Effect of Twist Angle on Mechanical Properties of Diffusion Bonded Joint Using Ni-Base Single Crystal Superalloy TMS-26

Osamu Ohashi[†], Susumu Meguro and Toshihiro Yamagata

National Research Institute for Metals, 1-2-1 Sengen, Tsukuba 305, Japan

The aim of this study is first to find suitable bonding conditions for joining Ni-base single crystal superalloy TMS-26 without the formation of a recrystallized zone at bond-line. Subsequently the study is carried out to investigate the relationship between misorientation angle at bonding interface and joint strength. Joints were made using samples prepared with a {100} crystal plane at the bonding surface. All the samples were joined in a vacuum of 4×10^{-3} Pa using radio frequency heating.

Single crystal TMS-26 can be bonded without the formation of voids and recrystallized zone at the bond-line. The quality of creep rupture of joints deteriorates due to formation of precipitates at the bond-line. There are a few inclusions at the bond-line when the twist angles of 0° up to 3° are used. Creep rupture life of the joints is long. When twist angles greater than 3° are used, many inclusions are formed at the bond-line and joint strengths are very low. The precipitates at the bond-line consist of α tungsten phase and oxides due to surface film.

(Received April 26, 1995)

Keywords: diffusion welding, diffusion bonding, single crystal, nickel-base superalloy, recrystallization, misorientation angle, creep rupture, inclusions, precipitates

I. Introduction

In recent times, active research and development work have focused on the joining of single crystal materials using diffusion bonding methods.

On the other hand some researchers have tried to investigate the effect of misorientation angle between bonding surfaces on joint properties, using single crystal materials.

Some previous work on the diffusion bonding of single crystal materials includes the joining of copper, molybdenum and silicon. In the case of copper⁽¹⁾, it was difficult to make joints without forming a recrystallized zone at bonds. Using single crystal molybdenum and silicon, the joint strength increased as the number of coincident lattice points between two bonding surfaces decreased⁽²⁾⁽³⁾.

There is a patent about liquid phase diffusion bonding of Ni-base single crystal superalloy. The patent is the bonding process that the joints are bonded below 5° as the total misorientation angle at each crystal axis between bonding surfaces⁽⁴⁾. One disadvantage of liquid phase diffusion bonding is the ease with which precipitates form at the bond-line.

The misorientation angle between two bonding surfaces is thought to affect the mechanism of diffusion bonding and hence, the properties of the bonded region. However, very little previous work has been undertaken to investigate the effect of misorientation angle on diffusion bonding behavior.

The aim of this study is at first to find suitable bonding

conditions for joining Ni-base single crystal superalloy without the formation of recrystallized zone at bonds. Subsequently the study is carried out to investigate the relationship between misorientation angle at the bonding interface and joint strength.

II. Material and Experimental Procedure

A Ni-base single crystal superalloy (TMS-26) was developed at National Research Institute for Metals and used in this study. The alloy has gamma prime dispersion of 66.1 vol% and gives a high quality of creep rupture life. Nominal compositions of the alloy used is shown in Table 1. The solution heat treatment recommended is 1330°C , 4 h, air cooling. The aging heat treatment is 982°C , 5 h, air cooling and 870°C , 20 h, air cooling. All heat treatments of the specimens were carried out, after encapsulating them in a quartz tube with argon gas.

Figure 1 shows the configuration of a creep rupture specimen bonded with a twist angle α . The specimens prepared for metallurgical examination were cut from the end of creep specimens. Ones were in the form of discs 13 mm in diameter and 5 mm in thickness.

A {100} crystal plane at the bonding surface was used and confirmed by a Laue X-ray diffraction method.

Surface preparation involved grinding to 600 grit finish

Table 1 Nominal compositions of single crystal Ni-base super alloy TMS-26 (mass%).

Cr	Co	W	Mo	Ta	Al	C	O	N
5.59	8.20	10.95	1.90	7.70	5.13	0.003	0.0008	0.0009

[†] Present address: Graduate School of Science and Technology, Niigata University, Ikarashi, Niigata 950-21, Japan.

Fig. 1 Configuration of a creep rupture specimen bonded with a twist angle α .

using water. Finally, specimens were degreased in acetone with an ultrasonic cleaner. Figure 2 shows a surface profile of specimens measured by a scanning tunnel microscope. The result shows that the formation of parallel rows of the scratches depends on polishing. A contact roughness profile meter gave a surface roughness value of $R_{\max} 0.3 \mu\text{m}$.

