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Transient Liquid Phase bonding of γTiAl using Al films and SPS method

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ABSTRACT

Purpose: Investigation of joining γ-titanium aluminides using Al interlayers.

Design/methodology/approach: : In order to achieve a proper joining of TiAl elements, a method known as Transient Liquid Phase (TLP) bonding was combined with Spark Plasma Sintering (SPS). All the experiments were performed under a uniaxial pressure of 7.4 MPa in a vacuum to prevent possible oxidation of liquid Al in the interlayer. The study analyzed the difference in microstructure of joints achieved with Al film thicknesses of 50, 100, 200 and 300 μm, as well as two different bonding temperatures of 900 and 1100°C. After bonding the samples were cross-sectioned, polished using 220-1200 mesh SiC paper, and 1/4 μm diamond dispersion, and analyzed using OM, SEM, EDS and EBSD, while Vickers hardness tests were performed to compare the hardness of the joint to that of the base material.

Findings: The analysis of the microstructures revealed that a joint has been created between the two starting γTiAl elements. The bond consisted of Ti_xAl_y phases, with some presence of Ti_2AlC and TiC. The Vicker's hardness tests reviled in some cases similar results to those of base material.

Research limitations/implications: Presence of carbon rich phases within the joint, are the result of graphite mold and filling what react with molten Aluminum. This can be counteracted by replacing graphite with prefabricated TiAl powder after SHS. That however results in increasing the amount of phases such as Ti_3Al, $TiAl_2$ and $TiAl_3$, different from the original γTiAl within the joint. An increase in bonding time and temperature could allow for faster homogenization in the joint, but would make the process more expensive and time-consuming.

Practical implications: The process allows for creation of strong joints, showing signs of homogenization. Low temperature and short time makes it a possible candidate for joining γTiAl elements.

Originality/value: The use of Al instead of commonly used Cu or Ni, allows minimalization of introduce impurities into the joint, while Al is still cheap and common and thus doesn't increase the cost of the process.

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Keywords: γTiAl; TLP bonding; Al films; SPS; Vickers hardness

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MATERIALS

1. Introduction

TiAl intermetallic, mainly γTiAl has been considered as a great engineering material due to its unique combination of properties, especially at elevated temperatures. It has already been assessed that use of γTiAl to replace the currently popular aerospace materials might result in an improvement in both quality and price. The reasons for that are some of γTiAl's properties including [1]:

- high melting point,
- low density,
- high specific strength and moduli,
- good oxidation and corrosion resistances.

On the other hand, low workability of γTiAl makes it hard to apply for creation of complex components. Hence, many conventional brazing techniques including induction brazing [2], gas tungsten arc welding [3] and diffusion bonding [4] were investigated for joining of γTiAl elements. It was found, however, that all of them have some limitations.

In recent years the method of Transient Liquid Phase (TLP) bonding has been implemented for joining of materials, including γTiAl. In TLP bonding as described elsewhere [5] a thin liquid interlayer is used to join two elements of base material together. The introduction of liquid phase can be achieved in numerous ways, including insertion of thin films with a low melting point between the working elements, and it's further melting. The liquid then fills the cavities formed by unevenness of the surface. With time diffusion of the elements across the liquid/solid interface leads to achieving localized element concentration, sufficient enough for creation of intermetallic phase. Those phases have higher melting point then the liquid, resulting in isothermal solidification. After solidification no traces of liquid phase should be seen, and the joint should become indistinguishable from other grain boundaries [5].

As mentioned above, attempts to join TiAl intermetallics have been done in the past [6-10], all of them using different forms of filler material for the interlayer, including Cu, Ni, Ti foils. Taking that into account this study investigates the possibility of TLP bonding of γTiAl elements, by using Al films. Al possesses a much lower melting temperature than these elements, which makes it a good choice for the interlayer material, while at the same time the Ti-Al phase diagram as seen in Fig. 1 shows us that diffusion of Al into the base material is expected to shift the phase composition in the area surrounding the joint into more Al rich Ti_xAl_y intermetallics, for example $TiAl_2$ or $TiAl_3$. The amount of Al introduced into the samples, must be small enough so that after homogenization, the overall shift in Al to Ti ratio does not the phase composition of the sample.

