OPTIMIZATION OF PROCESS VARIABLES DURING TLP BONDING OF NICKEL SUPERALLOYS

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ABSTRACT

In this study, the kinetics of isothermal solidification during TLP bonding of Inconel 625 and 718 superalloys with nickel based multi-component filler alloy BNi-2 have been studied through a combination of direct experimentation and mathematical modeling in order to determine the effect of process variables and, thus, to optimize the process. The predicted isothermal solidification completion times from the boron diffusion model and migrating solid/liquid interface model were in good agreement with the experimentally determined values and, they were found to be significantly less than those of other nickel superalloys with different nickel based filler alloys. Further, significant reduction of holding time was observed with increasing bonding temperature and with decreasing joint gap. No significant grain growth in the base metals has been observed in the temperature range being investigated (1325 – 1394K).

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INTRODUCTION

Inconel 625 and 718 superalloys are extremely versatile austenitic nickel based superalloys with excellent strength and good ductility at very high temperature. Typical applications include aero-engine hot section components, miscellaneous hardware, tooling and liquid rocket components involving cryogenic temperatures. However, like other austenitic nickel based superalloys that contain a substantial amount of Ti and Al, they are highly susceptible to heat affected zone cracking during welding [1,2]. Typical high temperature brazing with nickel based filler alloys, containing boron and silicon as melting point depressants, evolved as an effective way to join these superalloys. However, these melting point depressants form eutectic structures that are extremely hard and contain very brittle intermetallic compounds with nickel and chromium which are detrimental to the mechanical properties of the brazed joint [3-5]. One method to prevent the formation of these deleterious phases is transient liquid phase bonding (TLP), also known as diffusion brazing [6,7]. The TLP bonding process uses a low melting filler alloy to wet the contacting base material and that subsequently solidifies isothermally via a fast diffusing element, e.g. boron. Unlike conventional brazing, the thermal exposure used for the TLP bonding is sufficient to induce isothermal solidification at the bonding temperature [8]. Thus, at a relatively low melting temperature, diffusion brazing produces a joint that has a uniform composition profile and that is relatively more tolerant of surface oxides, geometrical defects and wide gaps [6,8]. These advantageous features have been exploited in a wide range of applications, from the production and repair of turbine engines in the aerospace industry to the connection of circuit lines in the microelectronic industry [6-8].

For a given operating temperature, transient liquid phase bonding process relies on the time required to complete the isothermal solidification to prevent the formation of the brittle eutectic phases in the resulting brazed joints. Boron composition reaches the solidus value during the holding period because of diffusion towards the base metal and thus, the formation of eutectic phases is avoided during cooling.

Tuah-Poku *et al.* [9] derived an expression for the holding time for silver/copper/silver sandwich joints based on stationary solid/liquid interface and their predicted values were found to be highly overestimated compared to the experimental findings. Lee *et al* .[10] suggested that diffusion of the solute atoms into the base metal could actually take place during liquid homogenization, which could result in the formation of second phase precipitates and thus the holding time required for complete isothermal solidification would be considerably reduced. Other models based on migrating solid/liquid interface and Fick's second law of diffusion have been used by several researchers [2,4,12-14] to predict the isothermal solidification completion times and the formation of second phase precipitates in the substrates for pure nickel, nickel based single crystal superalloys and Inconel 738 base metals with binary Ni-P and ternary Ni-Cr-B filler alloys, respectively, and good agreement with the experimental values have been reported. However, modeling studies and experimental investigations of isothermal solidification during TLP bonding of Inconel 625 and 718 superalloys with a multicomponent filler alloy, such as BNi-2, could not be found in literature.

Although TLP bonding is an excellent bonding technique, the time required to complete isothermal solidification is usually long enough to discourage their potential applications in many industries. Therefore, a better understanding of the effect of other process variables, such as bonding temperature and joint gap, on the time required to complete isothermal solidification, is imperative to reduce the time requirement and thus to optimize the process.