All the specimens were joined in a vacuum of 4×10^{-3} Pa using radio frequency induction heating. The twist angle between bonding specimens was measured from angular difference between marks made on the side of the specimens.

The quality of the joints was assessed by metallurgical and mechanical examinations. The bonds were inspected using light and scanning electron microscopy, and precipitates were identified using an electron probe micro-analyzer. The mechanical property of the joints was assessed by a creep rupture testing. The creep rupture tests were carried out at a temperature of 1040°C and a stress of 137 MPa.

III. Results

It is necessary to search for suitable bonding conditions for the joints without the formation of voids and a recrystallized zone at bonding interface. The effect of bonding temperature on the bond microstructures was studied, using a bonding pressure of 6.8 MPa, a bonding time of 1 h and a twist angle of 0° .

Figure 3 shows a light micrograph of the joint made at

Fig. 2 Surface profile by a scanning tunnel microscope.

Fig. 3 Light micrograph of joint produced under 1280°C , 60 min, 6.8 MPa with twist angle of 0° .

1280°C , 1 h, 6.8 MPa. In the case of joints made a bonding temperature of 1250°C , many voids could be seen along the bond-line. The joint made at 1280°C is free from voids and formation of recrystallized zone at the bond-line.

There are many sub-boundaries as seen in Fig. 3. They were also observed in the base metal before bonding, and the distribution and density of sub-boundaries did not depend on the diffusion bonding process.

Using a scanning electron microscope, a few voids could be found along the bond-line of the joint bonded at 1280°C . However, by carried out the post heat treatment at 1330°C for 4 h, the joint without voids could be produced. These bonding conditions and post heat treatment conditions were chosen as most suitable for the joints produced for subsequent creep rupture tests.

Figure 4 shows the relationship between creep rupture

Fig. 4 Relationship between creep rupture time and twist angle at the bonded interface.

time and the twist angle at the bonding interface. Time to rupture for joints was long when a twist angle of 0° was used. When twist angles greater than 3° were used, specimens fractured immediately on loading. Fracture of all joints occurred at the bonded interface. The creep rupture time of the base metal is 1728 h. That of the joint made using a twist angle of 0° was equivalent to about 8% for the base metal.

IV. Discussions

The results suggest that the quality of joints depends on twist angle at the bonding interface. To investigate the reason, the effects of twist angle and heat treatments on the bond-line microstructure of joints were checked using a scanning electron microscopy. Figure 5 shows micrographs of joints produced with a twist angle of 0° and 30° . The microstructure of joints in the as bonded condition shows that some gamma primes precipitate in a line at the bonded interface and the bond-line is also not clear. There is no apparent difference in the joint microstructures at the two twist angles used.

After a post bond heat treatment, no changes in microstructure were recorded when a twist angle of 0° was used. However many precipitates occurred along the bond-line of joints produced at a twist angle of 30° and heated after bonding. It was difficult to find voids along the bond-line. Heat treatment after bonding makes many

precipitates at the bond-line with large misorientation angle between two bonding surfaces.

At creep rupture testing, specimens were held at a temperature of 1040°C for 20 h to make temperature distribution of a specimen uniform, before applying a rupture stress of 137 MPa. To investigate a micro structure of joints at creep rupture testing, the joints were heated at 1040°C for 20 h and quenched in water.

Using the joints heated as described above, the precipitates at the bond-line were counted from photographs taken at a magnification of 5000 with a scanning electron microscope. Figure 6 shows the relationship between the number of precipitates per $10\text{ }\mu\text{m}$ of the bond-line and the twist angle at bonding interface under various heat treatments. In the as bonded condition there were few precipitates at the bond-line at twist angle of 0° up to 30° . When the bonds were heated, a few precipitates occurred at the bond-line at twist angles less than 3° . In the joints made with greater twist angles than 3° , many precipitates were found at the bond-line. The amount of precipitates increased with an increase of heating temperature and heating time.

Joints bonded with twist angles greater than 3° were found to give poor creep rupture time and therefore, the poor bond quality can be attributed to the formation of these precipitates at the bond-line of the joints.

