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Solid-state diffusion bonding of gamma-TiAl alloys using Ti/Al thin films as interlayers

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Abstract

Alternating nanometric layers of titanium and aluminium were used as filler material to promote joining between titanium aluminide samples. The improved diffusivity of these nanometric layers is thought to overcome the difficulties in solid-state joining of titanium aluminides without producing chemical discontinuities at the interface. In this study, a thin multilayer (alternating titanium and aluminium layers), 2 μm thick, was deposited by dc-magnetron sputtering onto the two surfaces to be joined. The effects of processing conditions and the thickness of nanometric layers on microstructure and chemical composition variation across the interface have been analyzed. Sound regions can be obtained at temperatures as low as 600 °C but higher temperatures (800–1000 °C) are needed to obtain completely sound joints. During processing, the as-deposited film evolves to a nanocrystalline TiAl layer which may explain why the bond region is slightly harder than the base material.

Keywords: A. Titanium aluminides, based on TiAl; B. Bonding; C. Joining; C. Thin films; F. Electron microscopy, scanning

1. Introduction

TiAl-based intermetallics have received special attention due to their potential for high temperature structural service, particularly for transport applications [1–3]. Their high specific strength, excellent creep strength and good oxidation resistance make these alloys particularly appropriate for the aerospace and automobile industries. The effective utilization of titanium aluminides has been delayed by their poor workability, and the development of reliable techniques to join TiAl to themselves and to other materials is indispensable to make these alloys more attractive for utilization in real components.

Despite some satisfactory results obtained with fusion welding methods [4–6], the processes present some difficulties, and very careful control of the welding parameters, namely the cooling rate, is required to avoid solid-state cracking.

Solid-state welding techniques, which overcome the solidification cracking problems, such as diffusion bonding [7–12], brazing [13–16] and diffusion brazing [17], are less problematic and have been applied in the production of high integrity welds of TiAl alloys.

Diffusion bonding is ideal for joining sheets and small components with simple joint geometry since it produces joints with no abrupt microstructural discontinuity; it induces a minimum deformation of the parts to be joined and does not require a liquid phase to promote bonding, which always causes some degradation of the base materials. The major disadvantages of the process are the accurate surface preparation and the prolonged high temperature stages required to promote bonding.

This paper describes joining experiments on γ-TiAl alloys by diffusion bonding using alternating nanometric layers of titanium and aluminium as filler material. Taking advantage of the improved diffusivity of these nanometric layers the

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production of bonds using lower temperatures and shorter times is evaluated. Both the microstructure and the hardness of the interface region are studied in order to characterize the joints obtained.

2. Materials and experimental procedures

The γ-TiAl alloys used in this investigation have a chemical composition of Ti–45Al–2Cr–2Nb at.%. Samples of these alloys were cut to $10 \times 10 \times 7$ mm and one of the surfaces was polished down to 1 μm diamond suspension.

Ti/Al multilayer thin films were deposited onto the polished surface of the TiAl samples by dc-magnetron sputtering using Hartec semi-industrial equipment with two cathodes. The thin films are alternating nanometric layers of titanium and aluminium with a total thickness of 2–2.5 μm and 4 nm period, the multilayer begins and ends with a titanium layer of 2 or 50 nm thickness and has an overall chemical composition close to 48–50 at.% of Al. The schematic illustration of the coated samples and the morphology of an as-deposited Ti/Al multilayer are presented in Fig. 1.

Diffusion bonding experiments were performed in a furnace with a vacuum level of less than 10^{-4} mbar. The samples were heated up to maximum temperature, ranging from 600 to 1000 °C, and held for 1 h under a pressure of 50 MPa. Heating and cooling rates of 10 °C/min were used for the lowest temperatures and 20 °C/min were used for 900 and 1000 °C.