The objectives of this work are, thus, to calculate the time required to complete isothermal solidification and to study the effect of process variables during the TLP bonding of Inconel 625 and 718 superalloys with nickel based filler alloy BNi-2, and to verify the predicted values with experimental investigations.

EXPERIMENTAL PROCEDURES

This research was conducted on both wrought Inconel 625 and 718 alloys. Wedge shape joint gap specimens with identical base alloys, shown in Figure 1, were utilized with a relative movement of 4 mm from each other to form an edge groove where the BNi-2 brazing filler paste was placed. The specimen was fixed by tack welds to form a variable brazing gap (0 – 250 μm).

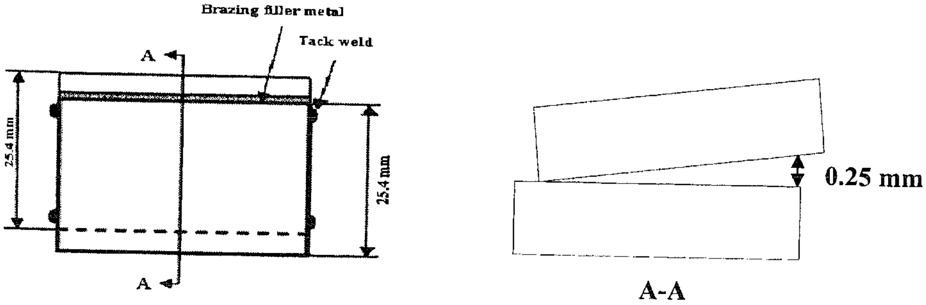


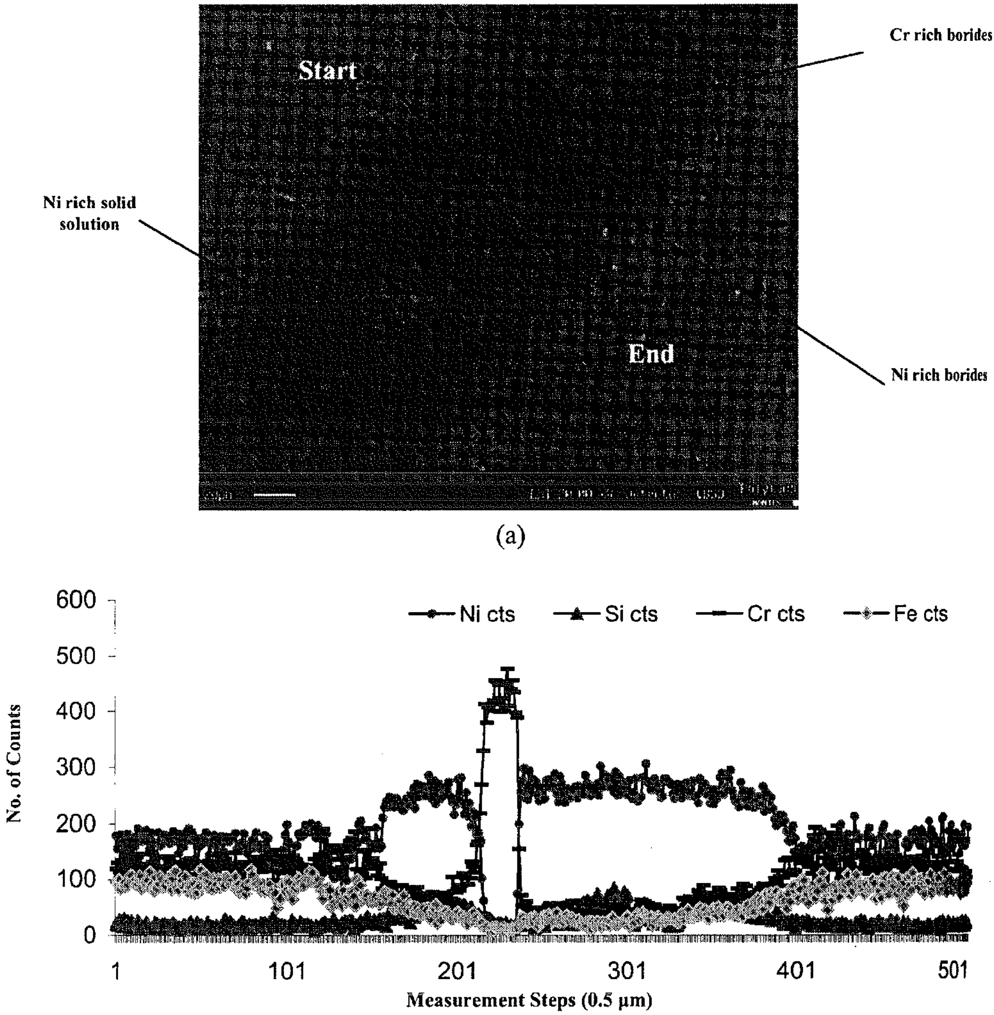
Figure 1 - The wedge shape joint gap specimen

