efficiency and saving costs.

2. TEST METHOD

Used to test materials are widely used titanium alloy Ti-6Al-4V thick plate, heat-resistant high-strength austenitic stainless steel 304 plate and pure copper foil (thickness are 90 mm, 40 mm), chemical composition of these materials in Table 1.

First, test with titanium and stainless steel cut into size the 24 × 20 × 10 mm the cuboid samples, machining titanium alloy and 304 stainless steel bonding surface and the surface of parallel to bonding surface, ensure that the four surfaces parallel to each other. Second, the titanium alloy and stainless steel vacuum diffusion bonding surface carry out accurate grinding and polishing after to wet the grinding by sand paper, confirmed after the bonding surface can not scrape a mirror as the bonding specimens. also been grinding on the copper foil. Preparation of samples for the thermal simulation can be carried out to accurate grinding. In ethanol cleaning and degreasing before bonding. Diffusion bonding test was carried out in vacuum diffusion bonding equipment, vacuum of 8×10^{-2} Pa. Diffusion bonding test of thermal simulation was carry out by dynamic thermodynamic physical simulation of NC machine (Gleeble 1500D) and inert argon gas environment. Through numerical analysis, select the following more appropriate diffusion bonding technological parameters shown in Table 2.

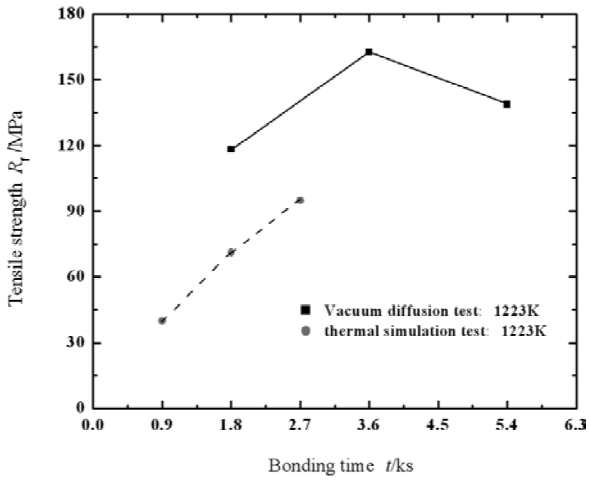

Fig. 1. Relationship between bonding time and tensile strength of Ti-6Al-4V/Cu/304 joints.

3. EXPERIMENTAL RESULTS AND ANALYSIS

3.1. The comparison of joints strength between thermal simulation and vacuum diffusion bonding

Relationship between tensile strength and bonding time is shown in Fig. 1. As the figure shows, under the bonding pressure of 5 MPa, the bonding temperature is 1223K. And as to the diffusion bonding of the thermal simulation, with the increasing thermal simulation time, the joint’s tensile strength continuous increase. The joint’s strength can reach 94.78 MPa in 2.7 ks. However, for the vacuum diffusion bondingwith the bonding time rising, the joint's tensile strength increased firstly and then decreased. The best joint's strength can reach 162.7 MPa in 3.6 ks.

As seen above, no matter thermal simulation or vacuum diffusion bonding, with the extension of bonding time initially, degree of close contacting with the interface improved, and atomic diffusion fully, it forms a continuous alloy layer, so the joint's strength increased gradually. But intermetallic compounds of generation become more thickness when bonding time extends and interface interdiffusion overly. And intermetallic compounds are brittle compounds. At the same time, with growing up of titanium alloy grain seriously, organizing of joints coarsens. Therefore, joint’s strength decreases significantly [9].

Table 1. Chemical composition of experiment materials (wt.%).

Materials	C	Si	S	P	Mn	Ni	Cr	Ti	Fe	Al	V	O	Cu	Impurity
304	0.67	0.54	0.01	0.036	1.10	8.10	17.06	0.20	other					
Ti-6Al-4V	0.03							other	0.10	6.2	4.3	0.15		
pure copper													>99.70	0.3

Table 2. Main bonding conditions.