The experiments were conducted inside a Spark Plasma Sintering (SPS) furnace, which allowed for stable application of pressure and heat, as well as a protective vacuum atmosphere for the process.

Optical Microscopy (OM), Scanning Electron Microscopy (SEM), Energy-dispersive X-ray spectroscopy (EDS) and Electron backscatter diffraction (EBSD), were used to analyze the samples. The results and appropriate images are shown in the result section, while at the same time micro hardness tests were performed for comparison between the joint area of bonded samples and base γTiAl.

The goal of this study is to investigate the possibility of the application of thin Al films as liquid interlayer/bonding agents for joining of γTiAl elements, under different temperatures and film thickness, using the TLP bonding method.

2. Experimental

Small pieces cut from a γTiAl ingot were used as the base material for the bonding process. The samples were approximately 10 mm x 10 mm x 10 mm in size. Thin 99.5% purity Al films were used as the material for liquid interlayer creation. Both the bonding surface as well as the Al film surface was polished using 500 to 1200 mesh SiC abrasive paper, in order to remove the possible contaminations, then cleaned ultrasonically in ethanol, and dried out using a hot blower prior to the experiment.

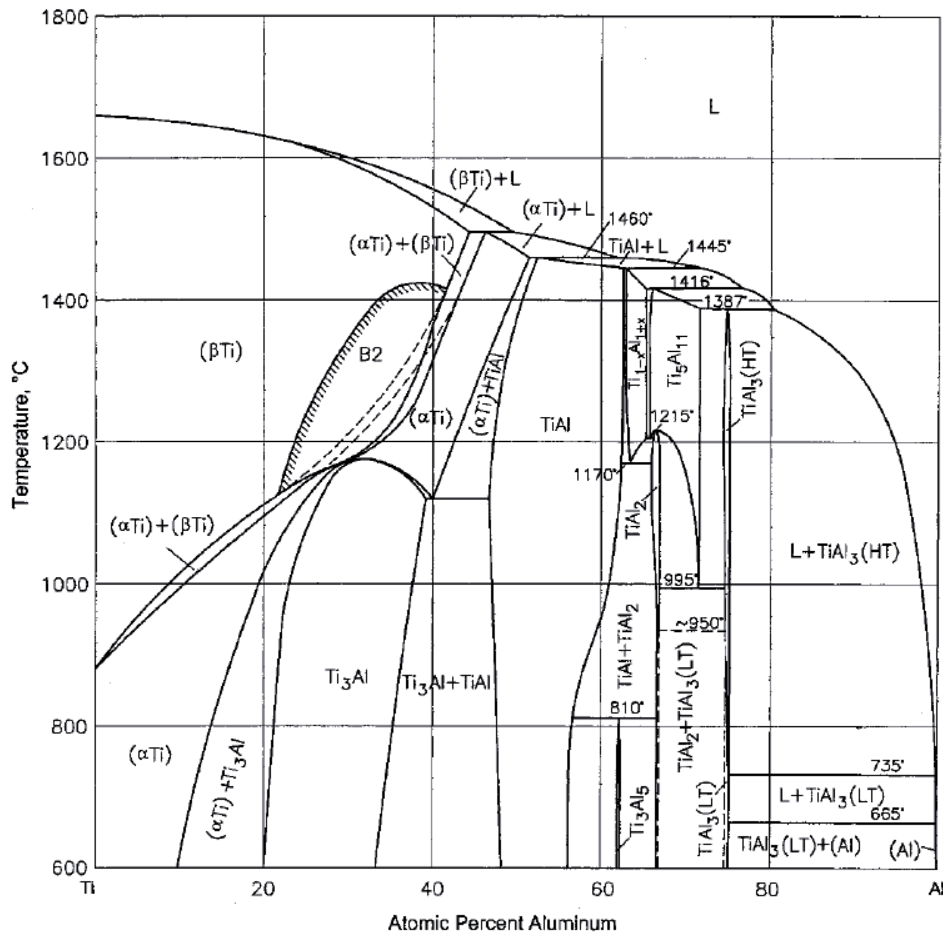

Fig. 1. Ti-Al phase diagram [11]

