Possibilities of using interlayers during diffusion welding of Ti Gr2 and AISI 316L

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Abstract. Joining of materials with different physical and mechanical properties is always very problematic. As one of the possibilities, there can be used method of joint creation in solid state – e.g. by the diffusion welding. With this method it is possible to joint very different materials – even metals with non-metals. Despite the unquestionable advantages of this method, there is necessary to take into account also its limits. Among of them, there is creation of brittle intermetallic phases on the boundary of materials. This occurs e.g. at creation heterogeneous joints between titanium and highly-alloyed austenitic steel. The major aim of this paper is to describe how it is possible to eliminate brittle intermetallic phases created on boundary between Ti Grade 2 and AISI 316L steel by using metal interlayers.

Keywords: Diffusion Welding, Interlayers, Titanium Grade 2, AISI 316L steel, Heterogeneous Welds

1 Introduction

Diffusion welding is fairly quite a new technology in the research field. It has numerous applications in distinct areas such as aerospace, nuclear, automotive, electronics and sensor industries. This has led to increasing attention for the researcher to develop new and advanced materials.

Diffusion, as every transport effect, can be described by two aspects. Phenomenological diffusion theory deals with the overall balance of transport process in solid substance undertaken in the given conditions. This theory takes into account only macroscopic perspective. Microscopic perspective is considered in the atomic diffusion theory. This theory respects internal composition of material including crystal lattice interferences. In the case of non-existing motive power on atoms, their jumps are random. However these jumps occur in specific crystallographic equivalent directions.

The diffusion process can be divided into two types: self-diffusion and hetero-diffusion. Self-diffusion takes place in pure metal. During self-diffusion, atoms migrate randomly throughout the crystal lattice. Due to the random movement of atoms in the grid, concentration gradient arises in crystal lattice but mass of pure metal isn't changed. Concentration gradient can be expressed by Fick's laws when the Fick's first law (eq. 1)

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indicates a variation of concentration in the x-direction per unit of time. The diffusion flux J [$\text{mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$] of atoms per unit time and unit area in the direction of the x-axis is proportional to the concentration gradient. Mathematically, it is expressed as:

$$J = -D \cdot \left(\frac{\partial C}{\partial x} \right) \quad (1)$$

Where D [$\text{m}^2 \cdot \text{s}^{-1}$] is diffusion coefficient, C [$\text{mol} \cdot \text{m}^{-3}$] is concentration of elements, x [m] is diffusion direction and t [s] is time. In most practical cases, diffusion processes are non-stationary ones, because diffusion processes are changing over time. It can be deduced as relation called Fick's second law (eq. 2). In mathematical form, it is represented as:

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x} \left(D \cdot \frac{\partial C}{\partial x} \right) = D \cdot \frac{\partial^2 C}{\partial x^2} \quad (2)$$

Hetero-diffusion occurs between two phases of the material. It is more complex phenomenon and possible only if atom has sufficient amount of energy to displace from one position to another one. When the atom moves in the grid, it creates a hole or vacancy in its node.[1,2]

2 Possibilities to joint Ti Gr2 and AISI 316L without interlayers

There are several methods how to create heterogeneous joints between titanium and highly-alloyed stainless steel. As the 1st possibility there is fusion welding, because both materials have similar magnitudes of melting temperatures. For own jointing can be used e.g. TIG method (however with precise shielding of weld pool), eventually electron-beam welding. However, in both cases takes place immixture of both materials at creation different intermetallic compounds (IMCs) in the weld metal. Mostly there are phases as TiFe and TiFe_2 that are typically of their high hardness and brittleness [3]. So as the aim of the welding process there is tendency to apply processes, which can eliminate quantity of areas, where are these IMCs created. Diffusion welding seems to be quite suitable possibility welding technology, because area of mutual diffusion of elements varies here just within the few tens of micrometers.