Materials	Bonding temperature *T*/K	Bonding pressure *R*/MPa	Bonding time *t*/ks	atmosphere/vacuum degree
Thermal imulation	1223	5.0	0.9	argon gas
Ti-6Al-4V/Cu/304	1223	5.0	1.8	
	1223	5.0	2.7	
Vacuum diffusion bonding	1223	5.0	1.8	8×10^{-2}
Ti-6Al-4V/Cu/304	1223	5.0	3.6	
	1223	5.0	5.4	
	1123, 1173, 1273	5.0	1.8	
	1223, 1273	5.0	3.6	

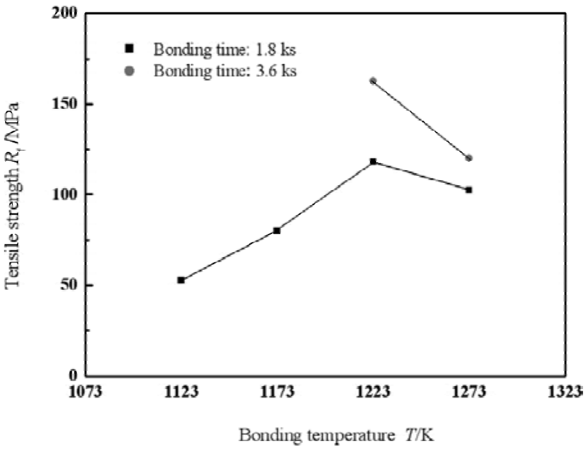

Fig. 2. Relationship between bonding temperature or time and tensile strength of Ti-6Al-4V/Cu/304 joints.

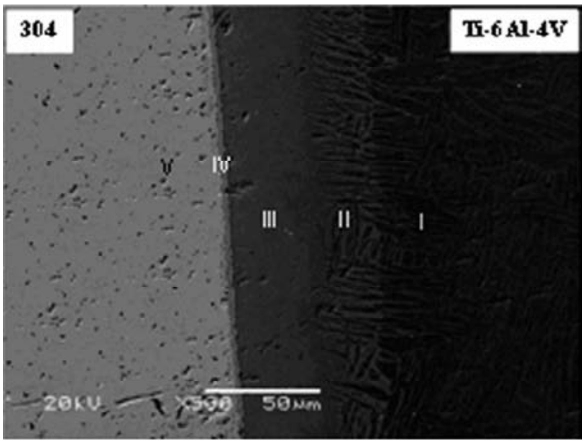

Fig. 3. SEM microstructure measure of Ti-6Al-4V/ Cu/304 bonding interface.

Relationship between bonding time or bonding temperature and joint's tensile strength is shown in Fig. 2. As the figure shows, under the bonding pressure of 5 MPa, the bonding time is 1.8 ks. Initially joint's tensile strength improved with increasing bonding temperature. But when it reaches to some extent, joint's strength decreases with increasing bonding temperature. The above phenomenon is because that activity of atomic near interface increase to improve atomic diffusion process with rising bonding temperature early. Because phase transformation of titanium turn up gradually, so that degree of close contacting with the interface improved obviously and degree of homogenization of connection improved. In the same time, the effective contact area of 304 with Cu also increases. Therefore, the joint's strength improved. But the rising bonding temperature or time overly, it makes the joints strength decrease, too. And when thickness of Cu interlay decreases from 90 to 40 μm, under the bonding time of 2.7 ks, the bonding temperature is 1223K, with reducing of thickness of Cu interlay, the joint's strength improved, but the rate of increase is not obvious.

3.2. Microstructure observation and testing of bonding interface and fracture

Under the bonding pressure of 4.9 MPa, the bonding time is 2.7 ks, and the bonding temperature is 1223K, test results of SEM and EDX of bonding interface is shown in Fig. 3 by thermal simulation. As the figure shows when bonding temperature is 1223K, not only surface of titanium alloy with Cu Copper contact closely by good ductility and deformability, but also 304 with Cu contact closed, too. The width of reaction layer of bonding interface showed bonding layer that it is different light and dark colors and clear. On the whole, interface structure can be divided into five regions, as follow: titanium alloy matrix (I), mesh diffusion zone (II), dark gray area (III), gray zone (IV), 304 matrix (V). Total thickness of diffusion layer (II~IV) is about 63 μm. It is shown that it forms different new phase compound layer which have boundaries significantly and different structure in the bonding interface.