The samples were placed into a cylindrical graphite mold with an inner diameter of 20 mm and the Al film in between the γTiAl base elements. The remaining volume of the mold was filled with graphite powder and sealed using the upper and lower punches. The whole setup was then placed inside of a Dr. Sinter Lab Spark Plasma Sintering system, which was used to achieve bonding conditions. In a closed chamber vacuumed to 10 Pa, an uniaxial pressure of approximately 7.4 MPa was exerted on the samples via the punches, and the heating process started. The heating curve as seen in Fig. 2 consisted of two areas with different heating rates, the first one of about 60°C/min was used to reach the temperature of 600°C which was used to allow the temperature within the mold, and at detector to stabilize. After that a constant heating rate of 20°C/min was implemented in order to reach the bonding temperatures. The samples were then held at that temperature for 10 min, to allow homogenization to start. After that the samples were cooled down, cross-sectioned, polished using different abrasive papers and diamond dispersions up to 1/4 μm and analyzed.

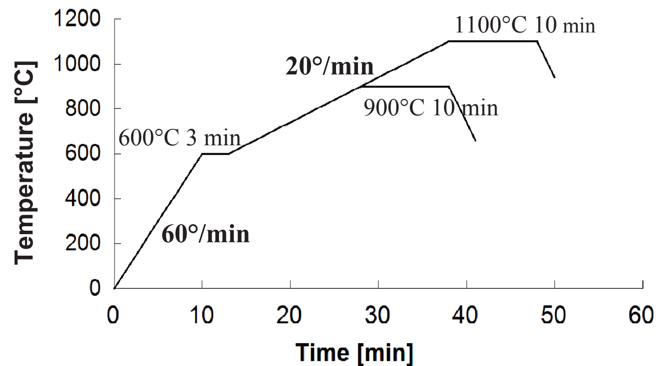

Fig. 2. Heating curve for the TLP bonding process

During the experimental process different initial film thicknesses of 50, 100, 200 and 300 μm were implemented in order to observe the difference in achieved joint microstructure. In addition two different bonding temperatures of 900 and 1100°C were used to determine the difference of bonding temperature on bond morphology.

The cross-sections of the joint were observed using VHX Digital Microscope Multi Scan system, and Keyence VH-Z500R RZx500-x5000 real zoom lens, in order to compare the microstructure of the joints, while the best set of sample was taken for SEM EDS and EBSD analysis using JEOL JIB-4600F/HKD Multi-Beam System to determine it's composition and microstructure. Vickers hardness test was performed on the samples using Future Tech microhardness tester Fm-300e with a load of 50 g to compare the hardness of the joint area to that of the base sample.

3. Results

The observation, and microstructure analysis of the samples achieved during the TLP bonding process, confirmed that a bond, joining the two base TiAl elements together was formed in all of the experimental setups. It was found that the initial Al film thickness plays a significant role in microstructure evolution, compared to the bonding temperature which however is also important.

In all samples we can easily notice the bonding area consisting of different phases. After further investigation it was discovered that the different phases present in the joint, are $TiAl_3$, $TiAl_2$, Ti_3Al, Ti_2AlC and TiC (USE subscript notation for the intermetallics). The existence of Ti contained in the phases in the interlayer, are associated with diffusion of Ti and possible surface melting. The multi-phase composition of the joint area occurs in all setups, although some differences can be found.

