THEORETICAL RESEARCH ON TRANSIENT LIQUID INSERT METAL

DIFFUSION BONDING OF NICKEL BASE ALLOYS

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Abstract

This paper is concerned with the theoretical consideration on the transient liquid insert metal diffusion bonding(TLIM bonding) process.

Main results obtained in this study are as follows;

(1) The dissolution phenomenon of base metal into liquid insert metal can be explained by Nernst-Brunner theory.

(2) The isothermal solidification process can be explained theoretically on Ni-B and Ni-P binary systems and the complete time of the process can be estimated by eq.(13).

The isothermal solidification process of Mar-M247, MM007, Alloy 713C, Inconel 600 joints with using Ni-15.5Cr-3.7B insert metal can be also interpreted by the same theoretical equation applied to the process of Ni-B and Ni-P binary systems.

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Introduction

Ni-base cast superalloys are a class of materials that are generally used hottest parts in aircraft or land-base gas turbines. As fair amount of γ' forming elements such as Al, Ti, Nb, Ta is contained, these alloys are very sensitive to hot cracking during fusion welding[1].

TLIM bonding has been recently developed to join hot cracking susceptible Ni-base cast superalloys and called 'TLP bonding' or 'Activated diffusion bonding'[2]. A liquid film temporarily forms at bonding interlayer during TLIM bonding and solidifies isothermally during interdiffusion of elements for depressing melting point of filler metal. Consequently, the excellent joint quality can be expected without any fusion welding defects. Therefore, TLIM bonding has been applied for bonding of Hastelloy X[2], Inconel X[3], Udimet 500[3],700[2], Alloy 713C[2], Mar-M247[4], Renè 80[5], IN-100[6], TD-NiCr[7] and MA 754[8].

The bonding process of TLIM bonding is composed of the following three processes. Namely, the first one is the melting process of base metal adjacent to the liquefied insert metal. The second one is the isothermal solidification process and the last one is the homogenizing process of alloying elements. It is essentially important to make clear these processes theoretically in order to understand the mechanism of TLIM bonding.

In this research, the melting process of base metal and the isothermal solidification process were studied theoretically.

Experimental Materials and Procedures

Experimental Materials

In this study, commercially pure Ni and Ni-base superalloy, MM007, Mar-M247, Alloy 713C, Inconel 600 were used for base metals. Amorphous alloys commercially available and electroless Ni-B plating were adopted as insert metals. MM007 and IM-l were used to study the melting process of base metal. Commercially pure Ni, MM007, Mar-M247, Alloy713C, Inconel 600 were used as base metals and IM-l, IM-2, Ni-B plating were used as insert metals to clarify the isothermal solidification process. Chemical compositions of these materials are presented in Table I.

Materials		Melting range(K)	Thick- ness(µm)	Chemical composition (wt%)														
				Ni	В	Co	Cr	A1	Ti	Mo	¥	H£	Ta	С	Si	Fe	S	P
	Ni			Bal.	-	-	-	-	-	-	-	-	-	0.02	0.19	0.04	0.01	_
Base	MM 007			Bal.	0.016	10.2	7.98	6.01	0.96	6.17	0.05	1.34	4.16	0.09	-	-	-	-
metal	MarM 247		1	Bal.	0.015	9.87	8.27	5.53	0.99	0.69	9.92	1.51	3.06	0.15	-	_	_	-
	Inconel 713C			Bal.	0.011	0.4	13.4	5.8	0.9	4.2	-	2.1	2.1	0.11	_	ĺ -	_	-
ĺ	Inconel 600	ĺ	[Bal.	-	-	15.5		_	-	-	- 1	-	0.03	_	7.7	-	-
Insert	IM-1	1293-1338	38	Bal.	3.7	-	15.5	-	-	-	-	-	-	-	-	_	-	_
metal	IM-2	1153	44	Bal.	-	-	-	-	-	_	_	-	_	-	-	_	-	11.5
	Ni-B	1353	20	Electroless plating														

Table I Chemical compositions of materials used(wt%)

Experimental Procedures

Bonding Procedures. A bonding equipment is illustrated in Fig.1. Specimens for bonding were held together with the characterized insert metal under slight compressive pressure as shown in Fig.1 and heated to the each bonding temperature in vacuum(13.3mPa) by the high frequency induction heating source and held for predetermined time. Prebond surface finishing of each specimen was achieved by #1500 polishing paper and its prebond cleaning was conducted in acetone by an ultrasonic cleaner. Each specimen

size of MMOO7, Mar-M247, Alloy713C and Inconel 600 is ϕ 10 X 10mm and that of commercially pure Ni is 5 X 10X 12mm.