Under the bonding pressure of 4.9 MPa, the bonding time is 2.7 ks, and the bonding temperature is 1223K, microstructure morphology of tensile fracture of thermal simulation joints is shown in Fig. 4. Fracture of stainless steel side and morphology of enlarged is shown in Figs. 4a and 4b, respectively. Fracture of titanium side and morphology of enlarged is shown in Figs. 4c and 4d, respectively. As the figure shows, no matter fracture of stainless steel side or fracture of titanium side is cleavage fracture basically, and both are brittle fracture. It is produced transgranular fracture along a particular crystallographic plane. Some regions take a certain amount of tensile stress in the tensile test. It can be seen from the magnification of the fracture that fracture developed into the direction of the trend of semi-plastic and semi-brittle. That is fracture of brittle fracture at macro level and fracture of semi-plastic and semi-brittle at micro level. It is shown in Figs. 4b and 4d obviously. The only difference is that there is material of titanium side in the bulge of stainless steel side. It is shown in EDX of section 3.4.

3.3. Hardness characteristics near the bonding interface and growth behavior of intermetallic compounds

Under different bonding time, the bonding temperature is 1223K, Vickers hardness for thermal simu-

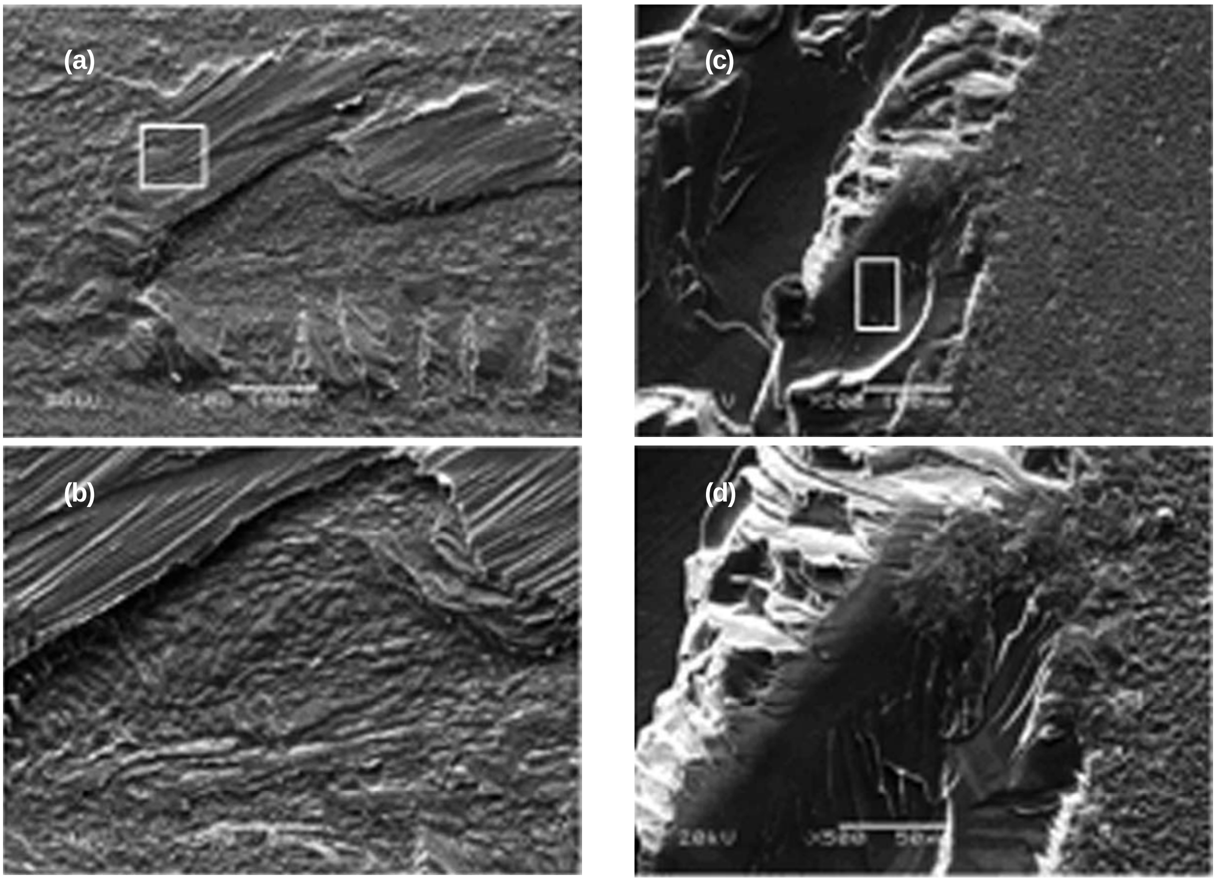

Fig. 4. Microstructure measure of fracture appearance of Ti-6Al-4V/Cu/304 joints. (a) 304 side; (b) 304 side; (c) TC4 side; (d) TC4 side.

