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Thermal Diffusion Bonding of Pure Titanium to 304 Stainless Steel Using Aluminum Interlayer

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As for thermal diffusion bonding of pure titanium to 304 stainless steel using aluminum interlayer, the temperature is ranged from 550 ℃ to 650 ℃ for 2 h under 2 MPa load in vacuum. The microstructures, chemical compositions and reaction products of the transition joints were revealed in SEM, EDS and XRD, respectively. The results have shown that the thickness of the diffusion layer increases with the increase of treatment temperature and the diffusion layer appears on the titanium side at the temperature of 650 ℃. In this process, aluminum not only plays a mid sole element to prevent iron diffusion to titanium master alloys forming brittle intermetallic phase Fe-Ti, but also makes iron elements, titanium and the aluminum element form a stable diffusion layer.

1. Introduction

Titanium and its alloy have been widely used in the aerospace industry, shipbuilding, nuclear industry and so forth, because it has many advantages, such as corrosion resistance, mechanical properties, high temperature resistance, low temperature resistance (Manesh and Taheri, 2003; Dey et al., 2009; Zhang et al., 2012; Kundu and Chatterjee, 2006; Kundu et al., 2005; Mousavi and Sartangi, 2008). On the other hand, stainless steel also has an excellent price (Mudali et al., 1995; Wan et al., 2012; Zhu et al., 2014). Composite plates using titanium and its alloys as surface material, stainless steel as master material, not only has good corrosion resistance, but also can reduce the economic costs. Therefore, titanium and stainless steel-clad plate composited with titanium will have potential and prosperous development.

However, some researcher has reported that, Fe-Ti, Ti-C and other metal brittle phases appeared on the interface if titanium and stainless-steel bind directly, and those metal brittle phases will greatly reduce the mechanical properties between metal binding (Özdemir and Bilgin, 2009; Yu and Zhan, 2014; Sun et al., 2010; Yu et al., 2014; Bemporad et al., 2007). Furthermore, using traditional welding means to connect dissimilar materials, will emerge stress concentration, microcracks and other problems on combined surface, which lead to crevice corrosion and fatigue fracture failure of composite plate (Kundu et al., 2007; Yuan et al., 2008; Elrefaey and Tillmann, 2009).

Diffusion coefficient difference between titanium alloy and stainless steel which lead to bonding crack, so bonding directly with adding interlayer metal is now largely used (Swarup et al., 2010). Aluminum metal has good corrosion resistance and plastic deformation (melting point and prices are lower than copper and nickel metal), using aluminum metal as an intermediate layer between clad plate of the titanium and stainless steel can prevent stress concentration, avoiding the generation of cracks, improving the corrosion resistance properties, reducing energy consumption and so on (Yu et al., 2015). In addition, thermal diffusion section between different metals involves complicated physical and chemical processes. The various constituent elements on the combined interface are the key of bonding strength of titanium steel clad plate. In this paper, we try to analyze the structure of bonding interface and research the composition varied with different temperatures.

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2. Experimental

The chemical composition of 304 stainless steel, commercially pure titanium and pure aluminum are given in table 1. Each material alloy (304 stainless steel, pure titanium and pure aluminum) were cut into 50 mm×50 mm×2 mm by DK7716 type linear cutting machine. Subsequently, all surfaces of the alloys were mechanically polished by 240 #, 360 #, 400 #, 600 #, 800 #, 1000 #, 1200 #, 1500 #, 1800 #, 2000 # paper. Then, all surfaces were cleaned in acetone and dried in air prior to bonding.

Table 1: The Chemical compositions of base metals (wt. %)

Material	C	Fe	Ti	Mn	Si	S	P	Cr	Ni	O	N	H	Al
Ti	0.02	0.10	Bal	-	-	-	-	-	-	0.18	0.012	0.003	-
stainless steel	0.04	Bal	-	2	4	0.03	0.035	17	8	-	0.02	-	-
Al	-	0.40	0.05	0.05	0.25	-	-	-	-	-	-	-	Bal

The oxide layer was easily formed on the metal surface, the titanium side was etched in an aqueous solution (100 ml H_2O) of 3 ml HF and 6 ml HNO_3. The stainless-steel side was etched by mixture of nitric acid alcohol solution 4%. It was etched by 10% NaOH solution for pure aluminum. After removal of the surface oxide layer, all plates were washed by acetone and deionized water, and then dried.

