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Journal of Alloys and Compounds 466 (2008) 363–367

Journal of ALLOYS AND COMPOUNDS

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# Microstructure and fracture properties of reaction-assisted diffusion bonding of TiAl intermetallic with Al/Ni multilayer foils

J. Cao, J.C. Feng *, Z.R. Li

*State Key Lab of Advanced Welding Production Technology, Harbin Institute of Technology, Harbin 150001, China*

Received 2 September 2007; received in revised form 5 November 2007; accepted 11 November 2007
Available online 19 November 2007

## Abstract

Reaction-assisted diffusion bonding of TiAl intermetallic was performed using Al/Ni multilayer foils as interlayers. The typical microstructure and the joining mechanism were investigated. The $Ni_3(AlTi)$, $Ni_2AlTi$, $NiAl_2Ti$ and (Ni,Al,Ti) reaction phases were observed at the interface between the Al/Ni multilayer foils and the TiAl substrate. The reaction products in the multilayer foils were mostly composed of $AlNi_3$. The joining between the multilayer and the substrate relied on the formation of Ti–Al–Ni compounds. The joining temperature played a strong influence on the microstructure of the joint. The fracture mainly took place between the $NiAl_2Ti$ layer and the substrate and the fracture presented a typical brittle characteristic.

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*Keywords:* Intermetallics; Thin films; Microstructure; Scanning electron microscopy

## 1. Introduction

The multilayer foils have been of great interest owing to their new structural, chemical, magnetic, optical and electronic properties. Especially, Al/Ni multilayers have attracted considerable attentions as metallurgical coating due to the excellent oxygen resistance [1]. The reaction-assisted diffusion bonding consists of introducing reactive foils as interlayer materials. The heat generated in the interlayer assisted the joining process and improved the joining quality [2]. The use of reactive multilayer foils as interlayers represents an exciting addition to the field of joining [3,4]. The energy generated by the exothermic reaction in the reactive multilayer foils assists the diffusion bonding process and enhances the joining efficiency [5,6].

Titanium aluminide is a potentially attractive material for high-temperature structural applications [7]. However, high-melting point of TiAl makes conventional fusion welding difficult [8]. Although some studies [9–11] have reported on the successful diffusion bonding of TiAl, few investigations on the reaction-assisted diffusion bonding of TiAl with Al/Ni multilayer foils have been systematically carried out. Accordingly, the objective of this work was to investigate the microstructure and the joining mechanism of the TiAl joint using Al/Ni multilayer foils as interlayers. In addition, the effect of joining temperature on the microstructure was discussed during the joining process.

## 2. Experimental

The TiAl-based alloys applied in this research had nominal composition of Ti–48Al–2Cr–2Nb (at.%). One surface was polished by SiC papers up to grit 1200 and ultrasonically cleaned by acetone. Al/Ni multilayer foils were deposited onto the polished surface of TiAl samples using magnetron sputtering, by rotating a substrate over fixed Al and Ni guns. The thin films were alternating submicron layers of nickel and aluminum with a total thickness of 20–30 μm. The morphology of as-deposited Al/Ni multilayer foils is presented in Fig. 1. In this photograph, the white zone was Ni layer and the dark zone was Al layer. The interface between Al and Ni layer was clear and no obviously intermixing region was found.

The schematic diagrams of reaction-assisted diffusion bonding experiments are shown in Fig. 2. The sizes of the substrate for metallographic observation introduced in Fig. 2(a) and (b) were Ø 10.0 mm × 1.5 mm. The dimensions of the TiAl specimens in Fig. 2(c) for strength test were 10.0 mm × 1.5 mm × 1.5 mm and 10.0 mm × 30 mm × 5 mm, respectively. The process was carried out in a high-vacuum heating apparatus at different heating rates varying between 20 and 30 K/min. The joining temperature was applied ranging from 973 to 1173 K and the holding time was kept at 10 min. The vacuum pressure was about $5 \times 10^{-6}$ Torr and the joining pressure varied from 35 to 55 MPa. After joining process, the microstructure of the products were characterized employing X-ray

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* Corresponding author. Tel.: +86 451 86416607; fax: +86 451 86416607.
*E-mail address:* feng_jicai@163.com (J.C. Feng).