Figure 2 shows commercially pure Ni joints for examining the isothermal solidification process. Ni stoppers were spot welded in order to keep the width of the bonding interlayer constant. These specimens were also heated to predetermined temperatures in vacuum by a high frequency inductionheating source.

Procedure for Measuring Dissolution Width of Base Metal. Prebond alignment of the piece being joined and bonded test piece are schematically illustrated in Fig.3. Dissolution width of base metal, X was calculated by the following equation.

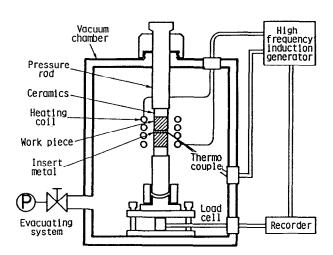


Fig.1 Schematic diagram of bonding apparatus

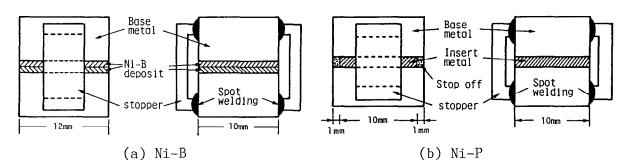


Fig.2 Schematic illustration of test specimen

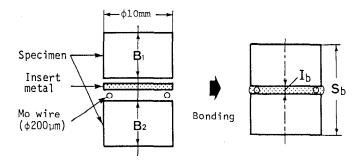


Fig.3 Prebond alignment of the piece being joined and bonded test piece

$$X = \{(B_1 + B_2) - (S_b - I_b)\}/2$$
 (1)

where B_1 , B_2 ;length of base metal before bonding, S_b ;length of bonded test piece and I_b ;width of interlayer. Mo wire(φ 200 $\mu\text{m})$ was used to maintain I_b constant and IM-l was used as an insert metal. B₁, B₂ and S_b were measured by a dial gauge-strain amplifier system(accuracy of the measurement;0.5 μm). I_b was measured by a microscopy.

Procedure for Measuring the Mean Width of Retained Liquid Phase in the Bonding Interlayer. Metallurgical observation(X 300) of the bonding interlayer was performed and the contour line of eutectic was traced at the central region(4.5mm length) of the bonding interlayer on each specimen heat-treated at predetermined conditions. Total area of the eutectic in the central region was examined by measuring area of the outlined eutectic figure using an area-analyser. The mean width of the eutectic was determined by dividing the total area of eutectic by the length of the central region of the bonding interlayer, 4.5mm. The mean width of retained liquid phase in the bonding interlayer was estimated by eq.(14).

Dissolution Phenomenon of Base Metal into Liquid Insert Metal

A kinetic study has been carried out to make clear the dissolution phenomenon of base metal into liquid insert metal. The isothermal phase transfer from solid to liquid can be broken up into two consecutive steps. The first is the surface reaction in which atoms go from the solid into the liquid phase. The second is the diffusion process in which the solute atoms diffuse from the interface into the bulk liquid.

The Nernst-Brunner theory [9] of dissolution has been applied to the dissolution phenomenon of solid to liquid. In this study, the dissolution phenomenon of base metal into liquid insert metal was analysed based on the Nernst-Brunner theory which was formulated as follows.

$$n=n_{s}[1-\exp\{-(KA/V)t\}]$$
 (2)

where n_s ,n ;solute concentrations at saturated state and at spontaneous time, t respectively, K ;dissolution rate constant, A ;surface area of the solid and V ;volume of the liquid.