We used HVHP-2 type vacuum hot press (the temperature range 550-650 ℃ for 2 h under 2 MPa load) for thermal diffusion bonding of pure titanium to 304 stainless steel (Figure 1). This is based on the equilibrium phase diagram; the melting point of aluminum metal will be close to 650 ℃ under 2 MPa load. Moreover, we try to research transient liquid phase bonding in comparison with the solid-state diffusion. In this experiment, the microstructures of the transition joints were revealed by scanning electron microscopy, the diffusion zone and their chemical compositions were determined by energy dispersive spectroscopy and the reaction products were confirmed by X-ray diffraction instrument.

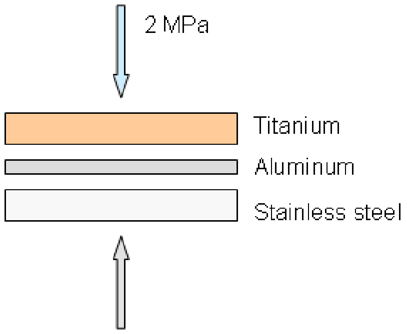


Figure 1: Experimental simulation

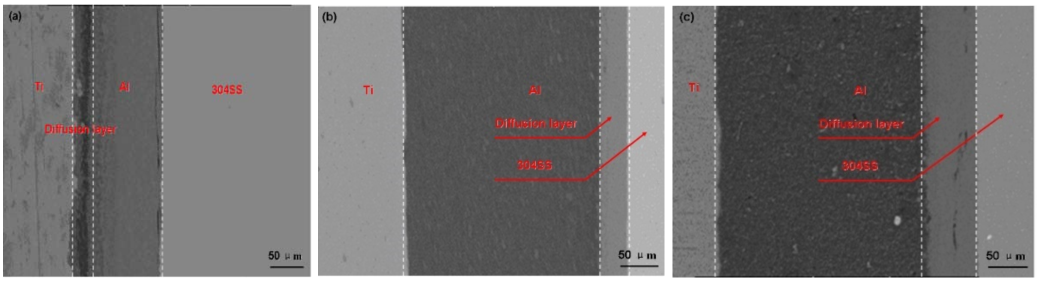
3. Results and discussion

3.1 Surface morphology of the transition joints of Ti-Al-304 stainless steel

Figure 2 shows the SEM image of the transition joints of Ti-Al-304 stainless steel. In Figure 2(a), A is the aluminum interlayer, B is the diffusion interlayer and C is the 304 stainless steel substrate. From these figures, we can clearly find out the diffusion intermediate layer between aluminum and 304 stainless steel, with a thickness of about 20-30 μm. It is observed that, the diffusion intermediate layer is very uniform and combine closely with each other. In addition, there are no obvious cracks in the bonding interface between the Ti/Al and Al/304 stainless steel.

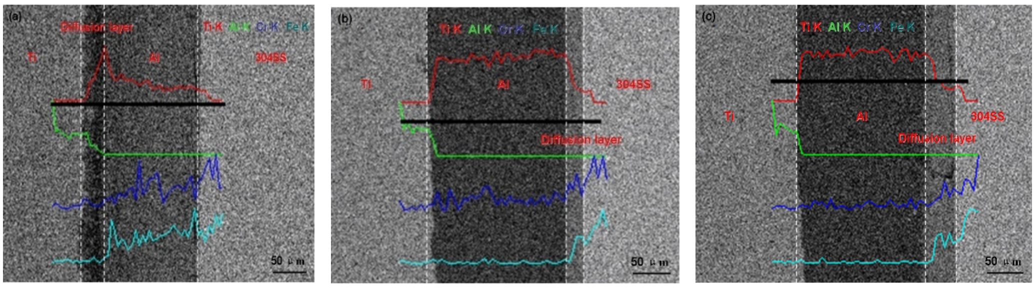
In Figure 2(b), A is the aluminum interlayer, B is the diffusion interlayer and C is the 304 stainless steel substrate. On one hand, at the temperature of 600 ℃, the diffusion intermediate layer is twice thicker than Figure 2(a) with the thickness of about 50-60 μm. This is because the diffusion temperature T is higher than Figure 2(a). On the other hand, the diffusion intermediate layer has passion for hot cracks and eager holes, because the treatment temperature increase will lead to increased vacancies and metal mobility. Thus, the diffusion temperature is an important parameter to determine the thickness and microstructure.