The base metal as shown in Fig. 3 has many sub-boundaries. To measure the misorientation angle between the

Fig. 5 Scanning electron micrographs showing changes in joint microstructure with twist angle 0° and 30° under two heat treatments. (Welding Conditions: 1280°C , 6.8 MPa, 60 min, Postheat Treatment Conditions: 1330°C , 4 h, AC, $+982^\circ\text{C}$, 5 h, AC, $+870^\circ\text{C}$, 20 h, AC.)

Fig. 6 Relationship between the number of precipitates per unit length at bond-line and twist angle at bonded interface under various heat treatments.

sub-boundaries, the base metal was checked using a X-ray Laue method. Figure 7 shows the result and each spot separates into a few small spots. The deviation between these small spots was attributed to about 3° of the maximum misorientation angle.

These results show that the single crystal TMS-26 changes into a polycrystal when the misorientation angle between the grains is above 3° . This alloy can be bonded without forming precipitates when a misorientation angle less than 3° is used.

The quality of the joints is influenced by the formation of precipitates at the bond-line. The precipitates at the bond line were investigated, comparing with the boundary of recrystallized grains and the grains in the selector part, used to control grain growth when making a single crystal.

Fig. 7 X-ray Laue photograph of base metal (distance: 35 mm).

The scanning electron micrographs show the formation of precipitates at grain boundaries as shown in Fig. 8. (A) is at single crystal recrystallized specimen, and (B) is at selector part for making a single crystal. Many precipitates can be seen at grain boundaries in both materials. The composition of precipitates was evaluated using an electron probe micro-analyzer by scanning the electron probe along the direction of the arrows. The composition of the precipitates at the grain boundaries was compared with the composition of the base metal. Table 2 shows the change in element concentration at each precipitate. The mark $\uparrow$ shows the increase of element concentration at precipitate, comparing the base metal. The mark $\downarrow$ shows the decrease and the mark $-$ shows a little change. Both specimens were measured after solution treatment at 1330°C , 4 h, aging treatment and treatment of 1000°C , 20 h, water quenching.

The precipitates seen at the recrystallization boundary were classified into two types. One type contained more aluminum and nickel than base metal and the other type had more tungsten and molybdenum. Therefore the former is considered to be gross gamma prime and the latter is α tungsten phase. The selector part also has the same precipitates, such as gamma prime and α tungsten phase. The results showed that there was no difference between the precipitates formed at the solid phase and at the liquid phase.

In Fig. 9 the scanning electron micrographs show the formation of precipitates at the bond-lines for: (A) twist

Fig. 8 Scanning electron micrographs showing the formation of precipitates at grain boundaries for: (A) recrystallization specimen of single crystal and (B) selector part for making a single crystal.

Table 2 EPMA analyses of precipitates at grain boundaries shown in Fig. 8(A) and (B). Measurement was carried out along direction of arrows.

(A) Grain boundary at recrystallization specimen.

No	Ni	Cr	Co	W	Mo	Ta	Al	O	Phase
1-1	↑	↓	↓	↓	↓	↑	↑	—	γ'
1-2	↓	—	↓	↑	↑	↑	↓	—	α W
2	↓	—	↓	↑	↑	↑	↓	—	α W
3	↓	—	↓	↑	↑	↑	↓	—	α W
4	↓	—	↓	↑	↑	↑	↓	—	α W
5-1	↑	↓	↓	↓	↓	↑	↑	—	γ'
5-2	↓	—	↓	↑	↑	↑	↓	—	α W
6	↓	—	↓	↑	↑	↑	↓	—	α W

(B) Grain boundary at selector specimen.

No	Ni	Cr	Co	W	Mo	Ta	Al	O	Phase
1-1	↑	↓	↓	↓	↓	—	↑	—	γ'
1-2	↓	↑	—	↑	↑	↑	↓	—	α W
1-3	↑	↓	↓	↓	↓	—	↑	—	γ'
2	↓	↑	↓	↑	↑	↑	↓	—	α W
3	↓	↑	↓	↑	↑	↑	↓	—	α W
4-1	↓	↑	↓	↑	↑	↑	↓	—	α W
4-2	↑	↓	↓	↓	↓	—	↑	—	γ'
5	↓	↑	↓	↑	↑	↑	↓	—	α W
6	↓	↑	↓	↑	↑	↑	↓	—	α W

Table 3 EPMA analyses of precipitates at bond-lines shown in Fig. 9(A) and (B). Measurement was carried out along direction of arrows.