In order to perform the microstructural and chemical characterizations of the interface, cross-sections of the joints were prepared using standard metallographic techniques. The interfaces were examined by scanning electron microscopy (SEM), under atomic number contrast, and chemically analyzed by energy dispersive X-ray spectroscopy (EDX) at an accelerating voltage of 15 keV. For transmission electron microscopy (TEM) thin foils were prepared by double jet electrolytic polishing in a solution of 60% methanol, 35% ethylene glycol and 5% perchloric acid. EDX analysis was also performed in TEM specimens under 200 KeV.

The mechanical characteristics of the interface were evaluated using ultramicrohardness tests. The experimental indentation tests were performed in a Fisherscope H100 computer-controlled ultramicrohardness testing system. A Vickers indenter was used in all indentation tests. The hardness tests consisted of a loading stage followed by a 30 s holding period at the maximum load and an unloading stage down to 0.5 mN. This load was then maintained for another period of 30 s. During the test, the load is increased (or decreased) in steps with a 0.5 s period until the nominal test load is reached. In this study, the number of steps was 60 and the maximum applied load was 30 mN. During all the tests, the environmental conditions were fixed (temperature $T = 22$ °C; relative humidity $H_{rel} = 50\%$) and the thermal drift in the penetration depth caused by temperature fluctuations was corrected. The corrected load/penetration depth data were introduced into a computer programme to determine the hardness [18].

3. Results and discussion

To obtain a sound bond, a good adhesion between the two matching surfaces and between the TiAl base material and the multilayers is necessary. In general, union between thin films and substrates was observed after the bonding experiments. Lack of union and porosity, when present, usually occurs at the centre of the joint between the matching interlayers. The adhesion between the matching interlayers is the dominant factor in obtaining a sound joint. Typical microstructures of joints resulting from processing at 600 °C using a multilayer with 4 nm period that starts and ends with a thicker (50 nm) titanium layer are presented in Fig. 2. The analysis of this figure clearly demonstrates that Ti/Al multilayers can produce regions with an apparent soundness, which means that they are a suitable filler alloy. However, as already stated in a previous study [19], the joints processed at 600 °C still present lack of union on the edges of the samples and some porosity in localized regions; an example is shown in Fig. 2(b). The joint also exhibits some non-uniformity in the interface; the brighter zones in Fig. 2(a) are associated with the presence of titanium rich regions that result from the fact that the two matching layers are composed of titanium. The transition between this enriched area and the rest of the interface is very diffuse, indicating that at this

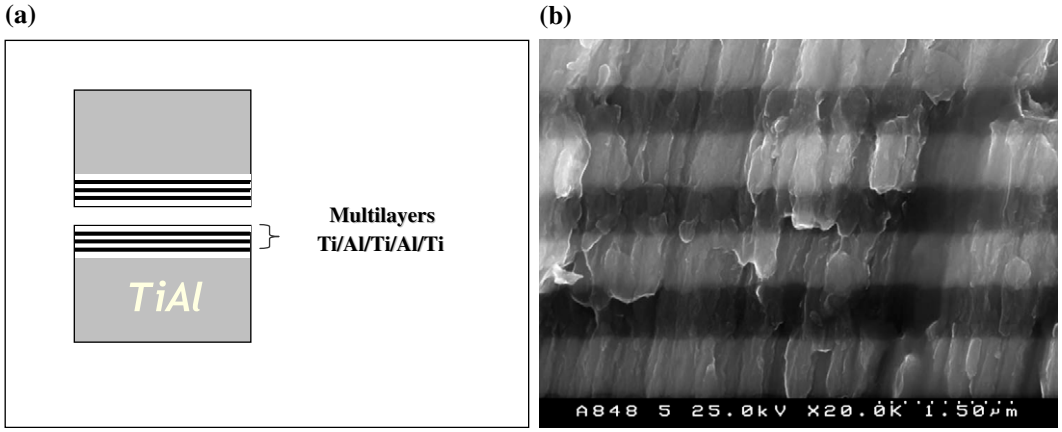

Fig. 1. (a) Schematic illustration of the coated TiAl samples and (b) SEM image of a Ti/Al multilayer produced by dc-magnetron sputtering.