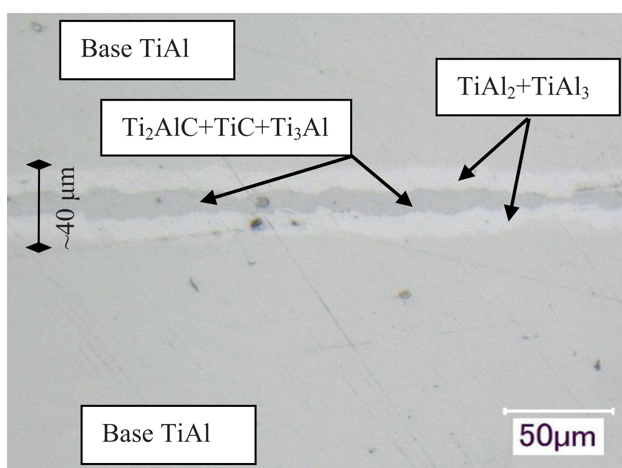

Fig. 3. Result of TLP bonding of TiAl with 50 μm thick Al film in temperature of 1100°C by SPS method

3.1. 50 μm thick Al film

In case of samples bonded at a temperature of 900°C the joint microstructure revealed existence of pores. This can be associated with small amount of Al in the interlayer, as well as low bonding temperature. During initial stages of TLP bonding, the interlayer reacts with the surface by solid state diffusion, that depletes the amount of Al, and when the liquid interlayer forms it's unable to fill in all the gaps in between the base elements [5,13]. Later during solidification, shrinkage increases the pore size.

In samples bonded at temperature of 1100°C, the porosity disappears, which is most likely due to higher wettability of liquid Al in higher temperatures, which results in it better filling the gaps in the bond. In case of both sample sets the microstructure closely resembles that seen in Fig. 3, with the bond being around 35-40 μm thick.

3.2. 100 μm thick Al film

For both 900°C and 1100°C bonding temperatures we can observe very similar microstructures, with creation of a non porous joint, and average thickness of 80-90 μm as seen in Fig. 4. The lack of pores can be associated with increased amount of Al during liquid stage. We also observed the difference in ratio of C and Al rich phases