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doi:10.1016/j.jallcom.2007.11.033

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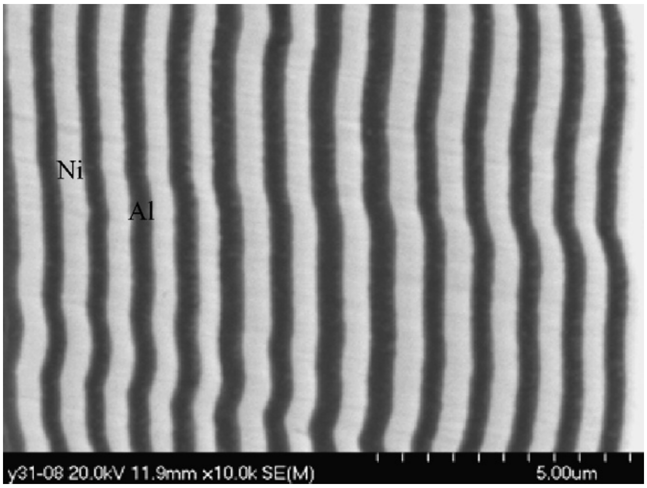

Fig. 1. The microstructure of the as-deposited Al/Ni multilayer foils.

diffraction (XRD) and scanning electron microscopy (SEM) with electron probe X-ray microanalysis (EPMA). Strength tests were performed at a constant speed of 0.5 mm/min by a universal testing machine (Instron 1186).

## 3. Results and discussion

The reaction characteristics in term of the reaction heat and ignition temperature were important factors to evaluate the multilayer foils. In order to release the residual stress, it was necessary for the reaction products to be homogeneous with the substrates in physical and chemical properties. The properties of Al–Ni products, which were similar to those of TiAl substrate, and the low-ignition temperature of Al–Ni system (similar with Ti/Al multilayer[12]) led to the selection of the Al/Ni multilayers for the joining. The contents of Al and Ni in the multilayer foils were selected to achieve the highest heat of reaction and the best-assisted effect. It was well acknowledged that the $Al_3Ni$ or $Al_9Ni_2$ phases were the first products in the reaction of Al/Ni multilayer foils and the reaction was exothermic [13–15]. After the total consume of Al, these Al-rich products may react with residual Ni to form AlNi. If some amounts of unreacted Ni still existed, the AlNi may further reacted with Ni to form $AlNi_3$. Because the reactions to form AlNi and $AlNi_3$ were both exothermic, the ratio of the Al to the Ni layer thickness was adjusted to produce a 1:3 atomic ratio.

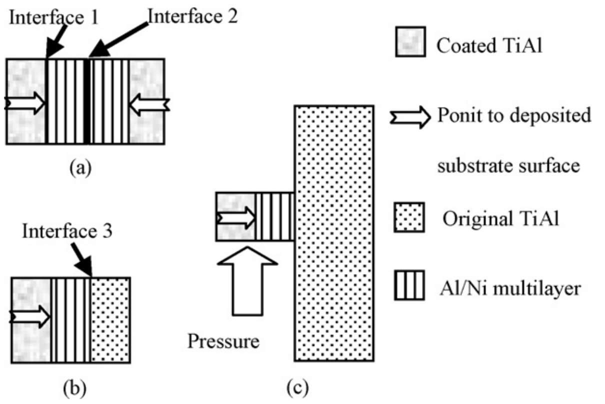

Fig. 2. Schematic diagram of the experiment for (a, b) metallographic observation and (c) strength test.

Table 1
Composition analysis of the different microstructural elements of the joint

| Zone | Color | Composition (at.%) | | | Phase |
|---|---|---|---|---|---|
| | | Ti | Al | Ni | |
| $A_1$ | Dark | 35.14 | 32.71 | 32.15 | (Ni,Al,Ti) |
| $A_2$ | Grey | 23.33 | 52.51 | 24.16 | $NiAl_2Ti$ |
| B | Grayish-white | 23.42 | 26.83. | 49.75 | $Ni_2AlTi$ |
| C | White | 18.53 | 19.37 | 62.10 | $Ni_3(AlTi)$ |
| D | Light | – | 25.67 | 74.33 | $AlNi_3$ |
| E | Grayish-white | – | 49.74 | 50.26 | AlNi |