The samples were nicro-blasted and then acid cleaned. To prevent the oxide build-up, the base alloys were pre-plated with very thin layer of nickel (nickel flash) and subsequently vacuum brazed in an argon atmosphere at a vacuum pressure of 106.6 Pa according to the matrix shown in Table 1. The brazed samples were prepared metallographically and studied under optical and scanning electron microscope (SEM) equipped with electron dispersive spectrometry (EDS) at Concordia University and the Université Polytechnique de Nantes, France.

Table 1 - Braze tests matrix

Temp. (K)	Holding Time (min)						
1325	10			50	60	70	
1358			30	50		70	90
1394	10	20	30	50			90

MICROSTRUCTURES OF THE BRAZED JOINT



(b)

Figure 2 - (a) SEM micrograph of Inconel 625/BNi-2 joint brazed at 1325K for 10 minutes showing centerline eutectics, (b) EDS number of counts versus the measurement step

A typical micrograph of the Inconel 625/BNi-2 brazed joint and the corresponding EDS analyses are shown in Figure 2. Intermetallic phases were formed along the centerline of the joint as the part was cooled before the isothermal solidification finished. The residual liquid that was present at the end of the temperature holding eventually transformed on cooling into eutectic constituents. EDS analyses suggest that the phase marked X1 is the pro-eutectic γ- nickel solid solution and the phases marked with X2 and X3 are Cr and Ni rich borides, which are in agreement with the findings of other researchers [2,3,5,15].

From the Ni-Si phase diagram [16], it is evident that Ni dissolves an average of 15 mol% Si over the brazing temperature range (1325K to 1394K), and thus it is expected to have little or almost no silicides. However, EDS compositional analyses in Figure 2 revealed a significant amount of silicon in the center of the joint that might form nickel silicides, which is in accordance with the findings of Jang *et al.* [5]. This can be understood from the following solidification phenomenon [3]: during brazing, γ- nickel first solidified isothermally from the faying surfaces into the melt. Upon cooling the primary γ- nickel solidified as nodular dendrites which enriched the remaining melt with boron, silicon and chromium. As cooling proceeded, binary eutectic of γ- nickel and nickel boride is encountered, further enriching the melt of chromium. Subsequently, binary eutectic of γ- nickel and chromium boride occurred. The melt, which is further enriched in silicon, will then transform into the ternary eutectic of γ-nickel, nickel boride and nickel silicides. Similar solidification phenomena are expected for the Inconel 625 and 718 superalloys with BNi-2 filler alloy when the holding times are not long enough to complete isothermal solidification.

MATHEMATICAL MODELING OF ISOTHERMAL SOLIDIFICATION TIME

Boron Diffusion Model

For unsteady state diffusion of a specie from a source with initial thickness *2w*, into a semi-infinite substrate, solute distribution in the substrate is represented by [17]:

$$C_{(y,t)} = C_m + \frac{1}{2}(C_0 - C_m)\left\{ erf\,\frac{y+w}{\sqrt{4Dt}} - erf\,\frac{y-w}{\sqrt{4Dt}} \right\} \qquad (1)$$

where, C_m = initial solute concentration in the base metal; C_o = initial solute concentration in the interlayer; $C_{(y,t)}$ = solute concentration as a function of distance from the centre of the interlayer (y) and time (t); D = diffusion coefficient of the solute in the substrate.