When n and n_s are expressed in weight %, C_t and C_s respectively, eq.(2) can be given by eq.(3). C_t and C_s of B can be given by eq.(4) in the case of the joint illustrated in Fig.3.

$$dc/dt = K(A/V)(C_s - C_t)$$
(3)

$$C_{t} = \{ph/(x+ph)\}C_{i}$$
 $C_{s} = \{ph/(x_{s}+ph)\}C_{i}$
(4)

where C_i ; initial concentration of B in liquid insert metal, p;(ρ_1/ρ_s), ρ_1 , ρ_s ; density of liquid insert metal and base metal, 2h; initial width of liquid insert metal and x ,x, dissolution width of base metal at spontaneous time,t and at saturated state, respectively. Eq.(5) is given by eq.(3) and (4).

$$Kt=h[ln\{x_{s}(x+ph)/ph(x_{s}-x)\}]=P$$
 (5)

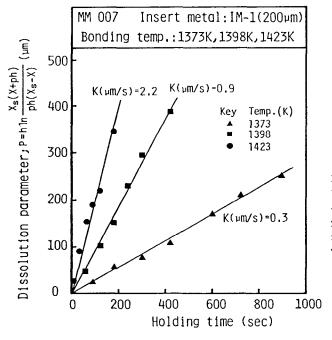
where P; dissolution parameter.

If dissolution rate constant, K is expressed as $K=K_0\exp(-Q/RT)$, following equation can be obtained.

$$1n K=-Q/RT + 1n K_{O}$$
 (6)

where Q ;apparent activation energy of dissolution of base metal into liquid insert metal, T ;temperature , $K_{\rm O}$; frequency factor and R ; gas constant.

The linear relationship between holding time,t and dissolution parameter,P can be expected during dissolution process of base metal from eq.(5) and Q can be estimated from correlation between ln K and l/T. Figure 4 indicates the relationship between holding time,t and P. As predicted from eq.(5), there is a good linear correlation between holding time,t and P. Figure 5 also shows a good linear relationship between ln K and l/T which is predicted from eq.(6). The apparent activation energy of dissolution, Q was estimated 163 kcal/mol from the result in Fig.5. It is concluded that the dissolution phenomenon of base metal into liquid insert metal can be explained by Nernst-Brunner theory.



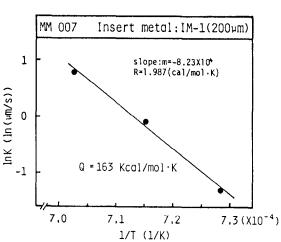


Fig.4 Dissolution parameter,P/ time,t relationships at various temperature

Fig.5 Temperature dependence of dissolution rate constant

<u>Isothermal Solidification Process on</u> Transient Liquid Insert Metal Diffusion Bonding

As shown in a schematical Ni-X binary phase diagram(Fig.6(a)), when the bonding pair is heated to the bonding temperature, $T_{\rm B}$, a liquid film is formed at the bonding region. Shaded area of Fig.6(b) indicates such a liquid film and a schematic X content profile at the bonding region is shown in Fig.6(c). At the bonding temperature, $T_{\rm B}$, a liquid phase of which X content is $C_{\rm 1}$ is in equilibrium with a solid phase of which X content is $C_{\rm S}$. Consequently, at the interface between the liquid phase and the solid phase, both X contents of the liquid phase and the solid phase should be kept constant. If X atoms diffuse into the solid phase from the liquid phase, the solid-liquid interface must move to the minus direction of x axis to make X content in the liquid phase constant, $C_{\rm 1}$.

At the solid-liquid interface, the concentration gradient of X in the solid phase is assumed to be constant even if the solid-liquid interface travels. And as the liquid film was very thin comparing with the length of base metal, one dimensional diffusion model was adopted. X flux, J, per unit area of the solid-liquid interface and unit time is given by eq.(7).

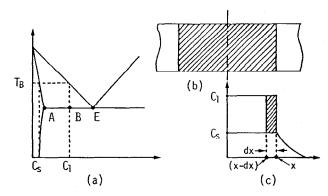


Fig.6 Schematic diagrams of Ni-X binary alloy and concentration profile of X at bonding region

If the solid-liquid interface moves dx for time interval, dt on account of X atoms diffusing into the solid phase, J is given by eq.(8).

$$J=-(D/V_{s})(dc/dx)$$
 (7)

$$J=-(dx/dt)(C_1/V_1-C_S/V_S)$$
 (8)