Fig. 3 depicts the representative microstructure of the joint of TiAl using Al/Ni multilayer foils as interlayers (Fig. 2(a) type). The joining process was performed at 973 K for 10 min. The integral microstructure of the joint is shown in Fig. 3(a). The reaction-assisted diffusion bonding of TiAl intermetallics was successfully achieved under these parameters. The porosity, which was inevitable, using powder compacts as interlayers, was not observed in the reaction products. Both Al and Ni in the foils took part in the reaction and no original multilayer structures were represented. The interfacial microstructure is shown in Fig. 3(b). Complex reaction phases were observed at the interface. According to EPMA results (shown in Table 1) of the joint, it was noted that the dark reaction layer (Layer A) consisted of two phases. The main phase ($A_1$) was confirmed to be $NiAl_2Ti$ and the other grey phase ($A_2$) was (Ni,Al,Ti) compound. The grayish-white reaction layer (Layer B) was composed of $Ni_2AlTi$ phase. The white layer (Layer C) at the interface was $Ni_3(AlTi)$ phase. Though the reaction in the Al/Ni foils was insufficient, the assisted effect of the reaction heat was significant because the direct diffusion bonding without the foils failed under these parameters. The light granular structures (Zone D) were $AlNi_3$ phases and large amounts of grayish-white phases (Location E) were AlNi phases. Although the Al/Ni atomic ratio in the foil was 1/3, the predominant phase in Fig. 3 appeared to be AlNi. The non-equilibrium and insufficient reaction derived from the heat transfer to the substrate resulted in this special phenomenon. The magnification microstructure at the joining interface is presented in Fig. 3(c). The sound joining regions were obtained under these parameters. However, the unbonded regions at the interface indicated the existence of unsatisfied joining zone.

The X-ray diffraction patterns of the reaction products and as-deposited multilayer foils are presented in Fig. 4. For as-deposited multilayer, Al and Ni were detected in the diffraction results and no compounds of Ni–Al systems existed. The results were in good agreement with the microstructure in Fig. 1. After the reaction in the multilayer foils was completed, $AlNi_3$ was the main reaction products. It was noted that the reaction was carried out in a freestanding foil without substrate. Therefore, the reaction in the freestanding foil was performed completely. In addition, a small amount of AlNi phase was identified and no peaks of unreacted multilayers were detected in the reaction products.

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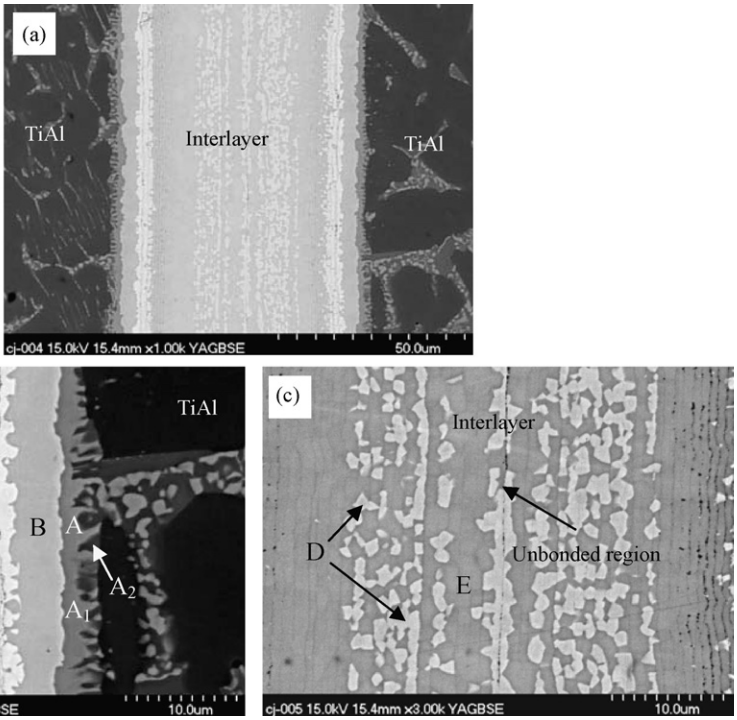

Fig. 3. Back-scattered electron images of the joint microstructure (973 K, 10 min, Fig. 2(a) type) (a) integral microstructure (b) near the substrate and (c) at the interface between the multilayers.

