The Influence of the Processing Temperature on the Microstructure of γ -TiAl Joints Brazed with a Ti-15Cu-15Ni alloy

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Abstract. The joining of Ti-47Al-2Cr-2Nb (at.%) by diffusion brazing at 1000, 1050 and 1100°C for 10 min, using a Ti-15Cu-15Ni (wt.%) braze alloy was investigated. The microstructure and the chemical composition of the interfaces were studied by scanning electron microscopy (SEM) and by energy dispersive X-ray scans (EDS), respectively. The reaction between the γ -TiAl alloy and the braze alloy led to the formation of layered interfaces, which should be essentially composed of α_2 -Ti₃Al and of Ti-Ni-Cu-Al and Ti-Ni-Cu intermetallic compounds. Raising the brazing temperature increases the amount of Al rich intermetallic compounds (Ti-Ni-Cu-Al) and the extension of the interface. The increase of the thickness of the interface is primarily due to the growth of the reaction layer formed near the γ -TiAl alloy.

Introduction

The implementation of γ -TiAl alloys as structural materials will require the development of reliable and cost-effective joining techniques. Diffusion brazing [1] and diffusion bonding [2-4] are joining techniques that allow to produce joints with mechanical properties comparable to those of γ -TiAl alloys at high temperature. Ideally, diffusion bonded and diffusion brazed joints have the same composition and structure of the parent material, and therefore the same mechanical properties. Diffusion bonding causes less disruption of the surface of the components to be joined and produces more homogeneous interfaces than diffusion brazing. However, diffusion brazing offers some advantages over diffusion bonding: joining is carried out at lower temperatures and without the need of applying elevated pressures to promote bonding. The mechanical behaviour of diffusion brazed joints is highly dependent on the features of the interface, namely on the chemical and mechanical characteristics of the reaction products and their distribution across the interface; these are in turn affected, for a given system, by the processing variables such as the brazing temperature, the brazing holding stage and the heating and cooling rates. Therefore, it is crucial to assess the influence of these variables in order to optimise the joining of γ -TiAl alloys by diffusion brazing.

Materials and Experimental Procedures

The γ -TiAl alloy (Ti-47Al-2Cr-2Nb, at.%) used in this investigation has a duplex microstructure and was produced from gas atomised elemental powders by Crucible Research. Samples of the intermetallic alloy with 10 x 7 x 3 mm were cut with a diamond saw and then ground with SiC emery paper down to 1200 grade. A Ticuni foil (Ti-15Cu-15Ni, wt.%) with a thickness of 90 μ m was used as filler metal. Ticuni is a clad-laminated alloy that consists in a sandwich of a Cu-Ni foil between two 37 μ m Ti foils. Prior to joining both alloys were degreased in acetone with ultrasonic agitation, dried in air and then assembled into a sandwich type. A small pressure (50 Pa) was applied to the γ -

TiAl alloy/Ticuni/ γ -TiAl alloy assemblage during the brazing thermal cycle. Brazing was carried out at 1000, 1050 and 1100°C for 10 min in a vertical furnace, which was evacuated to a vacuum level better than 10^{-4} mbar. For all processing conditions the heating and cooling rates were fixed at $3^{\circ}\text{C}.\text{min}^{-1}$. Cross-sections of the joints were prepared using standard metallographic techniques. The interfaces were examined by SEM and chemically analysed by EDS at an accelerating voltage of 15 keV.

Materials and Experimental Procedures

Each processing condition used in this investigation led to the formation of layered interfaces that are composed of three distinct reaction layers. These layers are labelled as *A*, *B* and *C* on the back-scattered electron images (BEI) of the interfaces shown in Figs. 1 to 4.

Figure 1 – Interface for joining at 1000°C.

Figure 2 – Interface for joining at 1050°C.

Figure 3 – Interface for joining at 1100°C.

Figure 4 – Layers *A*, *B* and part of layer *C*, for joining at 1000°C, where two types of dark zones are shown, labelled as D_1 and D_2 .

The extensions of the interface and of the reaction layers are listed, for all processing conditions, in Table 1. The interface grows from about 122 to 194 μm , when the brazing temperature is raised from 1000 to 1050°C. This increase results essentially from the growth of layer *A*, once the thickening of this layer represents approximately 60% of the total growth of the interface. However, the extension of the interface remains practically the same when the brazing temperature is raised from 1050 to 1100°C.