(A) Joint at twist angle 0°.

No	Ni	Cr	Co	W	Mo	Ta	Al	O	Phase
1	↓	↓	↓	↑	↑	↑	↓	—	α W
2	↓	—	↓	↑	↑	↑	↑	↑	Oxide
3	↓	—	↓	↑	↑	↑	↓	—	α W
4	↓	—	↓	↑	↑	↑	—	—	α W

(B) Joint at twist angle 30°.

No	Ni	Cr	Co	W	Mo	Ta	Al	O	Phase
1	—	↓	↓	↑	↑	↑	—	—	α W
2	↓	↓	↓	↑	↑	↑	↑	↑	Oxide
3	↓	↓	↓	↑	↑	↑	↑	↑	Oxide
4	↓	—	↓	↑	↑	↑	↓	—	α W

angle 0° and (B) 30°. Though it was difficult to find the precipitates at the bond-line for a twist angle of 0°, the micrograph shows a part of the bond-line that has a few precipitates.

Table 3 shows the change in element about each precipitate at the bond-lines for a twist angle of 0° and 30°. Some precipitates at the bond-line contained much oxygen. Auger analyses taken from the bonding surfaces before bonding show an oxidized surface covered with alumina. The bonding surface was oxidized and covered with alumina. Therefore some precipitates are oxides that contain alumina at the bonding surface.

The other precipitates seen at the bond-line are α tungsten phase. When a twist angle of 30° was used, much α tungsten phase was observed at the bond-line, as compared with bonds with a twist angle of 0°. The formation of precipitates at the bond-line of the joint with twist angle 0° seems to be due to a local increase of misorientation angle between the bonding surfaces that have a micro surface roughness.

Joint strengths of single crystal molybdenum decrease as the number of coincident lattice points between the bonding surface decreases, and the twist angle between the bonding surfaces is greater than 10°⁽⁴⁾.

On the other hand, joint strengths of Ni-base single crystal superalloy decrease due to the formation of precipitates such as a tungsten phase when the twist angle is greater than 3°. The twist angles at which the bond-line is free from precipitates equals the misorientation angle at the sub-boundaries. A misorientation of 3° is a characteristic value for single crystal Ni-base superalloy used.

V. Conclusions

The effect of misorientation angle between two bonding surfaces on the properties of the diffusion bonded joint was investigated using single crystal Ni-base superalloy TMS-26. The conclusions are as follows:

(1) Single crystal TMS-26 can be bonded without the formation of voids and a recrystallized zone at the bond-

(A)

(B)

Fig. 9 Scanning electron micrographs showing the formation of precipitates at bond-lines for: (A) twist angle 0° and (B) 30°.

line. The most suitable conditions for bonding are 1230°C, 1 h, 6.8 MPa and a post-bond heat treatment of 1230°C, 4 h.

(2) The quality of creep rupture of joints is attributed to the formation of precipitates at the bond-line. Many precipitates occur at the bond-line when twist angles above 3° are used. The amount of precipitates increases with an increase of heating temperature and time.

(3) The precipitates at the bond-line consist of oxides due to surface film and α tungsten phase.

(4) Single crystal TMS-26 can be bonded without the formation of precipitates when a misorientation angle of less than 3° is used.

(5) The misorientation angle at sub-boundaries of the alloy is maximum 3°. This alloy can be bonded without forming precipitates when a misorientation

angle less than 3° is used.

Acknowledgments

The authors would like to thank Mr. Kimihiko Sato of formerly graduate student of Shibaura Institute of Technology for the cooperation with the experiments, and Dr. Tahir Khan of visiting researcher for his fruitful discussion.

REFERENCES

- (1) T. Momono and K. Inoue: J. Japan Inst. Metals, **42** (1978), 211.
- (2) O. Ohashi and S. Suga: Quarterly J. Japan Welding Society, **10** (1992), 53.
- (3) K. Sato, H. Miyazaki, Y. Ikuhara, H. Kurishita and H. Yoshinaga: J. Japan Inst. Metals, **53** (1989), 536.
- (4) A. F. Giami, B. H. Kear: United States Patent, July, 5, 1977, 4033792.