lation and vacuum diffusion bonding was tested to further determine distribution of the Ti-Fe and Ti-Cu intermetallic compound series and the effect of joint strength. The results are shown in Fig. 5. As the figure shows, hardness near the bonding interface of welded seam no matter vacuum diffusion bonding or thermal simulation is higher than it both sides of the base metal obviously. At first, hardness will decrease fast. It will slow down until decrease to the same of base metal. Hardness of titanium alloy is higher than hardness of stainless steel obvious. Change rate of hardness is about 170 μm (it is higher than the thickness of SEM slight), and it is higher than stainless steel side (about 80 μm). With the increasing bonding time, hardness near the bonding interface increase on the whole, and distribution range of intermetallic compounds formed is bigger. In the stainless steel side, the hardness of 5.4 ks is not only higher than the hardness of 2.7 ks, but also peak move to the welded seam. In the 304 side, the hardness near the welded seam is higher base metal slight. And beginning from 80-100 μm, hardness close to the base metal along with the distance from welded seam. Those phenomena are shown that bonding interface of 304/Cu side and Cu/Ti-6Al-4V side form brittle and hard intermetallic compounds. It improves joint's hardness obviously [8]. Increase of hardness is reflected in the titanium side mainly. As following, it is explored that effect of Ti_xCu_y and Fe_xTi_y intermetallic compounds on performance of bonding interface.

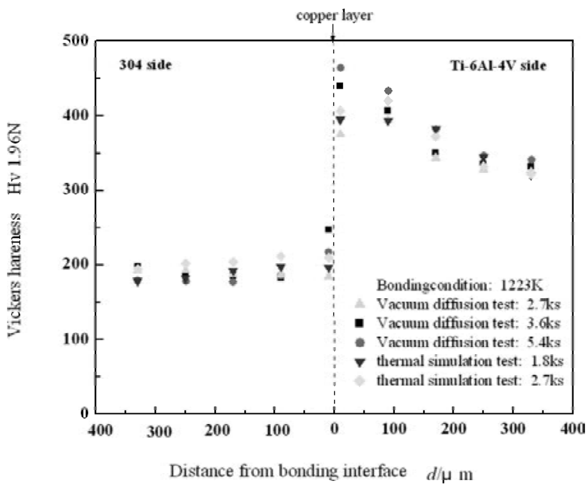

Fig. 5. The change of Vickers hardness along distance from bonding interface of Ti-6Al-4V/Cu/304.