In Figure 2(c), A is the titanium master, C is the aluminum interlayer, B is diffusion intermediate layer. From these figures, the diffusion intermediate layer sharply reduced and produced a stratified sheet. Moreover, it is different from the case of 550 ℃ and 600 ℃, the diffusion intermediate layer appears on the titanium side.



(a) 550 ℃, (b) 600 ℃, (c) 650 ℃

Figure 2: The SEM image of the transition joints of Ti-Al-304 stainless steel by diffusion bond.



(a) 500 ℃, (b) 600 ℃, (c) 650 ℃

Figure 3: The case of diffusion of the four main elements of Al, Ti, Cr, Fe, at the range of 550-650 ℃.

3.2 Elements diffusion of the transition joints of Ti-Al-304 stainless steel

3.2.1 Line scan analysis of the transition joints of Ti-Al-304 stainless steel

Figure 3 shows the diffusion of the four main elements of aluminum, titanium, chromium and iron, especially for the temperature range 550-650 ℃. As can be seen from the figures, there is one or more intermediate layer at transition joints. In those intermediate layers, under constant pressure conditions, at different diffusion temperature, the four-main component of the diffusion element changed slowly.

In Figure 3(a) and 3(b) (in the temperature range 550-600 ℃), temperature mainly affects the diffusion of iron and chromium elements, and has little effect on the titanium and aluminum element, producing a clear interface at the iron side. Moreover, the thickness of diffusion layer increases with the increase of treatment temperature.

In Figure 3(c) (at the temperature of 650 ℃), when the diffusion temperature is close to the melting point, the temperature has a great effect on iron and chromium element, but the effect on titanium element diffusion is still small. It is worth noting that the aluminum element becomes active and dominant, the formation of a multi-layer interface at the titanium side. This shows that the diffusion temperature is close to the melting point of aluminum element, the diffusion activation energy will make some changes.

All the results have been demonstrated that the thickness of the diffusion layer increases with the increase of treatment temperature and the diffusion layer appears on the titanium side at the temperature 650 ℃ due to the change of diffusion activation energy.

3.2.2 Point scan analysis of the transition joints of Ti-Al-304 stainless steel

Microelements properties have great effect on the material properties, so we used EDS point scan to analyze the elements of specific area. Figure 4(a)~(f) are element distribution graphs of titanium side at the temperature range 550-650 ℃. In Figure 4(b) and Figure 4 (d), the microelements did not change significantly, and the main elements of this specific area is titanium element. So we can determine that aluminum and titanium element almost no diffusion at the temperature range 550-600 v. Figure 4(f) shows that the content of aluminum element is greater than that of titanium, and the percentage of aluminum element is 24.16%. This is because the aluminum element diffusion occurs and forms some intermetallic compounds.

Figure 4(g)~(l) are element distribution graphs of iron side at the temperature range 550-650 ℃. Figure 4(h) and Figure 4(j) are the composition analysis region at the temperature range 550-600 ℃. It can be found that the content of aluminum element is 0.44 %, chromium element is 15.06 %, iron element is 83.96 % at the temperature of 550 ℃ (in Table 2). According to Figure 3, iron and chromium diffusion are dominant. This is because iron, especially, chromium element diffusion activation energy is smaller than aluminum and titanium element. The diffusion elements at the temperature of 650 ℃ as shown in Figure 4 (l). In this figure, the content of chromium element is 16.26%, aluminum element is 2.16 %, iron element is 81.58 %. It shows that the diffusion activation energy increases with the increase of diffusion temperature. Meanwhile, some diffusion has also occurred in aluminum.