From the above analysis in Fig. 4, it was observed that there were some unbonded zones at the interface. The high-joining temperature was applied to increase the joining quality. The microstructure with a higher joining temperature is shown in Fig. 5 (Fig. 2(a) type). The joint processed at 1173 K presented nearly invisible interfaces and almost no deficiency was observed. The reaction and diffusion in the multilayer foils was enough and the reaction products were homogenous. The interfacial reaction layer was thicker than that of low-joining temperature. The high temperature led to the sufficient reaction in the foils. At high temperature, the Ni-rich phase reacted with AlNi to form $AlNi_3$. Thus, the reaction products were $AlNi_3$ and only a small amount of Ni-rich phase was found at the interface.

There were two series of interfaces in the joint shown in Figs. 3 and 5. One interface lied between the Al/Ni multilayer and the deposited TiAl surface (named as interface 1), the other one distributed between the Al/Ni multilayers (named as interface 2). The joining at the interface 1 was much better than that at the interface 2 from the microstructural analysis. In the present case, this was most likely due to the close contact and the adequate reaction at the interface 1. The interface 2 was free surface and the deformation was difficult due to the hardening effect in the deposited process. Compared Fig. 3 with Fig. 5, it was concluded that the joining temperature played a strong influence on reaction products and interfacial microstructure. When the Al/Ni multilayer was heated to 973 K, the sequence of reactions in the multilayer was $Al + Ni \rightarrow Al_3Ni + Ni \rightarrow AlNi + Ni$.

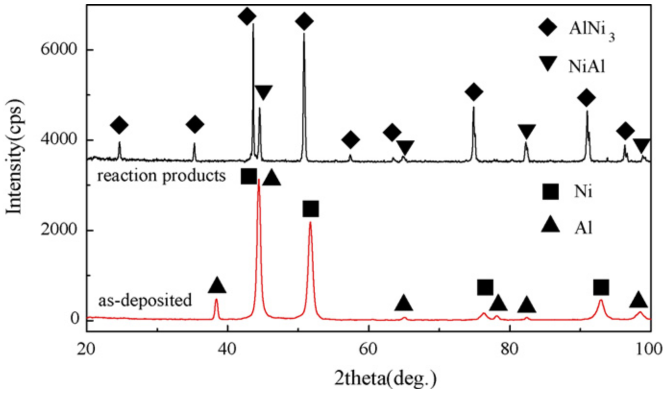

Fig. 4. X-ray diffraction pattern of the reaction products and as-deposited multilayers.

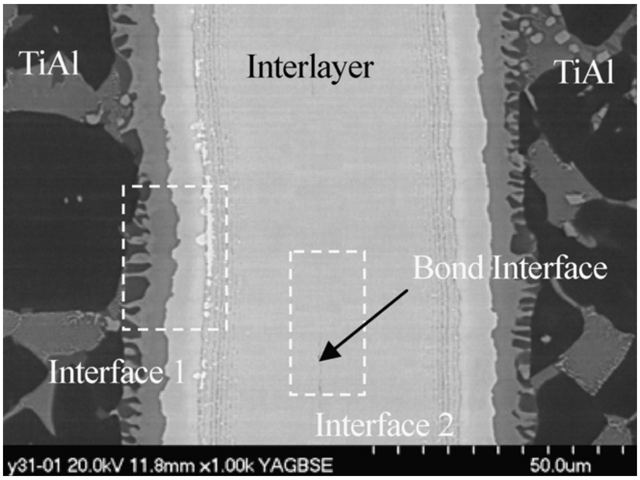

Fig. 5. Back-scattered electron images of the joint microstructure with higher joining temperature (1173 K, 10 min, Fig. 2(a) type).

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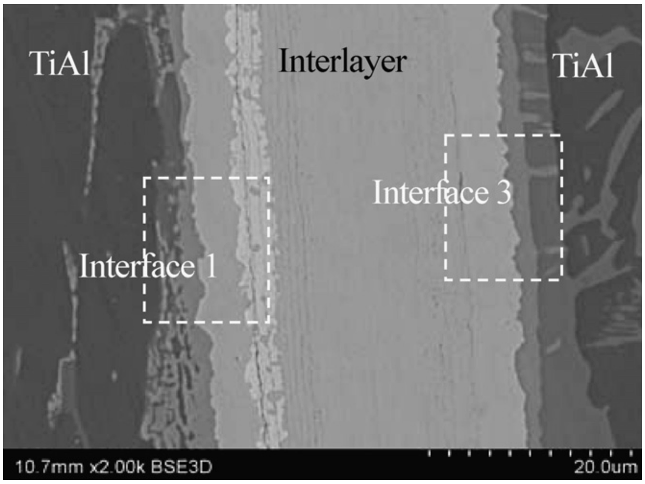

Fig. 6. Back-scattered electron images of the joint microstructure using original TiAl (1173 K, 10 min, Fig. 2(b) type).