Table 1 – Thickness (μm) of the reaction layers and of the interface for all processing conditions.

Layer	1000°C	1050°C	1100°C
<i>A</i>	21	42	44
<i>B</i>	8	17	16
<i>C</i>	64	73	74
Interface ($A \times 2 + B \times 2 + C$)	122	191	194

The compositions of the reaction products detected at the interfaces are listed in Table 2. All compositions are expressed, hereafter, in atomic percentage. For all processing conditions, the composition of the base intermetallic remained practically unaltered after joining. Small amounts of Cu and Ni, both of them never exceeding 0.6%, were detected in the γ -TiAl alloy but only near the interface (no farther than 6 μm away from the interface).

EDS does not allow the identification of the phases that constitute the interface. However, the heating and cooling rates were slow enough ($3^\circ\text{C}.\text{min}^{-1}$) to consider that a quasi-equilibrium state was achieved at the interface during the entire processing thermal cycle. Therefore, equilibrium phase diagrams could be used in conjunction with SEM and EDS as predictive tools of the nature of the phases that constitute the interface. Since the interfaces are essentially composed of Ti, Al, Ni and Cu the equilibrium quaternary phase diagram would provide valuable information, but unfortunately it has not yet been established. However, one could assume, as Lee *et al.* [5, 6] did that Ni and Cu atoms behave in a similar way since they both have similar atomic sizes and electronegativities, the same crystalline structure and unlimited solid solubility. In addition, Cu and Ni show practically the same distribution across the interface. Therefore, the Ti-Cu-Al and the Ti-Ni-Al ternary phase diagrams can be used as a last resort to bypass the lack of the quaternary phase diagram, as long as one adds Cu and Ni contents together.

All dark zones are essentially composed of Ti and Al ($89.2\% < (\text{Ti}+\text{Al}) < 98.4\%$) with Ti:Al atomic ratios comprised between 2.0 and 3.3. In layer *A*, two types of dark zones were detected; they will be designated as D_1 and D_2 (see Fig. 4). D_2 zones are brighter than D_1 zones and significantly richer in Cr and Nb. In layers *B* and *C*, only D_1 dark zones were detected. According to the Ti-Al phase diagram [7], the α_2 -Ti₃Al compound is stable below 500°C for Ti:Al atomic ratios comprised between 1.9 and 3.5 approximately. Therefore, layer *B*, which is an irregular and quasi-continuous dark zone (D_1) and all dark zones in layer *A* (D_1 and D_2) and in layer *C* (D_1) should correspond to the α_2 phase.

The white zones in layer *A* are plotted near the single phase field Ti(Ni,Cu)Al, on the isothermal sections of the Ti-Cu-Al and of the Ti-Ni-Al ternary phase diagrams presented in Fig. 5. Hence, layer *A* should be composed of mixture of α_2 -Ti₃Al (dark zones) and of Ti(Ni,Cu)Al (white zones).

Table 2 – Average composition (at. %) of some of the reaction products detected at the interface.

Temperature (°C)	Layer / zone	Ti	Al	Cu	Ni	Cr	Nb
1000	A / D ₁	73.3	25.1	0.9	0.3	0.3	0.2
	A / D ₂	60.5	29.4	2.0	1.2	3.9	3.0
	A / white	38.8	33.3	11.6	14.5	1.2	0.5
	B	73.6	24.5	1.1	0.4	0.3	0.1
	C / D ₁	73.9	24.3	1.0	0.5	0.2	0.2
	C / white	58.7	13.1	10.8	15.5	1.6	0.3
1050	A / D ₁	67.0	29.6	0.8	0.3	0.7	1.5
	A / D ₂	60.0	29.2	2.3	1.5	4.8	2.2
	A / white	39.6	33.2	10.5	13.7	2.0	1.0
	B	73.2	24.5	0.8	0.5	0.2	0.9
	C / D ₁	73.8	22.1	1.7	1.1	0.5	0.9
	C / white	58.2	11.7	11.7	15.3	2.6	0.6
1100	A / D ₁	65.5	31.1	1.0	0.2	0.8	1.4
	A / D ₂	60.2	30.5	2.5	1.2	3.2	2.4
	A / white	38.7	32.7	11.1	14.9	2.0	0.8
	B	73.8	24.3	0.8	0.3	0.1	0.8
	C / D ₁	73.0	24.2	1.1	0.7	0.2	0.9
	C / white	57.3	12.5	11.5	15.2	2.6	0.9