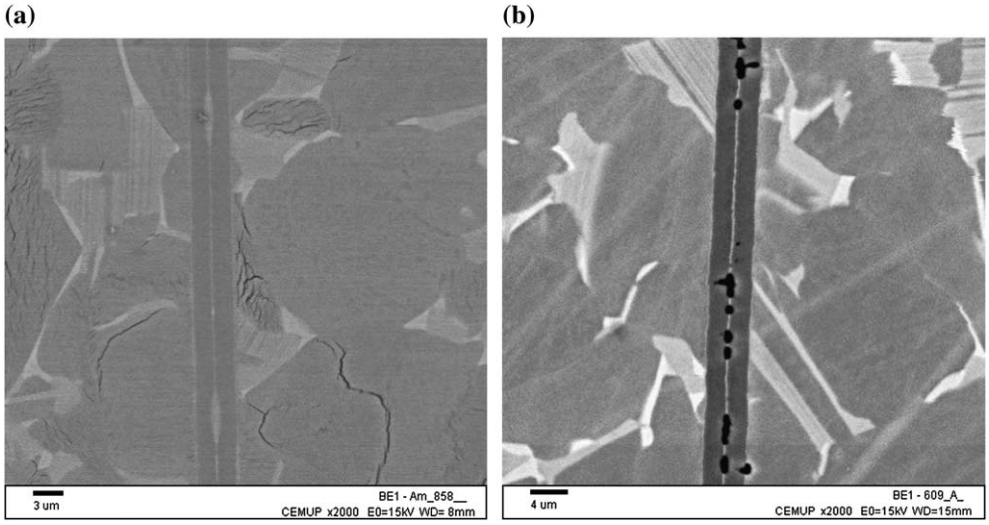

Fig. 2. Backscattered electron images of the joint of samples coated with a multilayer of Ti/Al with 4 nm period processed at 600 °C/50 MPa/1 h. The multilayer starts and ends with a thicker titanium layer (50 nm).

temperature the entire interface is composed of only one phase (γ-TiAl). This conclusion is corroborated by studies [20] showing that Ti/Al multilayers with nanometric periods (less than 200 nm) heat treated at 600 °C for 1 h evolve to γ-TiAl; small amounts of α_2-Ti_3Al are found in some samples. Similar results have been obtained by Kim et al. [21] whose research indicates that the crystallization of an amorphous $Ti_{53}Al_{47}$ thin film starts at 500 °C and γ-TiAl + α_2-Ti_3Al phases were formed during annealing at 600 °C for 1 h.

Tests at high processing temperatures and multilayers without the thicker superficial titanium layer were used trying to eliminate the porosity, lack of union and chemical heterogeneities present at the interface. The superior limit of the temperatures tested (1000 °C) was chosen in comparison to the lower temperatures used in diffusion bonding experiments without filler alloys [10].

The joints processed at 700 °C present a microstructure similar to the joints described above for 600 °C at these temperatures the number of successful joints is reduced. Fig. 3 shows the backscattered electron images of the SEM observations for samples processed at 800 °C. No problems concerning central porosity are observed but there is still a lack of union on the edges of the samples. At these two temperatures a thicker titanium surface layer (50 nm) is essential to obtain a sound joint, as it was the case for lower temperatures. Probably, this more deformable titanium layer not only reduces the importance of the surface conditions and improves bondability but it is also responsible for the formation of a titanium rich layer at the joint centre that might reduce mechanical properties.

