

# Research on Interlayer Alloys for Transient Liquid Phase Diffusion Bonding of Single Crystal Nickel Base Superalloy DD6

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# Abstract

Transient Liquid Phase Diffusion bonding (TLP bonding) is an effective method to achieve excellent joint of DD6, which is a new generation single crystal superalloy to manufacture aero-engine turbine blades. In this paper, the interlayer alloys for DD6 TLP bonding were designed. The alloy foils with thickness 40  $\mu$ m ~ 60  $\mu$ m, width 4 mm were prepared by using a single roller rapid solidification apparatus and the TLP bonding of DD6 was conducted. Then the joint microstructure and alloying elements diffusion behaviors were analyzed. The results indicate that microstructures of interlayer alloys prepared are fine and homogeneous, the melting point range of alloys from 1070°C to 1074°C and their melting temperature interval is merely 20°C, when the chemical composition of alloys are 1.5 ~ 2.0Cr, 3.2 ~ 4.0W, 3.7 ~ 4.5Co, 2.2 ~ 3.0Al, 0.7 ~ 1.0Mo, 3.2B, remain Ni (wt%). When the welding parameters are bonding temperature 1200°C, holding time 8.0 hour and welding pressure 0.3 MPa, the compacted joints obtained and the microstructure of TLP bonding seams were similar to base metal. The bonding joint is composed of weld center zone, isothermal solidification zone and diffusion-affected zone. Within joint, the elements diffusion is sufficient and borides in the diffusion zone are fewer.

# **Keywords**

Interlayer Alloy, DD6 Single Crystal Superalloy, TLP Bonding, Microstructure of Joint

# **1. Introduction**

Ni-based single crystal super alloys are key materials to manufacture aero-engine turbine blades owing to their fine thermostability, heat resistance and high-temperature structural stability. Considering thermo dissipation

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and energy saving, aero-engine turbine blades are always designed in complex hollow structures, and it's inevitably involves welding problems [1] [2] with during the manufacturing process of that hollow structures. Since the melting weld seam is apt to suffer from hot cracking and the brazed joining intensity is always insufficient, Transient Liquid Phase Diffusion (TLP) Bonding is an appropriate method to carry out the welding of Ni-based single crystal super alloys [3]. Except the process factors, the interlayer alloy's chemical components as well as its applied forms are closely related to the welding quality and joint microstructures of TLP bonding DD6 alloy [4] [5]. So according to the chemical composition of DD6 alloy and the requirement of property matching, in this paper, the composition of the interlayer alloy is designed and the alloy foils is prepared, which is used for DD6 TLP bonding, thus the excellent quality bonding joint of DD6 single crystal alloys would be obtained.

# 2. Design of Interlayer Alloys

In order to realize the TLP bonding of DD6 single crystal alloys, there are several sound principles to follow. First, assuring that the joint strength matching to the base metals, the main alloying elements of interlayer alloys must be as much as possible approximate to the parent metal [6]. Secondly, insuring that the welding process go without a hitch, the lower melting point elements and easy to diffusion elements would be added [7].

## 2.1. Selection of Lower Melting Point Elements

The grains of superalloy DD6 to begin coarsening at 1320°C, therefore, TLP bonding should be conducted lower than that temperature. References [8] [9] show that the appropriate TLP bonding temperature of single crystal alloy DD6 is perforce controlled in less than 1200°C.

Due to the fact that the TLP bonding temperature should be higher than that of the interlayer alloy's melting point  $100^{\circ}C \sim 150^{\circ}C$ , the applicable melting temperature of the interlayer alloy should be  $1050^{\circ}C \sim 1100^{\circ}C$ .

On the basis of the chemical composition of base metal, the interlayer alloys must contain some certain elements in order to bring about eutectic action with matrix metal Ni, so the melting point of interlayer alloy lowered. B, Si, Nb and Mn *et al.* are the alternative alloying elements to lower the melting point of interlayer alloys used to TLP bonding DD6 alloy. Among them, B has the most significant effect to drop interlayer alloy's melting point, due to B atom has the smaller radius with easy to diffuse conveniently and is able to react with Ni form eutectic with lower melting temperature. Whereby B is selected as main additive element to interlayer alloys to lower their melting points and the additive amount is set nearby to the Ni-B eutectic point. Ni-B binary phases diagram is shown in Figure 1.

