Transient Liquid Phase Bonding for Ni-base Superalloys, Mar-M247 and IN939

Masako Nakahashi*, Seiichi Suenaga*, Makoto Shirokane*
and Hiromitsu Takeda*

This study discusses TLP bonding of Ni-base superalloys Mar-M247 and IN939, using filler metals specially designed and fabricated into flexible foils by rapid solidification processes. Filler metals, selected by metallographic studies, produce almost the same microstructure in TLP bonds as in the base metal.

The mechanical properties for TLP bonds have been investigated by stress rupture tests, which are the severest tests used to evaluate bonding strength.

The results have also revealed that sufficient bonding pressure was indispensable for Mar-M247 to obtain high bonding efficiency. The TLP bonding efficiency for IN939 was obtained easier than Mar-M247.

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I. Introduction

TLP bonding⁽¹⁾ or Activated Diffusion Bonding⁽²⁾ is a bonding process that combines the manufacturing ease of brazing with the high efficiency of solid state diffusion bonding. TLP bonding is applicable to Ni-base superalloys that are difficult to bond by conventional fusion welding, because of their fusion cracking troubles. This process is carried out with vacuum furnace brazing using filler metals with a specific composition, usually Ni-base alloys containing boron. These filler metals temporarily melt and then resolidify at the bonding temperature according to the boron diffusion into the base metals. Moreover, it is said that by the post bond heat treatments, the elements composing base metal diffuse into the TLP bonds and make as same bond as base metal.

In manufacturing superalloy turbine blades, the metal bonding process is often employed to attain complex shaped turbine blades with excellent cooling ability. They are necessary to attain high temperature gas turbines, which achieve high thermal efficiencies and reduce fuel consumption.

This paper discusses TLP bonding for Ni-base superalloys Mar-M247 and IN939, which contain many elements to enhance high temperature strength, using filler metals specially designed and produced by rapid solidification. Influences of elements Al, W, Si in the filler metals and the bonding process upon bonding strengths are discussed.

II. Experimental

1. Base metals

Commercial conventional cast Mar-M247 and IN939 (produced by KOMATSU-HOWMET Corp.) 13 mm

dia. 100 mm length bars were used for this study. Mar-M247, one of the most difficult alloys for fusion welding, is hardened by approximately 60% volume fraction of γ' ($\text{Ni}_3(\text{Al}, \text{Ti})$) and matrix strengtheners Co, Ta, W, Mo, etc.

2. Filler metals

Table 1 shows filler metal elements and their roles. A symbol O shows that the addition of the element is effective to roll and X shows harmful. Al, Ti are γ' formers, Co, W, Ta are γ matrix strengtheners, Ti, W, Ta, Hf are carbide formers, Al, Cr are oxide formers and Ti, Cr, Mo concern the precipitation of a harmful phase, *e.g.* TCP phase. Group IV elements are used for glassy thin foil formers and melting point depressant. The B+Si amount should be 18–20 at% to make flexible amorphous filler metal.

Five filler metal alloy compositions, shown in Table 2, were determined in this experiment. The Co, Cr and Ni contents of the alloys were fitted to the base metal and main γ' former Al and carbide former W contents were changed, respectively. Ti was eliminated from the alloy, because it has been found to form harmful phases during TLP bonding. B was used for metalloid, except for F25, because of its rapid diffusivity during heat treatment. Only Si containing F25 was amorphous, but the other filler metals had sufficient flexibility to be treated as brazing foil. MBF80A is a commercial amorphous filler metal.

Filler metal alloys were initially melted and refined as 2 kg ingots in 10^{-3} Pa vacuum. They were rapidly quenched from the liquid state using a single roll under Ar atmosphere. Produced filler metals were 25–50 μm thick and 10 mm wide. Melting points of the filler metals are between 1373 and 1423 K.

3. Bonding procedure

Test piece bonding surfaces were polished by #600 emery paper and degreased prior to assembly. Bonding

* R&D Center, Toshiba Corporation, Kawasaki 210, Japan.

Table 1 Filler metal elements and their roles.

		Elements	Ni	Co	Cr	W	Hf	Nb	Ta	Ti	Al	C	B	Si
I	Strengtheners	Matrix class	○	○	○	○								
		γ' class					○	○	○	○	○			
		Carbide class			○	○	○	○	○	○		○		
		Grain-boundary class										○	○	
II	Oxide-scale subclass				○						○			
III	Unwanted phases	TCP phase class			×	×			×	×				
		Stable interface phase class								×				
IV	Metalloid	Glassy											○	○
		Melting point depressant				×	×	×			○	○	○	○

Table 2 Base metal and filler metal chemical compositions (mass%).