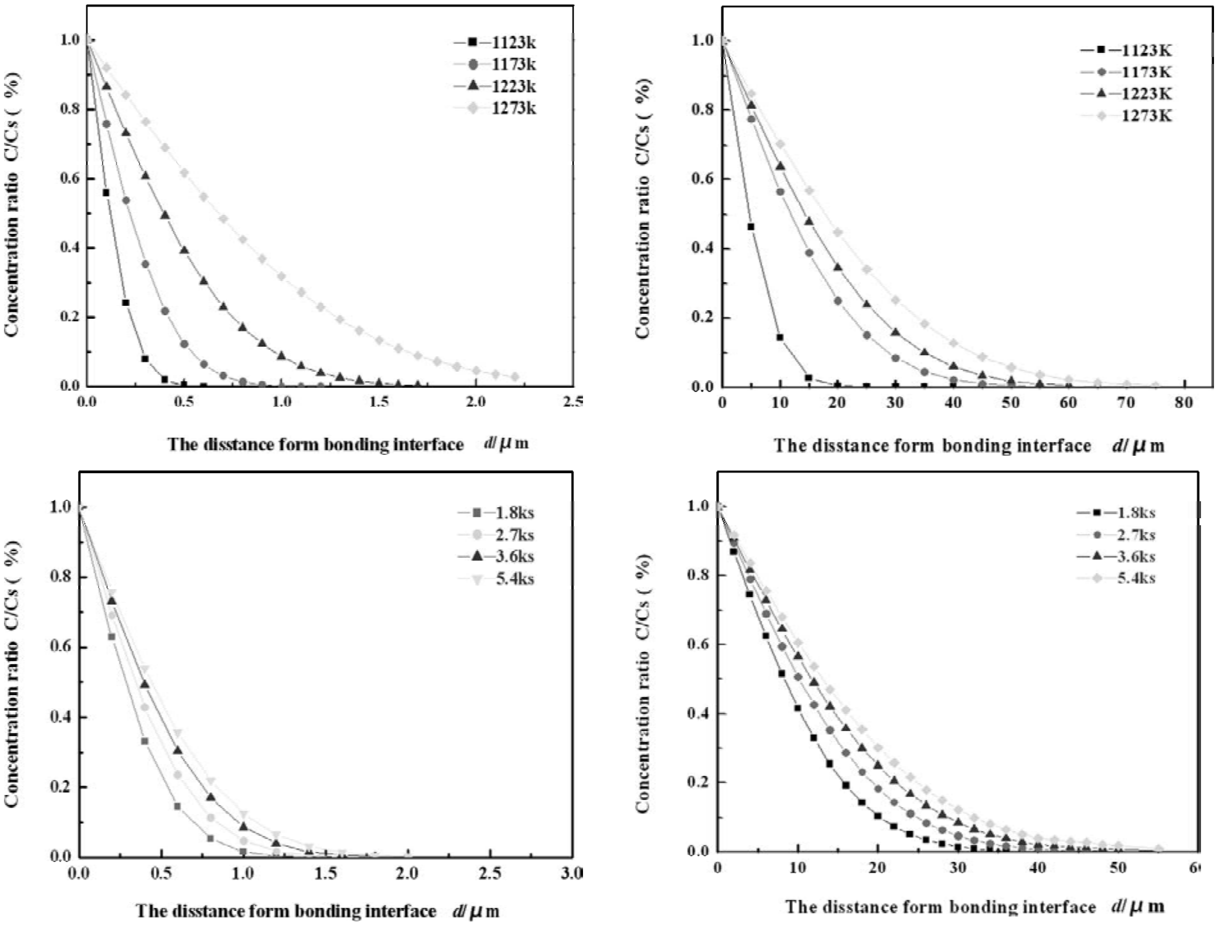

Fig. 6. Concentration distribution of Cu at Ti-6Al-4V and stainless steel side of Ti-6Al-4V/Cu/304 joints. Diffusion of Copper to Fe side: (a) 3.6 ks; (b) 1223K; Diffusion of Copper to Ti side: (c) 3.6 ks; (d) 1223K.