X concentration at arbitrary position x in the solid phase is described by eq.(9) under the condition of the solid phase at the solidliquid interface being constant, C_s . From eq.(9), (dc/dx) at x=h is expressed by eq.(10).

$$(C/V_S) = (C/V_S) \left[1 - \operatorname{erf}\left((x-h)/2\sqrt{Dt}\right)\right]$$
(9)

$$\left(\frac{dc}{dx}\right)_{x=h} = -C_{S} / \sqrt{\pi Dt}$$
 (10)

From eqs. (7), (8) and (10), the thickness of the liquid film, 2x at arbitrary time, t is given by eq.(11). If the gradient of linear relationship expressed by eq.(11) is represented as m, natural logarism of m is given by eq.(12).

$$2x=2h-(4C_{S}/\sqrt{\pi}V_{S})(C_{1}/V_{1}-C_{S}/V_{S})^{-1}\sqrt{Dt}$$
(11)

$$\ln m = \ln \left\{ (4C_s / \sqrt{\pi}V_s) (C_1 / V_1 - C_s / V_s)^{-1} \right\} + (\ln D_0) / 2 - (Q/2RT)$$
 (12)

From eq.(12), activation energy of X diffusion in the solid phase can be estimated approximately. C_1 is mol fraction of X in the liquid phase at T_B , C_s is mol fraction of X in the solid phase at the solid-liquid interface at T_B , V_1 is molar volume of the liquid phase at T_B , V_s is molar volume of the solid phase at T_B , V_s is molar volume of the solid phase at T_B , T_B is initial thickness of the liquid phase at arbitrary time, t, D is diffusion coefficient, R is gas constant

T is absolute temperature and \mathbf{D}_0 is frequency factor. The complete time(\mathbf{t}_f) of the isothermal process can be estimated by eq.(13).

$$\sqrt{t_f} = (4C_s / \sqrt{\pi} V_s)^{-1} (C_1 / V_1 - C_s / V_s) 2h / \sqrt{D}$$
(13)

As it was a little difficult to determine the solid-liquid interface at room temperature, the thickness of Ni-X eutectic was measured in this experiment and the eutectic thickness, We was converted into the thickness of the liquid film, W_1 using eq.(14).

$$W_1 = W_{e} \{ 1 + (BE/AB)(\rho_{e}/\rho_{s}) \}$$
 (14)

where ρ e; density of Ni-P eutectic and ρ s; density of Ni solution.

As expected from eq.(11), there are good linear correlations between reduced width of eutectic and square root of holding time at each holding temperature, in Ni-B and Ni-P systems, as shown in Fig.7.

Figure 8 shows the relation between 1n m and 1/T. As presumed from eq.(12), there are good linear correlations between 1n m and inverse of absolute temperature, T in Ni-B and Ni-P systems. Each activation energy of B and P diffusion in Ni, obtained from Fig. 8 is 226 KJ/mol and 284 KJ/mol, respectively. As the term, $(4\text{C}_\text{S}/\sqrt{\pi}\text{V}_\text{S})^{-1}(\text{C}_1/\text{V}_1\text{-C}_\text{S}/\text{V}_\text{S})$ in eq.(13) is nearly independent of the temperature in the temperature range from 1373K to 1523K on both Ni-B and Ni-P systems, the square root of t_f is directly proportional to $2h/\sqrt{D}$ according to eq.(13).

As indicated in Fig.9, there are good proportional relations between $\sqrt{t_f}$ and $2h/\sqrt{D}$ in both Ni-B and Ni-P systems. The disappearance time of liquid phase at bonding interlayer can be estimated by eq.(13) in both Ni-B and Ni-P systems. The isothermal solidification process on Ni-base superalloy joints were also examined.