Nevertheless, even there is very narrow diffusion area, IMCs are also created within it and hence there is important increase of hardness values as can be seen from Fig. 1 (measurement of hardness $\text{HV}_{0.02}$). Due to that, ductility of created joint is very low. So as a solution, there is utilization of interlayers from elements, which don't create hard and brittle IMCs with titanium. Their purpose is to create with both materials diffusion joint, but also to prevent mutual diffusion of Ti and Fe at the same time.

Fig. 1 Measurement of micro-hardness $\text{HV}_{0.02}$ over the diffusion area [3]

3 Possibilities to joint Ti Gr2 and AISI 316L by the interlayers

Based upon the performed *recherche* there was determined that for diffusion bonding of titanium and highly alloyed austenitic steels it is possible to use Ni, Ag or eventually Cu interlayer. Together with titanium, these elements create compounds that are tough enough and thus suitable to remove abnormal brittleness in the joint area. In all cases monitored in *recherche* was for the diffusion welding used thermal-mechanical simulator Gleeble and that is why the same device was also used for the experimental verification of the individual interlayers properties.

As the first interlayer used for the bonding there was used the Nickel one. From the research paper of Chatterjee and Kundu [4], it is investigated that the diffusion bonding between commercially pure Ti Gr 2 and AISI 304 steel with the help of Ni interlayer reveals good result on final welded joint in view of ductility. They used bonding temperature within range from 850 up to 950°C, but applied pressure wasn't mentioned in the article. Also Fang-Li Wang [5] and his colleagues used diffusion bonding between commercially pure Ti Gr 2 and AISI 304 steel with the help of Ni interlayer. They used impulse pressuring in range from 8 up to 20 MPa with duration of 60 up to 150 sec at temperature 850°C.

During the experimental verification of Ni interlayer application suitability was determined that maximal available pressure at temperature 870°C is 3.2 MPa. Then there is very rapid deformation on the side of pure titanium, which is unacceptable from the industrial utilization point of view. At temperature 900°C is maximal available pressure lower than 1 MPa and at temperature 850°C must be pressure below 6.5 MPa.

At diffusion welding in vacuum and using device Gleeble 3500 there was determined that at temperature 870°C and pressure 3 MPa is not bonding between Ni interlayer on one side and Ti + AISI 316L on the other side sufficient enough - even after keeping holding time 50 min at used temperature. Magnitude of deformation for these samples after 50 min achieved only 0.182 mm. Moreover, there was experimentally determined that diffusion bonding of sufficient quality with Ni interlayer is created not below temperature 1050°C, pressure 4.5 MPa and holding time 20 min. Nevertheless, this was possible to be verified only for steel AISI 316L, which can resist such temperature and pressure without any problems. The total deformation of sample under these parameters didn't exceed 0.2 mm.

By utilization of the impulse pressuring diffusion welding acc. to Fang-Li Wang [5] parameters it is also possible to achieve diffusion bonding of high quality. However, it is necessary to take into account deformation over 15% on the side of pure titanium, which is more than impracticable from the light of industrial utilization.

As the second interlayer used for the bonding Ti Gr2 and AISI 316L there was used the Ag interlayer. From the research paper of Yongqiang Deng and his colleagues [6] it is reported that using of Ag interlayer exhibits good result in light of micro-hardness distribution of bonded joint. Yongqiang Deng used device Gleeble and experiments were performed at temperatures 825, 850, 870°C and pressure 6 MPa. At experimental verification of Ag interlayer application suitability, there was carried out experiment at temperature 850°C, because at temperature 875°C and pressure 6 MPa would be again presented a very intense deformation of Ti Gr2.