3.4. The analysis of formation mechanism of bonding interface of atomic diffusion and reaction phase

In essence, bonding strength is determined by the interface structure [8]. In order to further reveal the composition of the reaction phase and formation mechanism. The EDS is carried on reaction zone of diffusion and tensile fracture, as show to Fig. 3 (I-IV). The results are shown in Table 3. It is known by the EDS that component is basically titanium alloy matrix near the zone α of reaction layer. Component of Ti is very high, and there is a little Cu and Fe in the zone α and β of diffusion-reaction layer. It maybe generated tittle solid solution of α-Ti (Cu) and some intermetallic compounds of Ti_2Cu. Near zone χ of 304 side, it maybe generate intermetallic compounds of $TiFe_2$. At last, the component is 304 matrix in the δ zone across the interface of 304/Cu. It is shown that interlayer of Cu copper can stop element of stainless steel (such as Fe, Cr, Ni and so on) from bonding to titanium alloy. It can improve joint's strength. But because of the strong activity of Ti, it is easy to react with a variety of metal. So that it reacted strongly between Ti and Cu element. And it is small for interlayer of Cu to stop Ti element from bonding to 304-matrix [9]. Therefore, it forms intermetallic compounds, such as $TiFe_2$ that it is gray diffusion layer in the interface of 304/Cu. For hardness testing show that affect of joint's quality is slightly smaller than Ti_xCu_y compounds. Under the thermal simulation time of 2.7 ks, it forms a little $TiFe_2$ (FeTi) in the diffusion layer of 304 side and much Ti_xCu_y in the diffusion layer of TC4. It does not improve joint's strength by a large extent.

To analyze the line of fracture expansion more clearly, the EDS is carried on box area of tensile fracture in Fig. 4. The results are shown in Table 3. As the fracture of 304 side of Fig. 4a shows, the content of Ti element is 80.53% in the heave area of 304-matrix by the box area enclosed. This shows that it is most of TC4 organization in the area. As the fracture of TC4 side of Fig. 4c shows, the content of Ti element is 56.72% and the content of Fe is 30.29% in the depressed area of TC4 matrix by the

Table 3. Chemical composition of area δ in Fig. 3 and square select the region of Fig. 4.

Condition	Position	Al	Ti	Cu	Fe	Cr	Ni
1223 K	I	08.72	91.28				
2.7 ks	II	08.70	87..81	02.50			
	III		87.44	06.96	04.77	00.45	
	IV		29.18		49.70	16.40	04.73
	V				72.95	19.94	07.11
Fracture	304 side	05.09	80.35		12.90		
	Ti-6Al-4V side		56.72	01.05	30.29	05.74	02.20

Table 4. Comparison of numerical analysis and experimental result.

Condition	Diffusion bonding of thermal simulation1223K, 2.7 ks		Vacuum diffusion bonding 1223K, 3.6 ks	
	Cu bond to Ti	Cu bond to Fe	Cu bond to Ti	Cu bond to Fe
Value of theoretical analysis	45 μm	1.8 μm	50 μm	2 μm
Value of test	60 μm	3 μm	72 μm	5 μm

box area enclosed. And the other content is Cr, Ni, and Cu. This shows that it forms intermetallic compounds in the diffusion layer, such as Ti_2Fe, Ti_xCu_y and so on. It is brittle phase. For interface distribution and hardness testing show that affect of joint's quality Ti_xFe_y is slightly smaller than Ti_xCu_y compounds.

It is conjectured from appearance of fracture and the results of EDS analysis that the heave area of 304 side is pull over from the titanium alloy side. Namely 304 side sticks some TC4 organization. As the results of EDS analysis show, solid solution may form in the zone α. Solid solution strengthening makes joint's strength that it is joints under the bonding temperature of 1223K and the bonding time of 3.6 ks more strongly in vacuum in this study. But intermetallic compounds form in the zone β and χ, such as Ti_2Fe, $TiFe_2$, and Ti_2Cu. This shows that fracture is cracking from the zone of titanium alloy side. Then it expands to intermetallic compounds diffusion layer of zone β and χ. This shows from the appearance of fracture in Fig. 4. Details still need to be further explored.

In addition, bonding interface and fracture of SEM and the results of EDS analysis of vacuum diffusion bonding is similar with the results of thermal simulation.