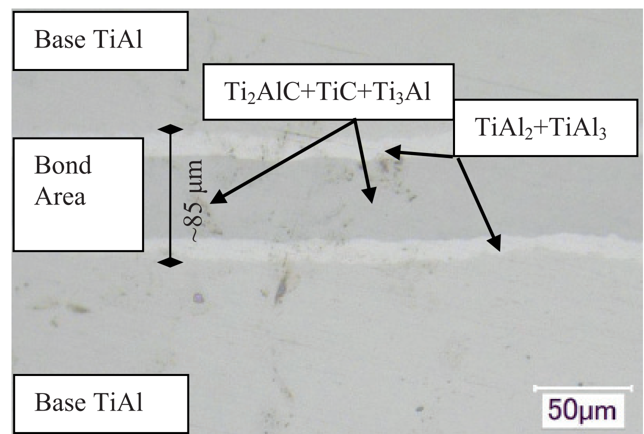

Fig. 4. Sample bonded with 100 μm thick Al film at 900°C

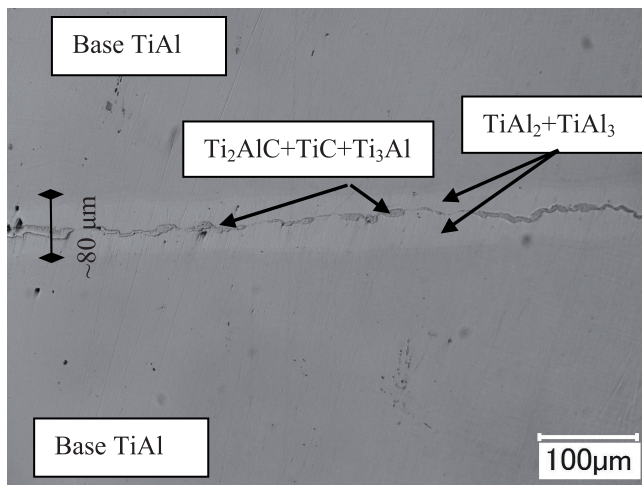

Fig. 5. Samples bonded with 200 μm thick Al film at 1100°C

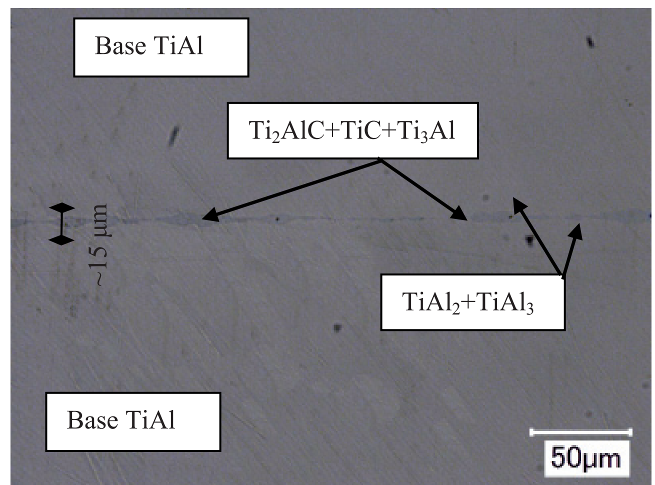

Fig. 6. Sample bonded with 300 μm thick Al film at 1100°C

in the joint, whit a much thicker carbon rich phase. That might be the result of increased amount of liquid Al reacting with the graphite filler. As described in [5,13] the solid state diffusion of Al film increases with its thickness, but at the same time the increased amount of liquid Al results in increased dissolution of graphite. The higher ratio of C rich phases in the microstructures, let us believe that those phases are produced easier then Al rich $TiAl_2$ and $TiAl_3$.

3.3. 200 μm thick Al film

Samples made with 200 μm thick Al film, have similar microstructures in case of both bonding temperatures of 1100 and 900°C. The image of the microstructure can be seen in Fig. 5. Compared to previously mentioned ones, we can observe a shift in the ratio of C and Al rich phases, resulting in Al rich phases being more dominant. The overall thickness of the joint did not increase, which is the consequence of some of the molten aluminum being pushed out from the interlayer due to pressure. However we can clearly see the dependence mentioned in previous paragraph, in which the solid state diffusion, resulting in creation of Al rich phases, increases with increase in initial thickness of Al film.

3.4. 300 μm thick Al foil

In case of these samples we can observe the biggest difference between the resulting joint structures. The samples made at 900°C follows the behavioral pattern of the previous samples, with thickness of approximately 90 μm, and a Al to C rich phase ratio similar to that of samples made with 200 μm Al foil. In case of samples bonded at 1100°C however, as seen in Fig. 6 the joint thickness is much smaller, close to 15-20 μm, while the existing Al rich phases are less noticeable, which suggests a closer composition to the base material. Its predicted that such behaviour is the result of strong solid state diffusion, as well as achieving optimal bonding conditions mentioned in [13].

3.5. SEM, EDS and EBSD

To further investigate the joint, microstructure and composition SEM, EDS and EBSD analysis were performed on the sample presented in Fig. 7. Due to very low detection rate of the elements even with very good polishing, the samples were etched using Kroll's reagent before the analysis. The results can be seen in Figs. 7-10.

The analysis made with SEM using the composition filter exposes the more complex phase structure of the joint, not seen using simple OM. An evident edge separates the base TiAl elements from the bond, however no porosity or defects were found in it. The existence of the edges is attributed to reaction of different phases with the etching reagent. Seeing as different phases are still present EDS analysis were conducted to determine the dominant elements throughout the joint, and it was confirmed that the 3 main elements in the bond were Ti, Al and C. The spacial distribution mapping of the elements is shown in Fig. 8.

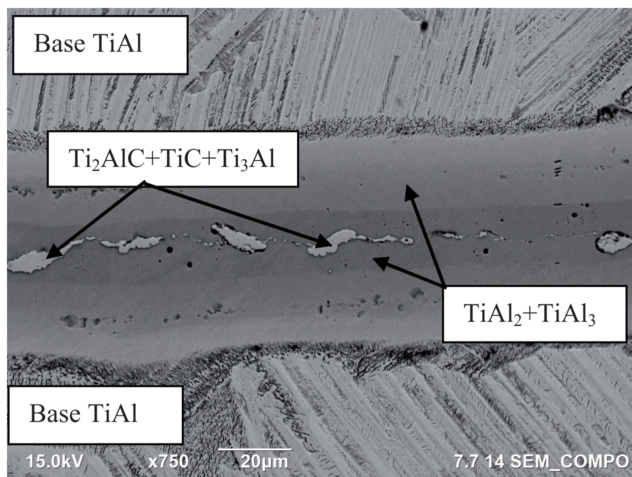

Fig. 7. SEM analysis of joint area for samples made with 300 µm Al film and 1100°C