Figs. 4 and 5 show the backscattered electron images of the SEM observations for samples processed at 900 and 1000 °C,

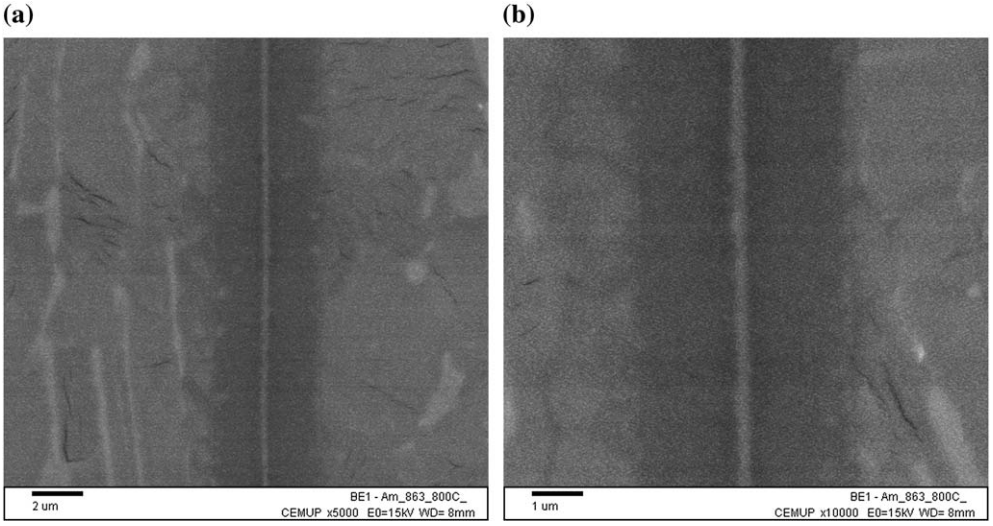

Fig. 3. Backscattered electron images of the joint of samples coated with a multilayer of Ti/Al with 4 nm period processed at 800 °C/50 MPa/1 h. The multilayer starts and ends with a thicker titanium layer (50 nm).

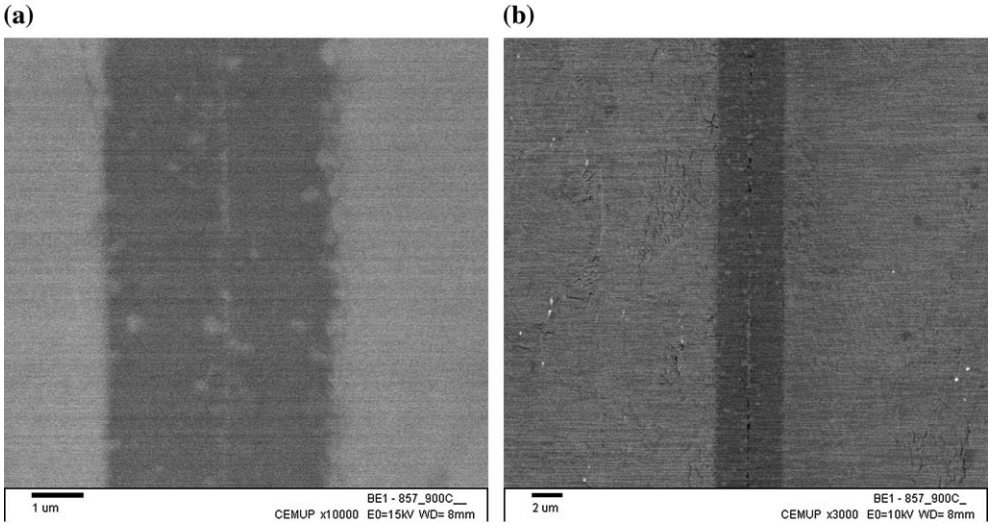

Fig. 4. Backscattered electron images of the joint of samples coated with a multilayer of Ti/Al with 4 nm period processed at 900 °C/50 MPa/1 h. White areas are titanium richer zones.

respectively. At these temperatures, sound joints can be obtained even without the thicker titanium layer at the multilayer surface. However, at 900 °C this final thicker layer is probably still important as the central region presents small pores, observable in Fig. 4(b). Finally, at 1000 °C the interface presents a continuous and regular joint and no porosity was observed.