Holding time can be estimated considering the fact that isothermal solidification is completed when the solute concentration at the centre of the interlayer is reduced to the solidus value C_α. Substituting $C_{(y,t)} = C_\alpha$ at $y = 0$ yields the following equation:

$$C_\alpha - C_m = (C_0 - C_m)\left\{ erf \frac{w}{\sqrt{4Dt_f}} \right\} \tag{2}$$

where, t_f is the time required to complete isothermal solidification.

For the Inconel 625/BNi-2 and Inconel 718/BNi-2 combinations, the values of activation energies and frequency factors for diffusion of boron into the base alloys were found to be 154 $kJmol^{-1}$ and 0.0002 m^2s^{-1}, and, 184 $kJmol^{-1}$ and 0.003 m^2s^{-1}, respectively.

Migrating Solid/liquid Interface Model

Modeling based on migrating solid/liquid interface gives the following expression [4]:

$$t_f^{1/2} = \frac{2h}{\gamma * 4 * D^{1/2}} \tag{3}$$

where, t_f is the time required to complete isothermal solidification, D is the diffusion coefficient, $2h$ is the maximum width of the brazed joint and γ is a dimensionless parameter that accounts for the moving boundary which can be determined from the following expression:

$$\frac{C_\alpha - C_m}{C_\beta - C_m} = \gamma\sqrt{\pi}\exp\gamma^2(1 + erf(\gamma)) \tag{4}$$

where, C_α and C_β are the solute concentration in the solid phase and the liquid phase at the interface, respectively. The value of γ was first calculated by taking C_α and C_β as the average values of boron concentration in solidus (0.3 at%) and liquidus (16.3 at%) of the Ni-B binary system over the brazing temperature range (1325 -1394K).

For $\gamma < 0.1$, a linear relationship exists between the square root of holding time and $2h/D^{1/2}$ [18]. This applies to the current results as can be seen in Figure 3. Nakao *et al.* [19] developed the following linear expression:

$$t_f^{1/2} = J\left(\frac{2h}{D^{1/2}}\right) \tag{5}$$

where J is a constant and is related to γ by equation (3). Plotting the values of $t_f^{1/2}$ against $(2h/D^{1/2})$, as shown in Figure 3, the value of J which is the slope of the straight line can be determined.

For the Inconel 625/BNi-2 and Inconel 718/BNi-2 combinations, the values of activation energies and frequency factors for diffusion of boron into the base alloys were found to be 142 $kJmol^{-1}$ and 0.000447 m^2s^{-1}, and, 168 $kJmol^{-1}$ and 0.0046 m^2s^{-1} respectively. The dimensionless constants *J* for the above mentioned two combinations were 23.9 and 24.2, respectively.