After analyzing the element mappings in Fig. 8 we can see that the area of the join contains large amounts of Al whit small areas in the very center filled with almost only Ti and C. The existence of carbon can be attributed to dissolution of graphite i the molten aluminum, while the presence of Ti, is the effect of diffusion or surface melting of the base elements. In order to determine if the carbon present in the joint is in elemental state or forms compounds, EBSD analysis was performed on the samples.

As can be seen in Fig. 9 the carbon within the bond area has joined with Ti and Al to form different phases, including TiC and Ti_2AlC. As already mentioned the liquid Al is in direct connection to the graphite, and as it dissolutes, it's distributed thorough the length of the joint due to it wanting to achieve equilibrium. The creation of Ti_2AlC and TiC was not anticipated as both of them typically require higher temperature to form [14-16]. It's also important to notice that no titanium was present in the interlayer area at the start of the process, while after the bonding process we do notice different Ti compounds. That would suggest that either Ti has diffused into the liquid interlayer and started forming different compounds, or some of the surface of the TiAl sample was melted during the process and thus introduced titanium into the interlayer. While the second idea is possible, it is more plausible to assume that its the diffusion of Ti that leads to this result.

While the amount and complexity of compounds found in the joint area confirm the diffusion of Ti into the interlayer, at the same time Fig. 10, shows us presence of $TiAl_3$ within the base material. That is a strong hint confirming the diffusion of Al from the liquid interlayer into the base material.