Thus the reaction products consisted of large amounts of AlNi phase. When the multilayer was heated to 1173 K, the reaction AlNi + Ni → $AlNi_3$ occurred and $AlNi_3$ phase was observed in the final products.

The simplification of the joining process is necessary in order to apply this method more widely. Depositing on the substrate is difficult to accomplish when the actual components have complex shape and huge dimension. Hence, the application of freestanding multilayer foils in diffusion bonding and the investigation on the interface between the Al/Ni multilayer and original TiAl substrate (named as interface 3) are meaningful. The type of joint in Fig. 2(b) is designed to compare the interface 3 with the interface 1 and the microstructure of this joint type is presented in Fig. 6. The reaction-assisted diffusion bonding of TiAl was successfully realized under the same parameters applied in Fig. 5. The sufficient reaction in the multilayer and the homogeneous reaction products were observed. The structure of the interface 1 was similar to that shown in Fig. 3. However, there were some unbonded regions observed at the interface 3. At the interface 3, the thorough reaction took place and thick reaction layer was produced. Due to the relatively poor surface contact, the heat transfer from the multilayer to the substrate was smaller than that at the interface 1. The main heat loss for the reaction-assisted process was the heat transfer to the substrate. Thus, the heat was concentrated in a very narrow region at the interface and the assisted effect of the foils was obvious. The reaction in the multilayer foils in the interface 3 side was sufficient and no unreacted Ni was observed at the interface 3.

In order to investigate the influence of the interface 1 and interface 3 on the joining quality, the quality test shown in Fig. 2(c) was carried out. Fig. 7 gives the SEM photographs of fracture surfaces joined with Al/Ni multilayers. The average shear strength of the joint was about 160 MPa. The overall fracture path is shown in Fig. 7(a). The fracture path was determined by combining the microstructural observation with composition