#### 2.2. Selection of the Diffuse Element

The essence of transient liquid phase diffusion bonding is elements' diffusion. During the TLP bonding process, with the temperature rising, the alloys at interface melt and the elements to lower the melting point diffuse toward the base metal rapidly, causing the melting point of base metal at the interface lower and the base metal melt, so the liquid phase area become wider. When the density of the elements to lower the melting point at the liquid/solid interface lower to the liquidus temperature, with the elements to lower the melting point diffusing further, the melting point of the liquid phase area at the liquid/solid interface rise, the liquid phase area begin to solidify from the interface to the center of welding seam. Hence, elements' diffusion must be made as precondition for successful TLP bonding and the interlayer alloys' elements to lower the melting point must possess the



**Figure 1.** Ni-B binary phase diagram [10].

character of diffusing rapidly. Generally speaking, diffuse elements are B and Si. B atom has small radius and high speed of diffusion. Besides lowering the melting point, B is the main diffuse element.

## 2.3. Selection of Main Alloying Elements

In order to obtain the joint whose structure and performance can be matched with the base metal, the kind of the elements which the interlayer alloys contain should be similar or close to that of the base metal's. The interlayer alloys for DD6 TLP bonding contain W, Cr, Co, Mo, Al as well as other alloy elements. Additionally, the interaction between the alloying elements and the elements to lower the melting point and diffuse should also be considered when determine the content of the alloying elements. If the main alloying elements can react with the elements to lower the melting point and diffuse into brittle compounds with high melting point, the joint mechanical properties will reduce dramatically so that the content of this element is unfavorable and overmuch. Contrary to that, the elements can be added appropriately. Whether the alloying elements can generate borides or not and the stability of the borides is the key to determining the content of alloying elements.

The criteria for judging the stability of the borides are:

1) According to the phase diagram, the lower the eutectic temperature of the alloys' elements and B is, the easier B melt and avoid generating the borides with high melting point. Therefore, the higher the eutectic temperature of the alloys' elements is, the less the content of the alloys' elements is.

From Figure 2, the Cr-B binary phases, it is found that the lowest temperature when the eutectic action of Cr-B take place is 1630°C and the reaction temperature is high, then the melting point of the generated  $Cr_2B$  is very high. Accordingly, the eutectic temperature of Co-B is 1100°C ~ 1350°C and the melting temperature of the generated borides is lower, shown in Figure 3.

2) According to the differential value of elements electronegativity, the lower the differential value of elements electronegativity of the alloys' elements and B is, their bond is weaker, the easier the generated borides resolve. However, the higher the differential value of elements electronegativity of the alloys' elements and B is, the more difficult the generated borides resolve, so the content of the alloys' elements should be controlled in lower range. The differential value of elements electronegativity of the alloys' elements and B is shown in **Table 1**. It can be seen from **Table 1** that the differential value of elements electronegativity of the alloys' elements and B is shown in **Table 1**. It can be seen from **Table 1** that the differential value of elements electronegativity of the alloys' elements and B is shown in **Table 1**. It can be seen from **Table 1** that the differential value of elements electronegativity of the alloys' elements and B is shown in **Table 1**. It can be seen from **Table 1** that the differential value of elements electronegativity of the alloys' elements and B is shown in **Table 1**. It can be seen from **Table 1** that the differential value of elements electronegativity of the alloys' elements and B from high to low can be ranged as Al > Cr > W > Co > Mo.

Referring to the chemical composition of the DD6 superalloy and considering the eutectic temperature and the



Figure 3. Co-B binary phase diagram [10].

Table 1. The electronegativity of elements.								
Alloys' elements X	The electronegativity of X	The electronegativity of B	The differential value of elements electronegativity of X and B					
W	2.36	2.04	0.32					
Al	1.61	2.04	0.43					
Cr	1.66	2.04	0.38					
Со	1.88	2.04	0.16					
Мо	2.16	2.04	0.12					

differential value of elements electronegativity's influence on the stability of the borides, the composition range of the interlayer alloy is designed as:  $1.0 \le Cr \le 2.0$ ,  $2.5 \le W \le 4.0$ ,  $3.0 \le Co \le 4.5$ ,  $1.5 \le Al \le 3.0$ ,  $0.5 \le Mo \le 1.0$ ,  $3.0 \le B \le 3.6$  and remain Ni (wt%). For easier research, the interlayer alloys with the composition above were divided into two groups for testing, shown in Table 2.

## 3. Preparation of the Interlayer Alloys

## 3.1. Preparation of the Interlayer Alloy Foils

The interlayer alloys' materials for testing are simple metals (99.9%): Ni, Al, W, Cr, Mo, Co and B. First, purify the alloys with  $B_2O_3$ . Then melt the interlayer alloys purified and B needed with high frequency induction heating equipment. As B cannot be dissolved completely when melt the interlayer alloys and B, the alloys should be weighted before melting and melt repeatedly, until the ratio of B and the alloy meets the requirement. Finally, The alloy foils with thickness 40  $\mu$ m ~ 60  $\mu$ m, width 4 mm were prepared by using a single roller rapid solidification apparatus. The testing parameters are shown in Reference [11].