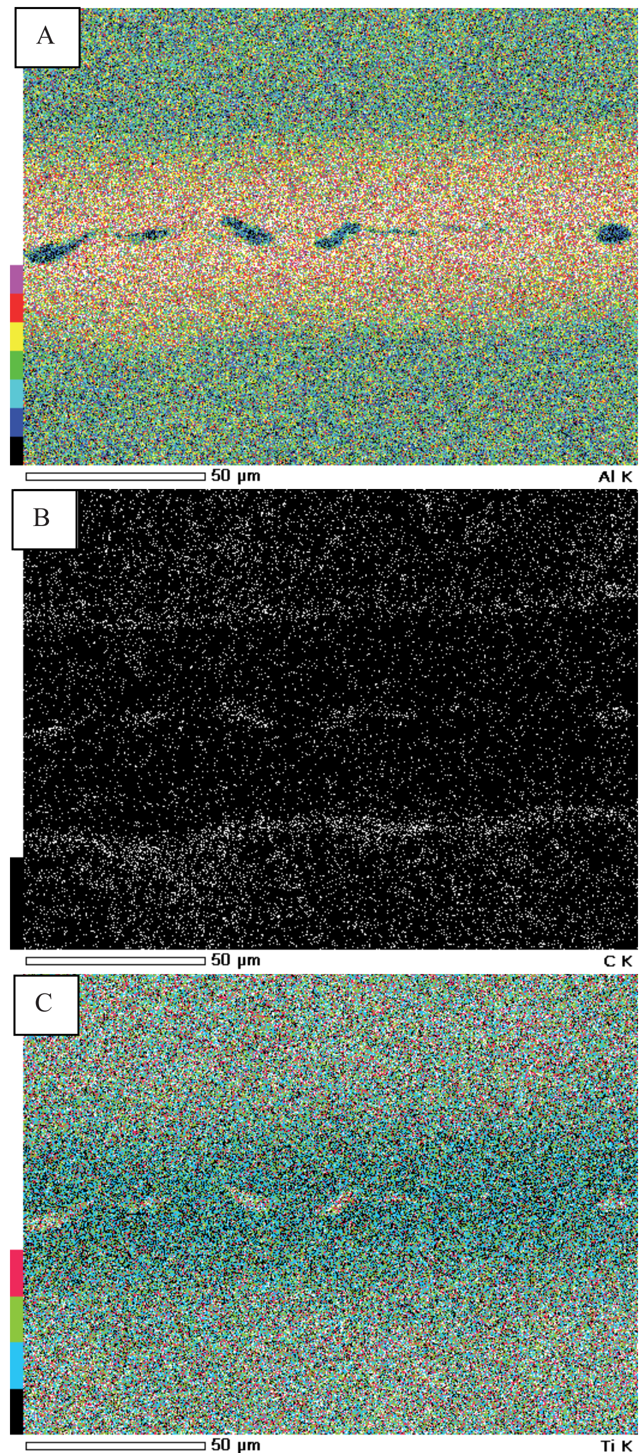

Fig. 8. EDS analysis of the joint area in a sample made with a 300 µm Al film and 1100°C: A, B and C shows spacial distributions of Al, C and Ti, respectively. The scale bar is 50 µm, and the left side bar, represents the presence of given element, with darker color representing less

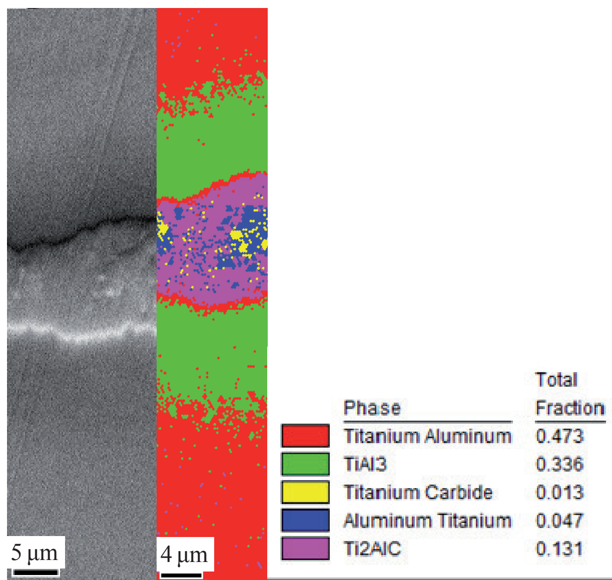

Fig. 9. EBSD analysis of sample made with a 300 µm-Al film and 1100°C. Titanium Aluminum represents TiAl, Titanium Carbide represents TiC, and Aluminum Titanium represents Ti_3Al

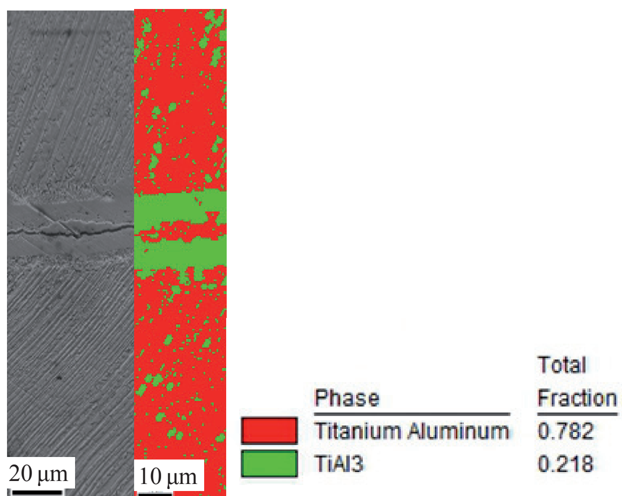

Fig. 10. EBSD analysis of sample made with a 300 µm Al film and 1100°C, showing diffusion of Al into the base sample. Titanium Aluminum represents TiAl

From the previously mentioned results we can know that the middle part in Fig. 10 contains TiC and Ti_2AlC, while like in other examples the outer green part of the joint consists of $TiAl_3$. The $TiAl_3$ spots seen in the base elements, are most likely due to diffusion of Al into the TiAl, along the diffusion paths, or due to already created $TiAl_3$ particles precipitation. The discontinuous distribution of the spots is most likely the effect of them forming in the most energy favorable positions.