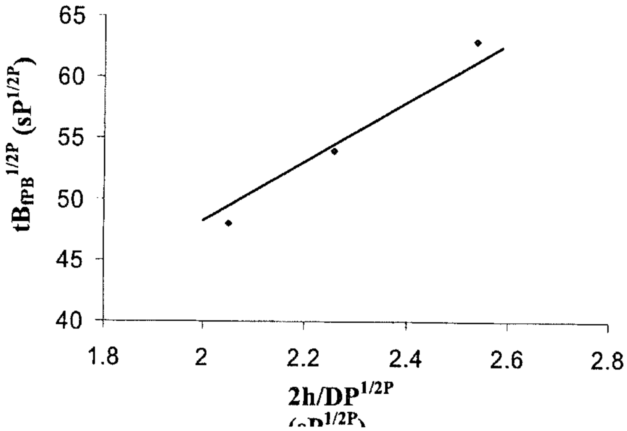


Figure 3 - Square root of holding time vs. $2h/D^{1/2}$ $(s^{1/2})$

EXPERIMENTAL VERIFICATIONS

In the wedge gap brazed joint, a distinction is made between areas of free of brittle phase and brittle phase containing seam sections as shown in Figure 4. The beginning of brittle phase marks the maximum brazing clearance (MBC) for the combination of base metal and filler alloy brazed at a particular temperature and holding time. Figures 5 (a) and (b) show the maximum brazing clearances for the Inconel 625/BNi-2 and Inconel 718/BNi-2 combinations, respectively, brazed at 1325K, 1358K and 1394K with different holding times ranged from 10 to 90 minutes. Conversely, if a specified MBC is taken, the corresponding brazing time will represent the isothermal solidification time for that brazing clearance.

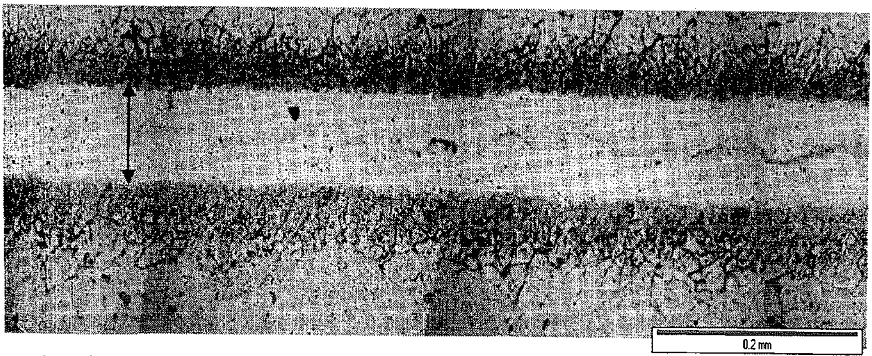


Figure 4 - Micrograph showing the initiation of brittle eutectic phase (Inconel 718/BNi-2 joint brazed at 1394K for 90 minutes)

Significant reduction of holding time was observed with increasing brazing temperatures and with decreasing joint gap. It has been observed that the predicted isothermal solidification completion times were in good agreement with the experimentally determined values, as shown in Figures 5 (c) through (f), with maximum error of ±9%. Besides the physical and chemical uncertainties associated with diffusion experiments, the small deviations could be attributed to the following model assumptions:

(i) The values of C_α and C_β for the complex multi-component melt are not available in literature and were assumed to be 0.3 at%, which is the maximum solubility of boron in nickel and remains almost constant over the bonding temperature range, and 16.3 at%, which is the average concentration of boron in the liquidus of the Ni-B system over the bonding temperature range, respectively. However, boron has a very low solubility in nickel and Ojo *et al.* [2] reported that the presence of substitutional alloying elements, like Cr and Co in nickel based superalloys and Cr in nickel based filler alloys, does not affect this solubility and the value of C_β significantly, and they can be taken as 0.3 at% and 16.3 at%, respectively. Similar approach was also used by Ohassa *et al.* [13] and Rhee *et al.* [20]. (ii) The assumptions associated with Fick's second law of diffusion might have resulted in some errors in the calculations.

The isothermal solidification completion times for Inconel 625/BNi-2 and Inconel 718/BNi-2 were found to be much less than other commonly used combinations, such as Inconel 738/Nicrobraz 150. For a joint gap of 75 μm and 1423K brazing temperature, the isothermal solidification time for Inconel 625/BNi-2 and Inconel 718/BNi-2 combinations are 34 and 38 minutes, respectively, whereas, for the Inconel 738/Nicrobraz 150 combination, it is 4.1 hrs [2].

No significant grain growth in the base metals has been observed in the temperature range being investigated (1325 – 1394K) and the optimum conditions occur when the solute diffusivities in the liquid and solid are increased.