Fig. 2 reveals SEM image with EDS scanning line of sample. Several distinct layers from left to right side (Ti/Ti s.s./TiAg/Ag/SS) were observed. Titanium solid solution (abb. Ti s.s) 19.55 μm , Ti-Ag IMC (TiAg) 28.33 μm and Ag interlayer 82.62 μm thicknesses were measured in the joint interface. From EDS analysis is evident that during holding time on temperature there occurs very intense diffusion between Ti and Ag. However, on the side of AISI 316L is diffusion minimal. Yongqiang Deng states that acc. to Ag-Fe/Cr/Ni binary diagrams [7], Ag cannot form IMCs with the main elements in SS. That is true, but there should be also state that solubility of Ag in Fe, Cr and Ni in solid state is almost on zero value. So it means that diffusion processes probably will not take place, which is also

confirmed by result of EDS analysis. Acc. to binary diagram Ag-Fe is solubility of Ag in iron at temperature 850°C lower than 0.01 wt%. Thus the bonding between surfaces is created only due to adhesion.

Fig. 2 SEM image and corresponding EDS scanning line of results for the bonded joint with Ag interlayer (Gleeble - 850°C; 6 MPa; 20 min)

To verify such presumption, there was performed experiment where samples Ti-Ag-SS were firstly diffusively bonded at device Gleeble 3500 at temperature 850°C, pressure 6 MPa and holding time on temperature 20 min. After cooling and unloading from Gleeble, samples were inserted into vacuum furnace for holding time of 10 hrs at temperature 850°C. The aim of experiment was to verify, whether there would be at least partial diffusion of Ag into material AISI 316L. Moreover, there was effort to determine diffusion rate of Ag into titanium. Results of experiments are shown in Fig. 3. It is obvious that high holding time at temperature 850°C resulted in fractures on the TiAg-Ag interface. They are (despite the high ductility of Ag interlayer) probably caused by differences between thermal expansion coefficients for titanium and silver. Furthermore, there was proved that even under 10-times extension of holding time, there isn't diffusion of Ag interlayer into steel AISI 316L. Thus there's no need to find or optimize diffusion welding process parameters for Ag interlayer.

Fig. 3 SEM image with EDS scanning line scanning of results for the bonded joint with Ag interlayer (Gleeble - 850°C; 6 MPa; 20 min) and 10 hrs in vacuum furnace at temperature 850°C

As another recommended type of interlayer for combination Ti-SS there is Cu interlayer. This problem is solved e.g. by S. Kundu and his colleagues at bonding the commercially pure Ti Gr 2 and AISI 304 steel. Cu interlayer reveals good result on final welded joint in view of ductility [8]. Therefore, it is anticipated that Cu can be estimated as a potential candidate to be used as interlayer to improve the joint ductility. S. Kundu performed the diffusion welding with Cu interlayer for temperatures 850 up to 950°C, pressure 3 MPa and holding time 90 min. Based upon the experiments, he states that at temperatures 900°C and higher, Cu interlayer can't stop diffusion of Fe, Cr and Ni into Ti and reversely Ti into SS. That is why there are created brittle IMC's in diffusion area, which have a crucial influence on increase of hardness and brittleness in the joint area. Because of that, there was decided to carry out experiment with Cu interlayer at temperature 850°C, pressure 3 MPa and holding times 30, 60 and 90 min.

There was only a partial diffusion and thus insufficient joint strength at temperature 850°C and holding time 30 min. This result is shown in Fig. 4. There was joint failure in the Cu interlayer, when part of interlayer remained on the SS part (Fig. 4 – left) and part on the titanium part.

Fig. 4 Failure of Cu interlayer in sample SS-Cu-Ti after diffusion welding (850°C; 3 MPa; 30 min)

As it can be seen from Fig. 5 (acquired by the optical microscope Olympus DSX 500) there was not achieve full diffusion bonding even after extension of holding time to 90 min. Nevertheless, from the measurement of micro-hardness HV0.1 it is clear that in the weld joint are not created hard and brittle IMC's and total hardness HV0.1 didn't exceed magnitude 265 HV. To create full diffusion welding through the whole cross-section, there is necessary to extend diffusion time or increase diffusion activation energy by higher temperature. Based upon these facts, there was decided to apply another holding time at temperature 850°C in the vacuum furnace for sample diffusively welded by device Gleeble (850°C, 3 MPa, 90 min). To be specific, holding time at temperature 850°C was now 5 hrs.