3.6. Vickers hardness

In addition to all that, Vickers hardness tests were performed on the joint area in most of the sample with 20+ measurements for each one. The results are shown in Fig. 11. As seen on the graph, the hardness of the joint area differs depending on the bonding condition of the sample. The increase in hardness for samples made with low film thickness, is the result of formation of hard phases, like $TiAl_3$, Ti_2AlC and TiC [14,17,18]. However in case of samples made with higher film thickness, where as already mentioned, solid state diffusion was stronger, we can observe that the hardness is getting closer to that of the base material.

Low hardness of samples made with a 100 µm-thick Al film at 900°C is the result of porosity in the joint. In addition to that the parallel line on the graph represents the hardness of the gamma TiAl. During the measurements it was discovered that the hardness of sample made with a 300 µm Al film at 1100°C is close to that of the base material. It is probably the effect of localized similarity of the joint to the base material.

4. Conclusions

The results of the OM analysis of the joint microstructure show us that bonding of γTiAl can be performed using TLP method with Al films as interlayers. Use of SPS furnace to apply pressure and temperature to the sample proved good. However the typical setup which included the graphite powder as filler material in the mold introduced contamination in form of carbide phases in the joint. Future experiments will be conducted in which the graphite powder will be replaced. The results of the SEM, EDS and EBSD analyses confirm the existence of different TiAl intermetallic phases within the joint, and even that of ceramic and cermet materials such as TiC and Ti_2AlC. The presence of those material results in a change of the mechanical properties of the joint, as was presented by the Vickers hardness test, however it could also be noted that some samples, hardness similar to that of base material could be achieved. Conducted experiments allow us to determine the proper TLP bonding parameters for joining of TiAl elements using Al films.

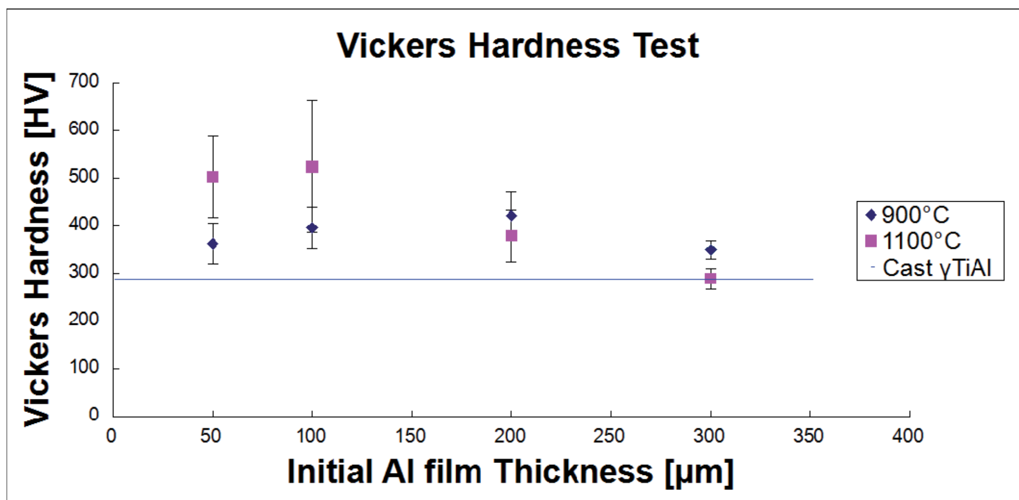

Fig. 11. Results of hardness tests, made on the joint area of the bonded samples and the base material. The line represents the hardness of base material, while the other dots the hardness of analyzed joints

Those are, film thickness of ~300 µm, bonding temperature of 1100°C, pressure of 7.4 MPa and protective vacuum atmosphere. EDS and EBSD analysis suggests active diffusion of both Ti and Al elements inside the bonding area, which is a good indicator for possible homogenization.

In conclusion we can say that TLP bonding of γTiAl using Al films by SPS method shows potential, for joining of the base elements, with creation of the bond even with low temperature and film thickness, as well as confirmed possibility of homogenization through diffusion of Ti and Al.

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