4. THEORETICAL ANALYSIS

For investigating the diffusion phenomenon of Fe, Ti, and Cu, it calculated on the relationship of bonding distance (X) and concentration (C) by application of Fick's second law. Namely:

$$C/Cs = \operatorname{erfc}\left(x/2\sqrt{Dt}\right).$$

The boundary conditions of found are shown.

When $X = 0$, $t > 0$. $C = C_S$; $X > 0$, $t = 0$. $C = C_0$.

In the formula: C_S is initial concentration, C is concentration of alloy, D is diffusion coefficient, X is bonding distance, t is bonding time [10].

Thus can resolve that relationship between bonding distance and concentration that diffusion of Cu atom to Fe and Ti side is shown in Fig. 6. Analysis contrast with test is shown in Table 4. As the theoretical analysis and test results show, joint's tensile strength of the vacuum diffusion bonding of TC4/Cu/304 is most when the bonding time is 36 ks under the bonding temperature of 1223K. At the moment, diffusion distance that Cu atom to titanium alloy side is 72 μm. And that Cu atom to 304 side is about 5 μm in fact. When the theoretical diffusion time is 3.6 ks, diffusion distance that Cu atom to titanium alloy side is about 50 μm. And diffusion distance that Cu atom to 304-side is about 2 μm.

Compared with test results have a little difference. But thermal simulation results of the shorter bonding time are similar with theoretical analysis. Therefore, it can guide practice combination of test to avoid more test and improve the efficiency of scientific research.

5. CONCLUSION

1. Thermal simulation diffusion bonding of Titanium alloy/Cu/Stainless Steel, under the condition that the bonding temperature was 1223K and the pressure was 5.0 MPa with the increasing thermal simulation time, the joint's tensile strength continuous increase, when time is 2.7 ks, there is a maximum tensile strength that is 94.78 MPa. However, the vacuum diffusion bonding, the joint's tensile strength increased firstly and then decreased, with bonding temperature and time rising, when bonding temperature is 1223K, bonding time is 3.6 ks, there is a maximum tensile strength that is 162.73 MPa. However, it will is disadvantageous to performance of the joints, when bonding temperature and time extended overly. But it can improve the tensile strength, When the reducing the thickness of the middle layer of copper properly.

2. Although it can prevent effectively Fe element and alloying elements from the stainless steel diffusing to titanium matrix when using copper foil to make interlayer. But it failed to prevent Ti element diffusing to the stainless steel. Therefore, it formed intermetallic compounds in the bonding interface, such as $CuTi_2$, Cu_3Ti, Cu_2Ti, CuTi, and FeTi. Ti_xCu_y is the main reason that joints fracture in the diffusion layer along the Titanium and Copper interfaces. Copper should not be too thick as the titanium intermediate layer directly. it formed multi-phase transition organizations by solid solution, intermetallic compounds in the bonding interface, such as $Ti_2CuTi_xCu_y$tie , $Ti_2FeTiFe_2$ and FeTi. Effect of Ti_xFe_y on strength of the joints is slightly inferior the Ti_xCu_y compound. But both's influence location must further discuss.

3. The titanium alloy side has produced the thick intermetallic compound , the break is brittle fracture. The fracture is mainly by the titanium alloy side region III for the source dehiscence, developping in region III-IV diffusion layer of intermetallic compound. Therefore the reasonable adjust technological parameter controls the intermetallic compound level thickness to be essential.

4. As can be seen from the comparison of the thermal simulation and the vacuum diffusion bonding, the thermal simulation diffusion bonding has superiority while the vacuum diffusion bonding has no. That is before bonding, the preparation accuracy requirement to bonding face of specimen is low; in thermal simulation testing, the rate of heating and the cooling are high, saving time, manpower, and cost. And the fracture's nature of thermal simulation joints is big to the plastic development's tendency, If uses the vacuum to have space that the raise joint's strength.

5. Diffusion bonding testing is closely with value analysis result, therefore take theory analysis as directional guiding practice, it may reduce the massive cockamamie experiments, thus may raise efficiency of scientific research.

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