Fig. 5 Results of measurement micro-hardness HV0.1 on the sample with Cu interlayer (Gleeble - 850°C; 3 MPa; 90 min)

From the result shown in Fig. 6, it is obvious that total diffusion time 6.5 hrs at temperature 850°C is sufficient to create quality diffusion bonding through the whole cross-section both on the side of Ti and on the side of SS. However, used diffusion time was too long, because there is diffusion of Ti through the whole thickness of Cu interlayer 0.125 mm into SS and simultaneously there is diffusion of Fe, Cr and Ni into Ti. Due to this fact there were created brittle IMC's, which not only increased micro-hardness HV0.1 up to 418 HV (see Fig. 7), but also caused fractures creation on the boundary with steel AISI 316L.

Fig. 6 SEM image and relevant EDS scanning line of results for the bonded joint with Cu interlayer (Gleeble - 850°C; 3 MPa; 20 min, vacuum furnace 850°C; 300 min)

Fig. 7 Results of measurement micro-hardness HV0.1 on the sample with Cu interlayer (Gleeble - 850°C; 3 MPa; 20 min, vacuum furnace 850°C; 300 min)

4 Conclusion

Diffusion welding is quite progressive technology at production heterogeneous welds created in solid state. On the other hand, this technology has also its limitations - mainly at welding materials, which create brittle IMCs on the materials boundary during diffusion. As example of such joint there is joint Ti-Fe – eventually with other elements as Cr, Al, etc.

One of the possibilities how to eliminate joint brittleness at diffusion welding there is usage of the metal interlayers. These interlayers have several objectives. 1st of them is to create transient layer, which prevent connection of elements that create hard and brittle IMCs during diffusion. Therewithal it should be valid for these interlayers that they have at least

partial solubility in solid state with the welding materials and also that they do not create own brittle IMCs with these materials and their major alloying elements.

In the paper were described possibilities to use 3 types of metal interlayers (Ni, Ag, Cu) at diffusion welding of pure titanium Gr 2 and austenitic highly alloyed steel AISI 316L. In light of requirements pose on these interlayers it seems that Ni interlayer is the most suitable one. Nickel has very high diffusion resistance in comparison to majority of basic alloying elements. Thus there is diffusion only in very narrow surface layer of Ni and due to the low diffusion rate, there aren't generally created intermetallic phase Ti_2Ni , which creates up to 40 wt% of Ti in Ni. On the other hand, nickel interlayer needs higher magnitudes of activation energy given by higher temperature that should be at least 1000°C for diffusion. However, titanium Gr 2 reveals very high amount of deformation already at 900°C and pressure 1 MPa. That is why it would be probably better to use this interlayer in alloyed alloys of Ti having higher strength under high temperatures.

The 2nd tested interlayer was Ag one. This interlayer creates with Ti solid solution whose hardness doesn't exceed pure Ti hardness. However, the problem about utilization of such Ag interlayer is on the side of AISI 316L, because solubility of Ag with Fe, Cr and Ni is almost on zero value at temperatures up to 1000°C. That is why on the boundary of these materials isn't created any bonding or just adhesive bonding.

The last tested interlayer was Cu one. Copper has very good solubility in solid state both with Ti and also with major alloying elements of steel AISI 316L. Nevertheless, problem here is to determine proper holding time on temperature so that there is sufficient diffusion through the whole cross-section of bonded sample and at the same time to prevent diffusion of Ti, Fe, Cr and Ni through the whole Cu interlayer thickness. However, in this case can be possible to apply suitable technological precautions as can be e.g. to increase Cu interlayer thickness at least to range from 0.25 up to 0.3 mm.

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