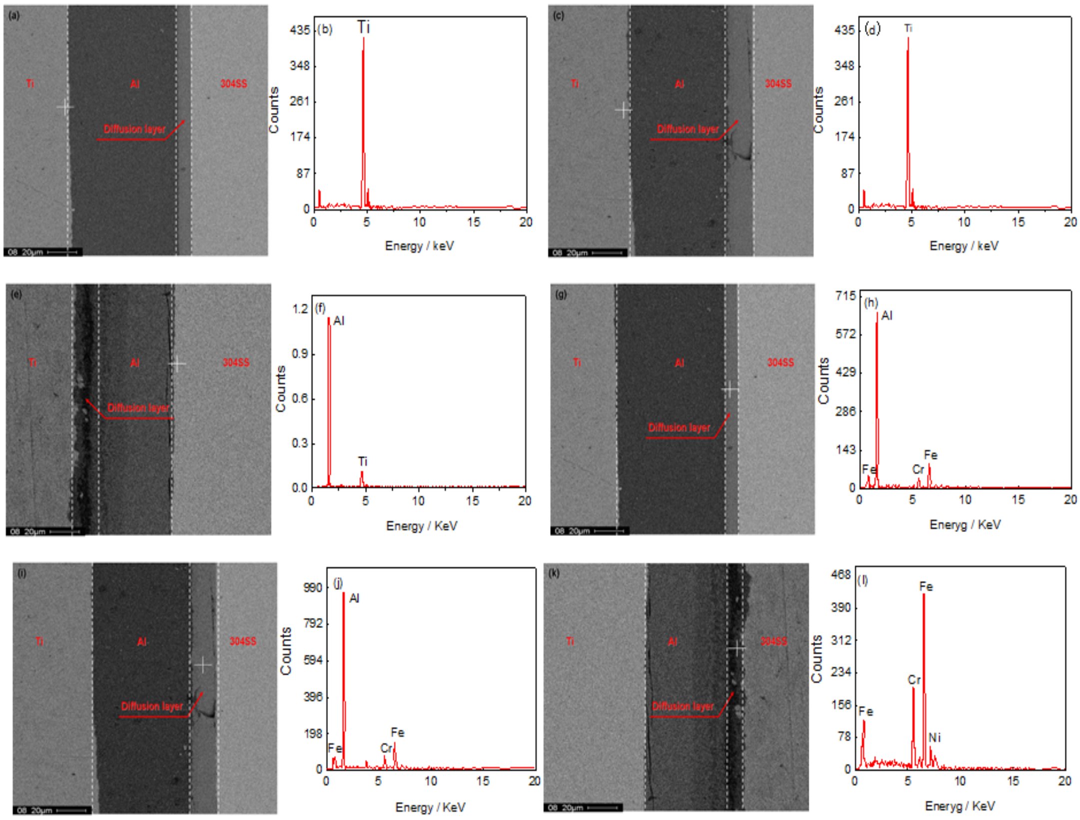


Figure 4: The EDS point scan of composite plat (Ti side: (a)(b) 550℃ (c)(d) 600℃ (e)(f) 650℃; 304SS side: (g)(h) 550℃ (i)(j) 600℃ (f)(g) 650℃)

3.3 Section analysis

According to Table 2-3, iron and chromium element are more active than titanium and aluminum element. Therefore, we choose to analyze the stainless steel side diffusion interface. Figure 5 is the XRD pattern of the phase composition analysis for the diffusion interface at the temperature of 550 ℃. We find that, as the temperature gradually increased, the diffusion element and the phase composition gradually increased.

Table 2: At the temperature of 550, 600, 650 ℃, the composition content at Ti/Al region

Temperature (^{o}C)	Ti (Wt%)	Al (Wt%)	Fe (Wt%)
550	100	0	0
600	100	0	0
650	75.84	24.16	0

Table 3: At the temperature of 550, 600, 650 ℃ the composition content at Al/304SS region

Temperature (^{o}C)	Al (Wt%)	Cr (Wt%)	Fe (Wt%)
550	0.44	15.06	83.96
600	0.21	15.87	83.92
650	2.16	16.26	81.58

Among them, the phase composition at the temperature of 550 ℃ is shown in Figure 5 (a). It can be found that the main phase of the interface is Al and Fe. This is because when the temperature is low, the diffusion activation energy is not high enough, and it has not produced metal compound. The phase composition at the temperature of 600 ℃ is shown in Figure 5 (b). In this figure, when the temperature increased, the main phase at the interface is $FeAl_6$, Fe_3Al and $FeAl_2$. This is because the increase of the diffusion temperature, which leads to the increase of the diffusion activation energy, the diffusion coefficient increases. Iron atoms enter the aluminum lattice to form metal compound.

In Figure 5 (c), temperature not only affect on the thickness of the diffusion, but also on the diffusion layer and the phase composition. The main phase of the interface is FeCr, Fe_3Ni_2. There is not Al element, that is because the treatment temperature is close to the melting point, the diffusion of aluminum is fast, iron and aluminum will not contact closely.