# 3.2. Analysis of the Interlayer Alloys Foils' Microstructure

The homogeneity of interlayer alloys' composition has great effect on the welding result. **Figure 4** shows the microstructure of the interlayer alloys foils along thickness, from which it can be seen that the composition of the interlayer alloy foils prepared is homogeneous and the microstructure of it is fine meeting the requirement for interlayer alloys for TLP welding.

#### **3.3. DTA Analysis for Interlayer Alloy Foils**

**Figure 5** shows the DTA curve of the interlayer alloys, it can be seen that the crystallization of interlayer alloys starts at 454°C and finish at 466°C showing a crystallization peak. With the temperature rising continuously, alloy foils begin to melt when it is up to 1074°C and finish at 1094°C. Their melting temperature interval is about 20°C. The melting temperature of the interlayer with small melting temperature interval ranges from 1050°C to 1100°C meeting the operating requirement for DD6 TLP welding.

# 4. DD6 TLP Bonding

# 4.1. Welding Method and Process

## 4.1.1. Material for Testing

The base metal for testing is as-cast DD6 with dimension of  $\Phi 16 \times 7$  mm and its chemical composition is: 4.34Cr, 9.01Co, 1.98Mo, 8.44W, 6.14Ta, 5.4Al, 3.02Re, 0.98Nb, 0.12Hf, remain Ni (wt%).

#### 4.1.2. Weldment Assemble

Before welding, remove the oxide film on the surface of the prepared base metal mechanically and clear the weldment in the acetone under ultrasonic vibration, then assemble them according to **Figure 6**. Particular way is: first, put several interlayer alloy foils with width about 4 mm between the end face for welding of the two weldments parallelly and horizontally. Then, fix the weldments in heat-resistant ceramic moulds, which are put in vacuum furnace. Finally, press weights on the assembled weldments with welding pressures 0.3 MPa on.

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No	The chemical composition of interlayer alloys (wt%)							
NO. —	Cr	W	Со	Al	Мо	В	Ni	
1	$1.0 \sim 1.5$	2.5 ~ 3.2	3.0 ~ 3.7	1.5 ~ 2.2	$0.5 \sim 0.7$	3.2	balance	
2	$1.5 \sim 2.0$	$3.2 \sim 4.0$	3.7 ~ 4.5	2.2 ~ 3.0	$0.7 \sim 1.0$	3.2	balance	



Figure 4. Microstructure of interlayer alloy.



Figure 5. The DTA curve of interlayer alloy.



#### 4.1.3. Welding Process

TLP bonding parameters are: welding temperature 1200°C, holding time 8 h, welding pressures 0.3 MPa, vacuum degree  $5 \times 10^{-2}$  Pa. Heating specifications are: heating to 1000°C at the heating rate of 10°C/min, holding time 30 min; welding temperature 1200°C, holding time 8h,cooling down to room temperature within the furnace.

## 4.2. Analysis of Joints' Microstructure

By standard metallographic techniques, the metallographic specimens of the welding joints' cross-section were prepared in the welding bars' longitudinal direction. The surface of joints should be etched with reagent (Compositions: 5 ml HNO<sub>3</sub> + 15 ml HCL) for about 3 s. DTA analysis the interlayer alloys with CRY-2P Differential Thermal Analyzer. Study the microstructure of the joints with JSM-6700F scanning electron microscope (SEM)

Table 2. The chemical composition of int

Q. Y. Zhai et al.

## and EDAX.

#### 4.2.1. DD6 TLP Bonding Joints' Structure

**Figure 7(a)** shows the structure of a typical joint made with DIF2 interlayer alloys for DD6 TLP bonding under the condition of welding temperature 1200°C, holding time 8h and pressure 0.3 MPa. The welding joint is composed of three zones of weld center zone, isothermal solidification zone and diffusion-affected zone whose amplifications were shown in **Figures 7(b)-7(d)** respectively. Among the three, the weld center zone is composed of fine grey  $\gamma$  phase, black  $\gamma'$  phase (consistent with the isothermal solidification zone) and the larger black  $\gamma'$ phase. The diffusion-affected zone contains white acicular W-Mo-Boride enriched MyB phase. Each phase' EDS testing point is shown in **Figure 7(a)** and analysis results shown in **Table 3**. The larger black  $\gamma'$  phase in weld center zone precipitates in the cooling process of residual liquid phase because the isothermal solidification time is not enough. The boride phase in the diffusion-affected zone is formed because B diffuses into the base metal in the interlayer. The brittle phase borides are emerged when the content of B is higher than the solubility limit of B in the base metal.



Figure 7. Microstructures of DD6 alloy TLP welding joint. (a) The structure of joint; (b) Amplification of weld center zone; (c) Amplification of isothermal solification zone; (d) Amplification of diffusion-affected zone.