At the two higher temperatures, the joint reveals some white regions, indicated by the white arrows in Fig. 5(a). Due to the small dimension of these regions it was not possible to determine its chemical composition. However, these regions could be associated with the presence of α_2-Ti_3Al phase as an EDX analysis shows Ti enrichment. The EDX analysis of the interface reveals the presence of Cr and Nb, which confirms the occurrence of an intense diffusion. In fact, these elements are absent in the deposited multilayer and are not detected when the joints are processed at the lower temperatures.

Fig. 6 shows the TEM observations for the sample processed at 1000 °C. The figure also reveals a sound joint without discontinuities either in the interfaces of multilayer/base material or multilayer/multilayer. The higher temperature used has promoted a total crystallization of the multilayers and a grain growth, as expected.

Nanocrystalline grains with sizes ranging from 50 to 300 nm replaced the alternated layers of titanium and aluminium. EDX analyses show that the interface region is mainly composed of γ-TiAl grains with a chemical composition of 47.09 at.% of Ti (average composition of grains marked γ in Fig. 6(b)) and a few α_2-Ti_3Al grains with 62.84 at.% of Ti (composition of the grain marked α in Fig. 6(b)). The selected area diffraction pattern of interface grains corroborates these results and Fig. 6(d) shows the [011] zone-axis diffraction pattern of the grain marked as γ_1 in Fig. 6(b) and (c).

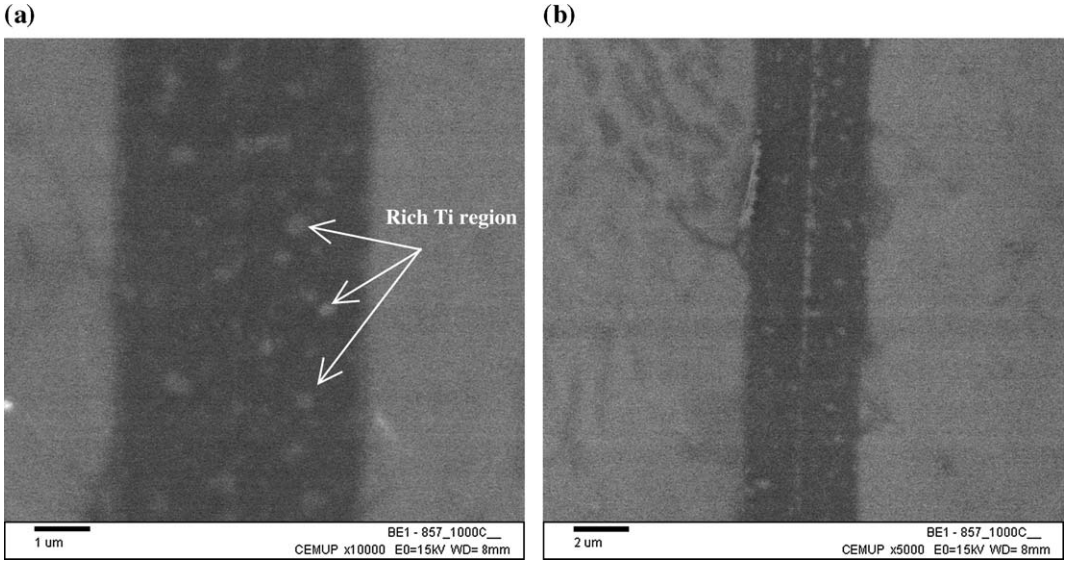

Fig. 5. Backscattered electron images of the joint of samples coated with a multilayer of Ti/Al with 4 nm period processed at 1000 °C/50 MPa/1 h.