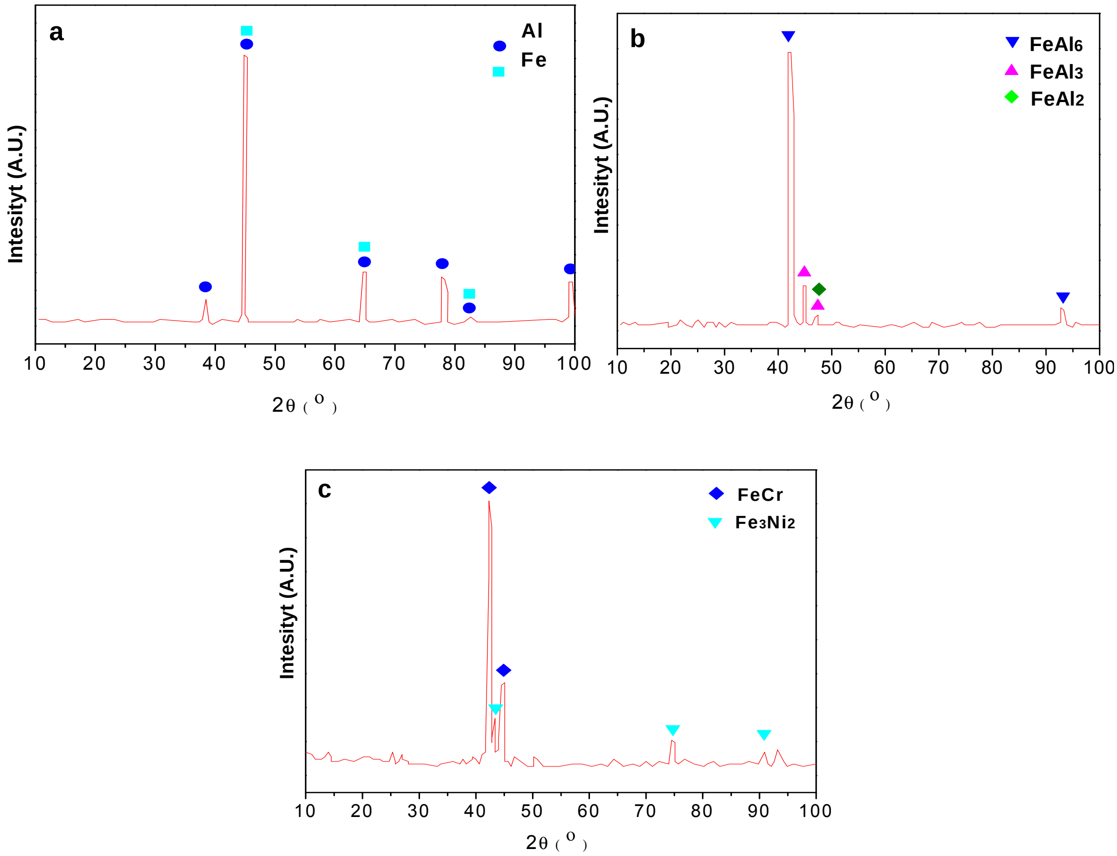


Figure 5: X-ray diffraction pattern of fracture surface at 550℃, 600v, 650℃

4. Conclusions

Aluminum and titanium elements are not substantially diffusion at the temperature range 550-600 ℃, the main phases at the interface are $FeAl_6$ when the temperature increased Fe_3Al and $FeAl_2$. At the temperature of 650℃, the thickness of the diffusion, the diffusion layer and the phase composition are affected by temperature. The main phases at the interface are FeCr, Fe_3Ni_2. In general, aluminum metal not only plays a mid sole element to prevent iron diffusion into titanium master alloys forming brittle intermetallic phase Fe-Ti, but also makes iron element and the aluminum element form a stable diffusion layer.

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Reference

Bemporad E., Sebastiani M., Casadei F., Carassiti F., 2007, Author links open the author workspace. Modelling, production and characterisation of duplex coatings (HVOF and PVD) on Ti–6Al–4V substrate for specific mechanical applications, Surface & Coatings Technology, 201(18), 7652-7662. DOI: 0.1016/j.surfcoat.2007.02.041

Dey H.C., Ashfaq M., Bhaduri A.K., Prasad K.R., 2009, Author links open the author workspace. Joining of titanium to 304L stainless steel by friction welding, Journal of Materials Processing Tech, 209(18), 5862-5870. DOI: 10.1016/j.jmatprotec.2009.06.018

Elrefaey A., Tillmann W., 2009, Solid state diffusion bonding of titanium to steel using a copper base alloy as interlayer, Journal of Materials Processing Technology, 209(5), 2746-2752. DOI: 10.1016/j.jmatprotec.2008.06.014