#### 4.2.2. Influence of Alloying Elements on the Joints' Structure

Table 3 Chemical compositions of phases by EDS analysis (wt%)

The structure of welding joints is mainly related with the diffusion of B. Under the condition of the fixed parameters, the diffusion of B is mainly controlled by the contents of the alloying elements in interlayer alloys.

The contents of the alloying elements have certain effect on the structure of the diffusion-affected zone in welding joint which is shown in **Figure 8**. It can be seen that the acicular borides in diffusion-affected zone increases with the increasing of the alloying elements in interlayer alloys which demonstrates that the more the alloying elements in interlayer alloys are, the greater they prevents the solute atomic diffusion and the slower the solute atomic diffusion is. For the same holding time, the more the residual borides in the diffusion-affected zone of the welding joints, the more the brittle borides are generated, which is more harmful to the welding joints.

Therefore, when the welding parameters are welding temperature 1200°C, holding time 8 h and welding pressure 0.3 MPa, the brittle phase borides are fewer in the diffusion zone of the bonding joint made with the interlayer alloys for DD6 TLP bonding with lower contents of alloying elements.

## **5.** Conclusions

1) The microstructures of interlayer alloys prepared with the composition range:  $1 \le Cr \le 2$ ,  $2.5 \le W \le 4$ ,  $3 \le Co \le 4.5$ ,  $1.5 \le Al \le 3$ ,  $0.5 \le Mo \le 1$ ,  $3 \le B \le 3.6$  and remain Ni (wt%) are fine and homogeneous. The melting point of the alloy foils having better welding performance with thickness 40 µm ~ 60 µm ranges from 1070°C to 1074°C and their melting temperature interval is merely 20°C.

2) Under the condition of welding temperature  $1200^{\circ}$ C, holding time 8h and pressure 0.3 MPa, the structure of a typical joint made with the interlayer alloys for DD6 TLP bonding when the chemical composition of alloys are  $1.5 \sim 2$ Cr,  $3.2 \sim 4$ W,  $3.7 \sim 4.5$ Co,  $2.2 \sim 3$ Al,  $0.7 \sim 1$  Mo, 3.2B, remain Ni (wt%), is composed of weld center zone, isothermal solidification zone and diffusion-affected zone.

3) Under the condition of welding temperature 1200°C, holding time 8 h and pressure 0.3 MPa, the brittle

Tuble 5. Chemical compositions of phases by ED5 analysis (with).								
Chemical compositions (wt%)						Phase		
Ni	Co	W	Та	Al	Cr	Mo		
66.5	5.7	5.5	13.0	7.4	1.9	-	γ'	
77.7	7.1	5.7	-	4.1	3.8	1.6	$\gamma'$	
75.0	6.8	8.0	-	6.8	3.4	-	γ	
26.8	4.9	39.6	13.9	-	2.1	12.7	$M_yB$	
	Ni 66.5 77.7 75.0 26.8	Ni Co   66.5 5.7   77.7 7.1   75.0 6.8   26.8 4.9	Ni Co W   66.5 5.7 5.5   77.7 7.1 5.7   75.0 6.8 8.0   26.8 4.9 39.6	Ni Co W Ta   66.5 5.7 5.5 13.0   77.7 7.1 5.7 -   75.0 6.8 8.0 -   26.8 4.9 39.6 13.9	Ni Co W Ta Al   66.5 5.7 5.5 13.0 7.4   77.7 7.1 5.7 - 4.1   75.0 6.8 8.0 - 6.8   26.8 4.9 39.6 13.9 -	Ni Co W Ta Al Cr   66.5 5.7 5.5 13.0 7.4 1.9   77.7 7.1 5.7 - 4.1 3.8   75.0 6.8 8.0 - 6.8 3.4   26.8 4.9 39.6 13.9 - 2.1	Ni Co W Ta Al Cr Mo   66.5 5.7 5.5 13.0 7.4 1.9 -   77.7 7.1 5.7 - 4.1 3.8 1.6   75.0 6.8 8.0 - 6.8 3.4 -   26.8 4.9 39.6 13.9 - 2.1 12.7	



Figure 8. Microstructures of the diffusion zone of DD6 TLP bonding joint at 1200°C for 8 h with various interlayer alloys.

phase borides are fewer in the diffusion zone of the bonding joint made with the interlayer alloys for DD6 TLP bonding when the chemical composition of alloys are  $1.5 \sim 2$ Cr,  $3.2 \sim 4$ W,  $3.7 \sim 4.5$ Co,  $2.2 \sim 3$ Al,  $0.7 \sim 1$ Mo, 3.2B, remain Ni (wt%).

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