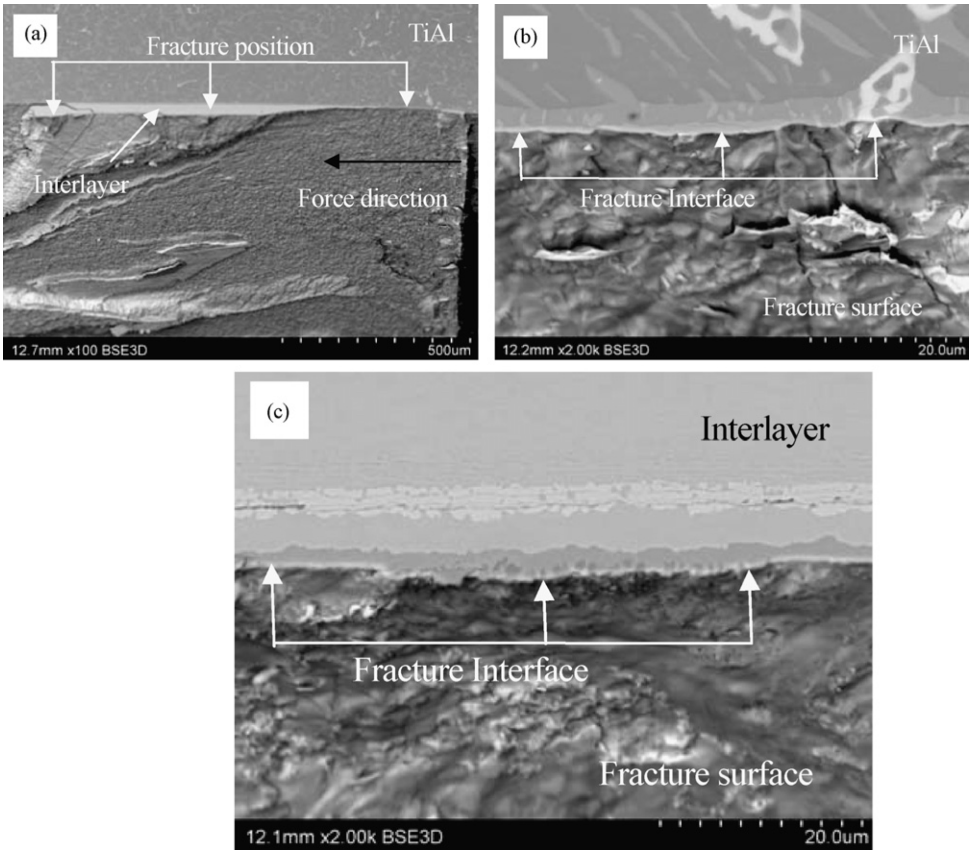

Fig. 7. Micrograph of fracture surface (a) overall fracture path, (b) at the interface 3 and (c) at the interface 1.

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analysis. The failure originated at the interface 3, but the fracture did not spread along the interface 3 completely. Partial fracture extended to the interface 1. Fig. 7(b) presents the main fracture surface. Combined with the EPMA result, it was confirmed that the accurate fracture mainly took place between the $NiAl_2Ni$ reaction layer and the TiAl substrate. The fracture belonged to a typical brittle fracture. The location of the fracture at the interface 1 is shown in Fig. 7(c). The fracture also occurred between the $NiAl_2Ni$ reaction layer and the TiAl substrate. No crack propagated in the multilayer foils. The evidence showed that reaction products were the strong joining parts in the reactive joint. It was worth noting that partial fracture extended in the substrate. The results did not represent intrinsic shear strengths of the bonds but indicated reasonably strong joining in spite of some microstructural defects.

According to the results mentioned above, it was concluded that the reaction-assisted diffusion bonding with Al/Ni multilayers was an effective way to join TiAl compared with traditional diffusion bonding method. For the direct diffusion bonding of TiAl, the high temperature and long-holding time were necessary to obtain no defect joint [16]. When the Ni/Al multilayer foils were applied as interlayers, the diffusion bonding process was successfully realized with relatively low temperature and short time. When the solder was sandwiched between the substrate and the Al/Ni multilayer, the heat of reaction in the foils melted the solder in a short time [5]. Based on this, the assisted effect of the reaction heat on the joining process was significant. Because the reaction products were the mixtures of different intermetallics and no phases with low-melting point existed, the joint could be applied in a high-temperature condition. Hypothetically considering further improvement of the joining efficiency without requirement for high-temperature application, the simplest way was to place solder or metal with low-melting point between the multilayer and the substrate, as demonstrated [6].

From the microstructural observation, it was found that complex Ni–Al–Ti compounds were produced between the Al/Ni multilayer foils and the substrate. In the joining systems, the interfacial joining for the interfaces 1 and 3 depended on the formation of Ni–Al–Ti ternary compounds, while the joining for the interface 2 was attributed to the diffusion of Al–Ni reaction products. The diffusion bonding between the same materials usually achieved more satisfied results. Thus the fracture tended to occur at interfaces 1 and 3. From the analysis of element content, the Ti and Al content decreased correspondingly with the gradation increase of the Ni content from the substrate to the multilayer. The gradation diffusion at the interface led to the formation of interfacial structure (shown in Fig. 3). Ni was the main diffusion element in the reaction system because of its largest concentration gradation between the Al/Ni multilayer foils and the TiAl substrate.

## 4. Conclusions

To summary, the reaction-assisted diffusion bonding of TiAl using Al/Ni multilayer foils as interlayers was successfully achieved. The effect of joining temperature on the microstructure was studied. The final reaction products in the multilayers were $AlNi_3$ phase. The interfacial reaction phases between the multilayers and the substrate were confirmed to be $Ni_3(AlTi)$, $Ni_2AlTi$, $NiAl_2Ti$ and (NiAlTi), respectively. Some unbonded regions were observed when the joining temperature was 973 K. With the increase of the joining temperature, the reaction and diffusion at the interface became adequate and sound bonds were produced at 1173 K. Interfacial Ti–Al–Ni compounds connected the Al/Ni multilayer foils with TiAl substrate. The fracture analysis showed that the failure mainly took place between the $NiAl_2Ti$ reaction layer and the TiAl substrate.

## Acknowledgement

The authors gratefully acknowledge the financial support from Project 50325517 supported by National Natural Science Foundation of China.

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