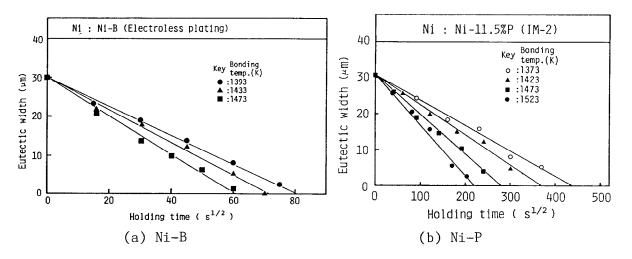


Fig. 7 Effect of holding time at each temperature on eutectic width

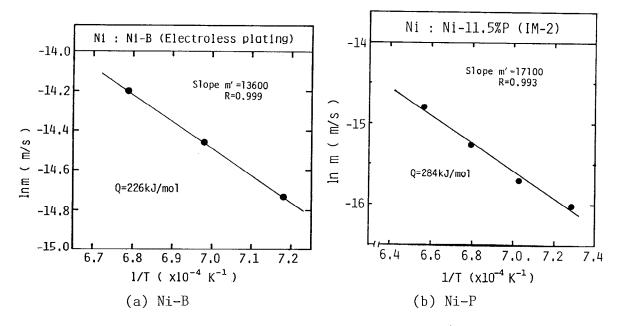


Fig. 8 Relation between 1n m and 1/T

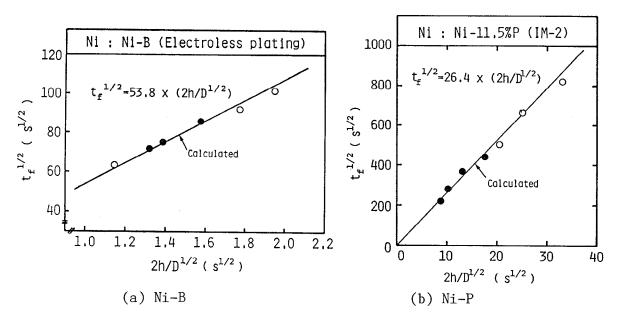


Fig.9 Comparison between calculated value of $\sqrt{t_f}$ and experimental ones

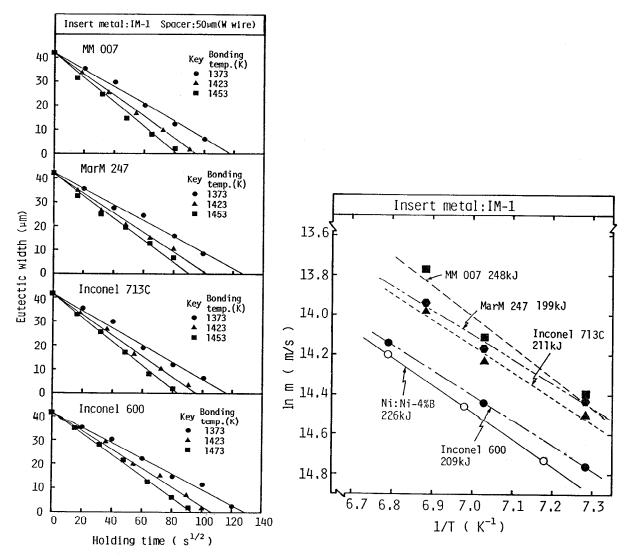


Fig.10 Effect of holding time at each temperature on eutectic width in various commercial alloys

Fig.11 Relation between 1n m and 1/T in various commercial alloys

Figure 10 indicates that there are also good linear relationships between reduced width of Ni solid solution- B eutectic in MMOO7, Mar-M247, Alloy713C and Inconel 600.

In these Ni-base superalloy joints, linear relationships between \ln m and 1/T are also obtained as shown in Fig.11.

Based on the experimental results mentioned already, it was made clear that the solidification process of liquid film during TLIM bonding was controlled by the diffusional process of dipressant element in the base metal.

Conclusions

In this study, the bonding process on the transient liquid insert metal diffusion bonding(TLIM bonding) of Ni-base alloys was considered theoretically.

Experimental results obtained in this study are as follows;

- (1) The dissolution phenomenon of base metal (MM007) into liquid insert metal(Ni-15.5Cr-3.7B) can be explained by Nernst-Brunner theory.
- (2) The isothermal solidification process can be explained theoretically on Ni-B and Ni-P binary systems and the complete time of the process can be estimated by eq.(13).

The isothermal solidification process of Mar-M247, MM007, Alloy713C, Inconel 600 joints with using Ni-15.5Cr-3.7B insert metal can be also interpreted by the same theoretical equation applied to the process of Ni-B and Ni-P binary systems.

(3) It is clarified that the isothermal solidification process of TLIM bonding is controlled by the diffusional process of dipressant element in the base metal.

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