Ghosh S.K., Chatterjee S., 2010, On the Direct Diffusion Bonding of Titanium Alloy to Stainless Steel, Materials & Manufacturing Processes, 25(11), 1317-1323. DOI: 10.1080/10426914.2010.520793

Kundu S., Chatterjee S., 2006, 2006 Interfacial microstructure and mechanical properties of diffusion-bonded titanium–stainless steel joints using a nickel interlayer, Materials Science & Engineering A, 2006, 425(1–2), 107-113. DOI: 10.1016/j.msea.2006.03.034

Kundu S., Chatterjee S., Olson D., Mishr B., 2007, Effects of Intermetallic Phases on the Bond Strength of Diffusion-Bonded Joints between Titanium and 304 Stainless Steel Using Nickel Interlayer, Metallurgical and Materials Transactions A, 38(9), 2053-2060. DOI: 10.1007/s11661-007-9273-8

Kundu S., Ghosh M., Laik A., Bhanumurthy K., Kale G.B., Chatterjee S., 2005, Diffusion bonding of commercially pure titanium to 304 stainless steel using copper interlayer, Materials Science & Engineering A, 2005, 407(1–2), 154-160. DOI: 10.1016/j.msea.2005.07.010

Manesh H.D., Taheri A.K., 2003, The effect of annealing treatment on mechanical properties of aluminum clad steel sheet, Materials & Design, 24(8), 617-622. DOI: 10.1016/S0261-3069(03)00135-3

Mousavi S.A.A.A., Sartangi P.F., 2008, Effect of post-weld heat treatment on the interface microstructure of explosively welded titanium–stainless steel composite, Materials Science & Engineering A, 494(1–2), 329-336. DOI: 10.1016/j.msea.2008.04.032

Mudali U.K., Dayal R.K., Gnanamoorthy J.B., 1995, Corrosion Behavior of Weldments of Ti and Ti- 5Ta for Nuclear Fuel Reprocessing Plants, Journal of Materials Engineering and Performance, 4(6), 756-760. DOI: 10.1007/BF02646452

Özdemir N., Bilgin B., 2009, Interfacial properties of diffusion bonded Ti-6Al-4V to AISI 304 stainless steel by inserting a Cu interlayer, The International Journal of Advanced Manufacturing Technology, 41(5), 519-526. DOI: 10.1007/s00170-008-1493-6

Sun D.Q., Gu X.Y., Liu W.H., Xuan Z.Z., 2005, Welding consumable research for austempered ductile iron (ADI), Materials Science & Engineering A, 402(1–2), 9-15. DOI: 10.1016/j.msea.2005.01.034

Wan W., Xiong J., Guo Z., Dong G., Yi C., 2012, Effects of Cr 3 C 2, addition on the corrosion behavior of Ti(C, N)-based cermets, International Journal of Refractory Metals & Hard Materials, 31(6), 179-186. DOI: 10.1016/j.ijrmhm.2011.10.013

Yu X., Rong J., Zhan Z., Liu Z., Liu J., 2015, Effects of grain size and thermodynamic energy on the lattice parameters of metallic nanomaterials, Materials & Design, 83, 159-163. DOI: 10.1016/j.matdes.2015.06.019

Yu X., Zhan Z., 2014, The effects of the size of nanocrystalline materials on their thermodynamic and mechanical properties, Nanoscale Research Letters, 9(1), 1-6. DOI: 10.1186/1556-276X-9-516

Yu X., Zhan Z., Rong J., Liu Z., Li L., 2014, Vacancy formation energy and size effects, Chemical Physics Letters, 600(600), 43-45. DOI: 10.1016/j.cplett.2014.03.046

Yuan X.J., Sheng G.M., Qin B., Huang W.Z., Zhou B., 2008, Impulse pressuring diffusion bonding of titanium alloy to stainless steel, Transactions of the China Welding Institution, 59(7), 930-936. DOI: 10.1016/j.matchar.2007.08.003

Zhang B., Wang T., Chen G., Feng J., 2012, Contact Reactive Joining of TA15 and 304 Stainless Steel Via a Copper Interlayer Heated by Electron Beam with a Beam Deflection, Journal of Materials Engineering and Performance, 21(10), 2067-2073. DOI: 10.1007/s11665-012-0132-4

Zhu Y.P., Li C.Y., Zhang L.Y., 2014, Corrosion resistance and antibacterial activity of different zones in TA2 weldment by TIG welding, Rare Metals, 1-8. DOI: 10.1007/s12598-014-0395-z