OPTIMIZATION OF PROCESS PARAMETERS

Research works [4,21,22] were carried out to determine the effect of process parameters on the brazed joint quality and to eliminate the brittleness, resulting from the formation of eutectic phases due to incomplete isothermal solidification, as much as possible by changing the process techniques. Several researchers [2,13,14] obtained the activation energies and frequency factors for the diffusion of melting point depressants in the base alloys being used in their studies; however, they could not validate their parameters over a wide range of bonding temperatures and joint gaps due to the lack of experimental data for the isothermal solidification times. Also, neither modeling nor experimental studies of isothermal solidification times for the Inconel 625/BNi-2 and Inconel 718/BNi-2 were found in literature. In this study, the obtained diffusion parameters, such as activation energies and frequency factors, for diffusion of boron into

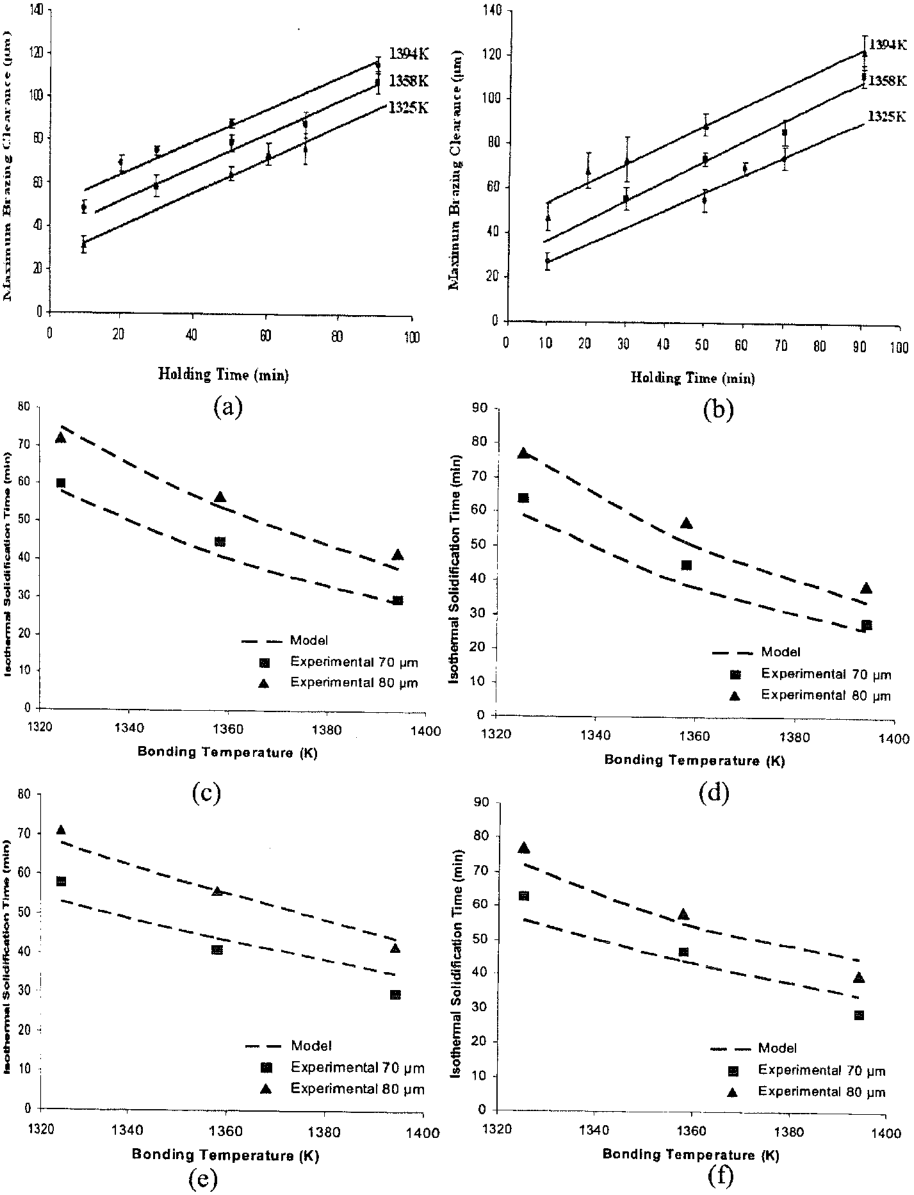
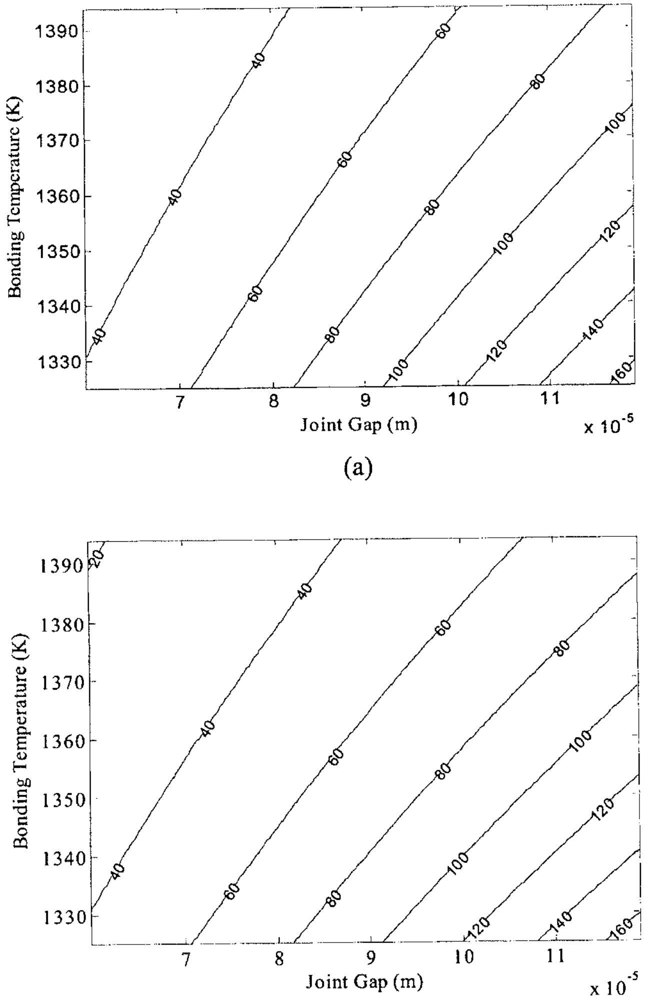