	Ni	Co	Cr	W	Ta	Ti	Al	B	Others
Mar-M247	Bal.	10	8.5	10	3	1	5.5	0.015	Hf1.4
IN939	Bal.	19	22.4	2	1.4	3.7	1.9		
F24	Bal.	10.8	8.8	3.9	3.0		3.0	2.5	
F25	Bal.	9.8	8.6	8.2				2.4	Si3.0
F26	Bal.	9.2	8.6		2.7		4.9	3.6	
F27	Bal.	9.8	7.5	1.9				3.3	
F20	Bal.	17.6	21.6	3.1	1.3		2.8	2.2	
MBF 80A	Bal.		15.0					4.0	

temperature was 50–100 K higher than the filler metal melting point. Bonding was carried out in vacuum. After that, diffusion heat treatment and aging treatment $1353 \text{ K} \times 14.4 \text{ ks} + 1146 \text{ K} \times 72 \text{ ks}$ were carried out in Ar atmosphere. Specimens for tensile test and creep rupture test (shown in Fig. 1) were machined before the aging heat treatment.

III. Results and Discussion

1. Bond microstructures

First, metallographic examinations were made on the TLP bonds in Mar-M247 using F24–F27.

Figure 2 shows the Al content effect on γ' precipitation in the bond region. Dark portions are γ' precipitates.

Fig. 1 Specimen (in mm) for tensile test and stress rupture tests.

When Al less filler metal (F27) was used, Al diffused from the base metal and fine γ' precipitated all over the bond. When 4.9% Al was used, the bond region was occupied by condensed γ' , as shown in Fig. 2. By the morphological measurement, volume fractions of γ' were 44% in the former, 82% in the latter and 53% in the base metal. The volume fraction of γ' in the bond area, using Al-less filler metal, was smaller than that for base metal.

Ohashi *et al.*⁽³⁾ investigated the γ' precipitation in Mar-M247 TLP bonds and reported that only fine γ' precipitates were observed in the bond made with Al-less filler metal. Also, longer diffusion treatment was needed to precipitate as much γ' in the bond region as base

Fig. 2 Al content effect on γ' precipitation in the bond region.

metal. In this research, it was found that Al containing filler metal can precipitate a large quantity of fine and also coarse γ' in a short heat time. The Al content, required to precipitate as much γ' (60 vol%) as base metal, was calculated, assuming that base metal consisted of Ni and Al, and that they didn't solve each other. The calculation led to 7.3 mass% was retained. Therefore, 4.9 mass% Al was not sufficient to precipitate as much γ' as base metal. Actually, however, excess γ' appeared at the TLP bond. It is considered that (1) the Al solubility in Ni is approximately 8 mass%, but Al, insoluble by short time heating, remain in the bond region (2) the solidification of the filler metal starts at the interface of the filler metal/base metal, according to the diffusion of the melting point depressant element B into the base metal, and ends at the center of the bond. Finally, the low melting point phase γ' solidification occurs at the center of the bond. It is considered that an adequate quantity of Al addition to the filler metal is effective in instigating

fine and coarse γ' precipitation.

Next, primary carbides were examined metallographically by electron probe microanalysis on TLP bond. The bond with F25, which contains only 1.9 mass% W as carbide former, indicated that a smooth transition on W composition was achieved across the bond region, after diffusion treatment at 1473 K for 86.4 ks. However, less primary carbides existed at the TLP bond than in the bond with 8.2 mass% W filler metal F27. Figure 3 shows the primary carbides in those two bonds. W addition to filler metal seemed effective to precipitate primary carbides, but large quantities of W might be the cause of a melting point increase.

Ti addition to the filler metal seemed to precipitate a harmful Ti-rich phase at the bond.

The Si addition effect on the bond was investigated by the hot corrosion test. Figure 4 shows TLP bond sections with F25 and F27 after heating at 1373 K for 10.8 ks in air with 20 mg/cm² of the V₂O₅-Na₂SO₄ mixture on the

Fig. 3 W content effect on carbide precipitation in the bond region.

Fig. 4 Bond section after the corrosion test.

bonds. The corrosion rate for a bonding region with Si-containing F25 was larger than that for Si-less F27.