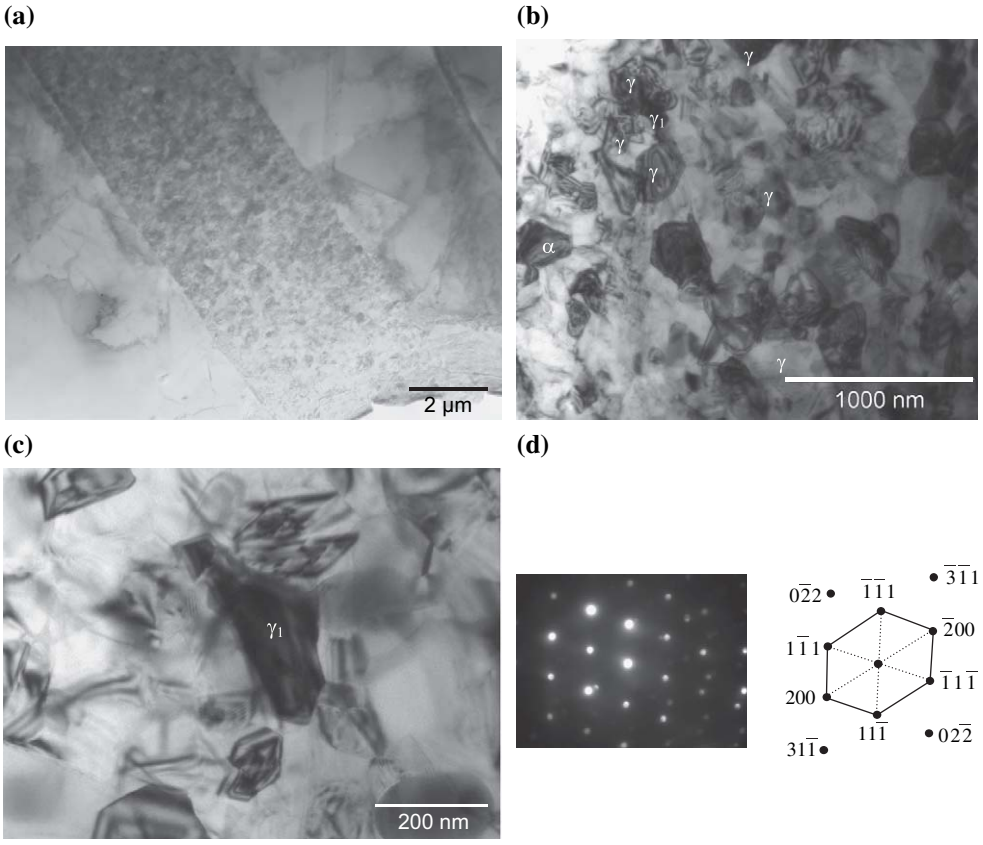

Fig. 6. (a), (b) and (c) TEM observations of nanocrystalline TiAl grains observed in the interface region of a joint processed at 1000 °C/50 MPa/1 h; grains marked as γ are γ-TiAl and the one marked as α is α_2-Ti_3Al. (d) Selected area diffraction pattern of γ_1 grain in (b) and (c).

The evaluation of the interface hardness was performed by ultramicrohardness tests. The hardness of the TiAl base alloy is 5.59 ± 0.69 GPa, independent of joining temperature. Joining at 600 and 800 °C produces interfaces with similar hardness values, 10.18 ± 1.07 and 10.20 ± 0.88 GPa, respectively. These hardness values are almost double the TiAl alloy one, 10.19 versus 5.59. Joints obtained at higher temperatures do not present such an abrupt change at the interface, after joining at 900 and 1000 °C the hardness drops to 7.37 ± 0.39 and 8.60 ± 0.54 GPa, respectively. The lower hardness after joining at higher temperatures is consistent with the grain growth observed on the TEM microstructure presented in Fig. 6.

4. Conclusions

This study investigated the joining of γ-TiAl alloys using alternating nanometric layers of titanium and aluminium as filler material. The results indicate that these nanometric layers are promising filler alloys for diffusion bond titanium aluminides at low temperatures and shorter times.

For low joining temperatures, 600–800 °C, results are inconsistent and a thick titanium layer at the two matching surfaces is essential to promote bonding.

At 900 °C bonding occurs without the need for a thicker titanium layer at the interface, the joint still presents some porosity and lack of union.

No defects were observed at the joints processed at 1000 °C, an interlayer of nanocrystalline grains assures the bonding.

Acknowledgement

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