Although most the white zones in layer C appear to be single-phase, the plots on the isothermal sections invariably indicate that they are composed of three phases, $Ti_2(Ni,Cu) + Ti(Ni,Cu)_2Al + Ti_3Al$. This inconsistency could be explained by the volume interaction on the EDS analysis of the zones that are below and around the analysed area, namely the interaction due to the black zones. So most of the white zones should be in fact richer in Cu and Ni and poorer in Ti than it is indicated by the EDS analysis. Therefore, in comparison to the plots on Fig. 5, their composition should be shifted towards the $Ti_2(Ni,Cu) + Ti(Ni,Cu)_2Al$ field. The detection of coarse white particles located at the centre of layer C confirms the formation of these two compounds: for joining at 1000°C (28.7Ti-24Al-20.5Cu-26.6Ni) these particles should correspond to $Ti(Ni,Cu)_2Al$ (see Fig. 6), however, in a previous experiment, it was found that for joining at 980°C, these particles (68.5Ti-1.7Al-5.7Cu-24Ni) should correspond to $Ti_2(Ni,Cu)$ [8]. When joints are processed at 1050 and 1100°C, coarse white particles as those detected for joining at 1000°C are observed not only at the centre of layer C but throughout it. In addition, as shown in Fig. 7, layered white zones that are composed of a mixture of these two compounds are also detected. The centre of these zones (28.8Ti-23.7Al-19.9Cu-27.4Ni) is surrounded by a thin layer that has a different composition (68.2Ti-1.7Al-11.7Cu-15.3Ni). The centre and the surrounding layer should correspond to $Ti(Ni,Cu)_2Al$ and to $Ti_2(Ni,Cu)$, respectively.

Figure 5 – Ti-Ni-Al and Ti-Cu-Al isotherms [9] in which the compositions of some of the reaction products detected at the interfaces are plotted. For all plots, Ni and Cu contents are added together.

Figure 6 – Interface for joining at 1000°C, showing coarse elongated white zones located at the centre of layer C.

Figure 7 – Interface for joining at 1050°C, showing layered white zones in layer C.

We presume that, for all processing conditions used in this investigation, most of the white zones of layer C are in fact composed of a mixture of two phases: $Ti_2(Ni,Cu)$ and $Ti(Ni,Cu)_2Al$. When the brazing temperature is raised the dissolution of the γ -TiAl alloy and the diffusion coefficient of Al are higher, thus more Al is able to reach layer C, favouring the formation of Al richer compounds. This should result in the increase of the amount of $Ti(Ni,Cu)_2Al$ particles.

Conclusions

(1) Joining a Ti-47Al-2Cr-2Nb alloy at 1000, 1050 and 1100 °C for 10 min with a Ticuni braze alloy produces multilayered interfaces that should be essentially composed of α_2 - Ti_3Al , Ti-Cu-Ni-Al and Ti-Ni-Cu intermetallic compounds.

(2) For all processing conditions the interface could be divided into three distinct reaction layers that are essentially composed of α_2 -Ti₃Al + Ti(Ni,Cu)Al (layer A) / α_2 -Ti₃Al (layer B) / α_2 -Ti₃Al + Ti₂(Ni,Cu) + Ti(Ni,Cu)₂Al (layer C), starting from the γ -TiAl surface towards the centre of the interface.

(3) Raising the brazing temperature induces a more intense dissolution of the intermetallic alloy. As a consequence, more Al is incorporated into the joint area and the amount of Ti(Ni,Cu)₂Al increases; Ti(Ni,Cu)₂Al coarse particles are detected throughout layer C when joints are processed at 1050 and 1100°C and not only at its centre as for processing at 1000°C.

(4) Raising the brazing temperature also increases the extension of the interface. The increase is mainly due to the growth of layer A. However, the growth rate slows when the processing temperature is raised from 1050 to 1100°C.

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