The Si concentration C_p of the bond after heat treatment was calculated by eq. (1)⁽⁴⁾.

$$C_p = C_0 h_0 / 2 \sqrt{\pi D t} \quad (1)$$

C_0 : initial content, t : time

h_0 : filler metal thickness

$D = D_0 \exp(-Q/RT)$

D_0 of Si in Ni = $1.5 \times 10^{-4} \text{ m}^2/\text{s}$ and $Q = 258 \text{ kJ/mol}$ were used. C_p after 1473 K for 86.4 ks heat was almost 0.265 mass%.

It is considered that corrosion rate difference was due to the Si remaining. Si seemed to cause an undesirable effect on the hot corrosion.

F24 filler metal compositions were determined for Mar-M247 and F20 for IN939, for the reason mentioned above. Only B was added to both filler metals for melting point depressant.

2. Bond mechanical properties

Initially the bonding temperature and bonding pressure effects were examined.

TLP bonding is a kind of brazing. The molten filler metal makes good contact to both base metals. Therefore, bonding pressure is unnecessary in principle. Figure 5 shows the cross sections of Mar-M247 bonds made at 10 and 0.1 MPa bonding pressures. The 0.1 MPa bond showed a straight boundary at the center, even after the 1503 K, 259.2 ks diffusion heat treatment. On the other hand, the 10 MPa bond microstructure was

nearly equivalent to that for base metal. The stress-rupture properties of these bonds indicated a marked difference in the time to failure, as shown in Fig. 6. The fracture surfaces were almost flat on the specimen for the 0.1 MPa bonding pressure and jagged for the 10 MPa bonding pressure. It is considered that when the bonding pressure was small, stable compounds as oxides remained at the surface of the bonds and restricted the boundary

Fig. 6 Bonding pressure and filler metal composition effect on time to failure for Mar-M247 bonds.

Fig. 5 Cross sections of Mar-M247 bonds. Bonds showed high bonding strength, 10 MPa bonding pressure and low bonding strength, 0.1 MPa.

movements. It was supported by the morphology of the fracture surfaces.

The bonding temperature was not so effective for the rupture life, if the bonding pressure was sufficient (10 MPa), as illustrated in Fig. 7. However, the bond with non tailored (simple composition) filler metal MBF80A showed 1/3 the life of the bond with a tailored filler metal. Large bonding pressure and tailored composition filler metal are necessary to get as long a rupture life as the base metal has.

It is considered that the bonding pressure is effective for pushing out such impurities as oxides from the bond or reducing the filler metal thickness, which decreases the

heat treatment time. Another effect is assumed to be that due to pushing out of the liquid filler metal with impurities, fresh partially melt and softened solid surfaces are directly contacted and make a strong metallic bond.

High temperature (1033 K) tensile tests showed that the 0.1 and 10 MPa bonded Mar-M247 with F24 were 984 and 950 MPa, respectively. They were almost the same as those of base metal 907–942 MPa. This might support the concept that grain boundary strength is predominant over the stress rupture property and the transgranular strength is predominant over tensile strength.

Figure 8 shows bonding pressure and filler metal composition effects on the time to failure for the IN939 bond.

Fig. 7 Bonding temperature effect on the time to failure for Mar-M247 bonds.

Fig. 8 Bonding pressure and filler metal composition effect on time to failure for IN939 bonds.

Fig. 9 Diffusion time effect on time to failure for Mar-M247 bonds.

Fig. 10 Diffusion time effect on time to failure for IN939 bonds.

High Cr and low Al, Ni-base alloy IN939, which had smaller high temperature strength than Mar-M247, indicated a higher bonding efficiency than Mar-M247.

Both bonding pressure and bonding temperature, as well as filler metal composition were scarcely effective for the time to failure in IN939.

The relations between diffusion heat treatment time and the time to failure were examined for Mar-M247 and IN939. As expected, Figs. 9 and 10 showed that bonding efficiencies increased as the diffusion heat treatment time increase.

Maximum Mar-M247 efficiency 74.5% was obtained when treated at 1503 K for 86.4 ks. IN939 indicated almost the same stress rupture properties as base metal by only 3.6 ks diffusion heat treatment.

IV. Conclusion

TLP bonds, using specially designed filler metal in Mar-M247 and IN939, were examined. The conclusions

are as follows.

(1) Microstructures of Mar-M247 TLP bond, almost the same as those for base metal, were attained with the filler metal containing adequate quantities of strengtheners 10.8Co-8.8Cr-3.9W-3.0Ta-3.0Al-2.5B. Al addition to the filler metal promoted the precipitation of γ' .

(2) Sufficient bonding pressure was also indispensable for Mar-M247 TLP bond to achieve high stress rupture properties.

(3) IN939 TLP bond showed better bonding efficiency than Mar-M247 TLP bond.

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