Figure 5 - Effect of holding time on the maximum brazing clearance of (a) Inconel 625/BNi-2 and (b) Inconel 718/BNi-2 combinations; comparison between models and experiments: (c) and (d) Solute distribution law, Inconel 625/BNi-2 and Inconel 718/BNi-2, respectively; (e) and (f) Migrating solid/liquid interface model, Inconel 625/BNi-2 and Inconel 718/BNi-2, respectively



(a)

(b)

Figure 6 – Isothermal solidification time contours (in minutes) for (a) Inconel 625/BNi-2 and (b) Inconel 718/BNi-2 combinations

the Inconel 625 and Inconel 718 base alloys were validated with experimental data over a wide range of bonding temperatures and joint gaps. These parameters were then used to obtain the isothermal solidification time contours, as shown in Figure 6. These contours can be used to select the optimum process parameters for a joint to be solidified isothermally.

SUMMARY AND CONCLUSIONS

Diffusion modeling based on solute distribution law and migrating solid/liquid interface predicted the isothermal solidification times for the Inconel 625/BNi-2 and Inconel 718/BNi-2 combinations successfully.

Isothermal solidification time contours were obtained for both the combinations for the purpose of process parameters optimization. The optimum conditions occur when the solute diffusivities in the liquid and solid are increased.

Unlike other currently used combinations, the isothermal solidification times for Inconel 625/BNi-2 and Inconel 718/BNi-2 were found to be much less. Further, significant reduction of isothermal solidification time has been observed with decreasing joint gap and with increasing bonding temperature.

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