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Mathematical Modeling and Experimental Investigations of Isothermal Solidification during Transient Liquid Phase Bonding of Nickel Superalloys

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Abstract. Mathematical model, based on Fick's second law of diffusion, was used to predict the time required to complete isothermal solidification and to determine the effect of process variables during the transient liquid phase bonding of Inconel 625 and 718 superalloys with nickel based brazing filler alloy BNi-2. Experimental investigations were carried out in the range of 1325 – 1394K to verify the model and the predicted times were in excellent agreement with the experimentally determined values. The obtained activation energies for diffusion of boron were very close to the ones reported for other nickel base polycrystalline superalloys; however, it was observed that the time required for complete isothermal solidification are significantly less than that of other nickel based superalloys with different nickel based brazing filler alloys. Because of this advantage, these combinations of base and filler alloys are expected to replace other currently used ones. Further, significant reduction of holding time was observed with increasing brazing temperature and with decreasing joint gap. The composition of the joints at the end of holding period, when the holding time was not sufficient to complete isothermal soldification, has been determined in order to predict the amount of brittle eutectic phases in the final joint microstructures.

Introduction

Inconel 625 and 718 superalloys are extensively used in aero-engine hot section components and in many other applications that require excellent strength and ductility at elevated temperature. However, because of their susceptibility to heat affected zone cracking during welding [1], transient liquid phase bonding evolved as an effective way to join these superalloys. TLP bonding is a derivative of high temperature brazing [2]; however, unlike conventional brazing, the thermal exposure used for the TLP bonding cycle is sufficient to induce the complete isothermal solidification to avoid the formation of deleterious eutectic phases. Thus, it is imperative to know the isothermal solidification time for the combination of base and filler alloys being used.

Tuah-Poku *et al.* [3] derived the 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 overestimated compared to the experimental findings. Lee *et al.* [4] suggested that diffusion of the solute 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 [5-6] and Fick's second law of diffusion have been used by some researchers [7-9] to predict the isothermal solidification completion times for pure nickel, nickel base single crystal superalloys and Inconel 738 base metals with binary Ni-P and ternary Ni-Cr-B filler alloys, respectively, and good agreements with the experimental values have been reported. However, TLP bonding associated with a multicomponent filler alloy, such as BNi-2, and for Inconel 625 and 718 base alloys 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 brazing 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 the isothermal solidification and to study the effect of process variables during the TLP bonding of Inconel 625 and 718 superalloys with nickel based brazing filler alloy BNi-2, and to verify the predicted values with experimental investigations.

Experimental Procedures

In this work, wedge shape joint gap specimen model, shown in Fig. 1, was utilized. The specimen models consisted of two identical wrought Inconel 625 and 718 alloys 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).

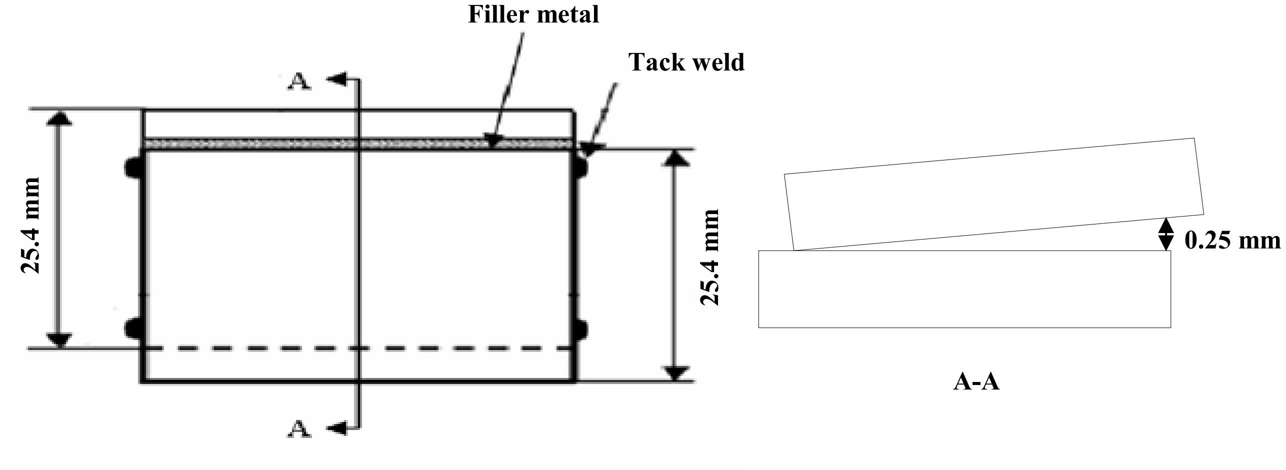


Fig. 1 The wedge shape joint gap specimen

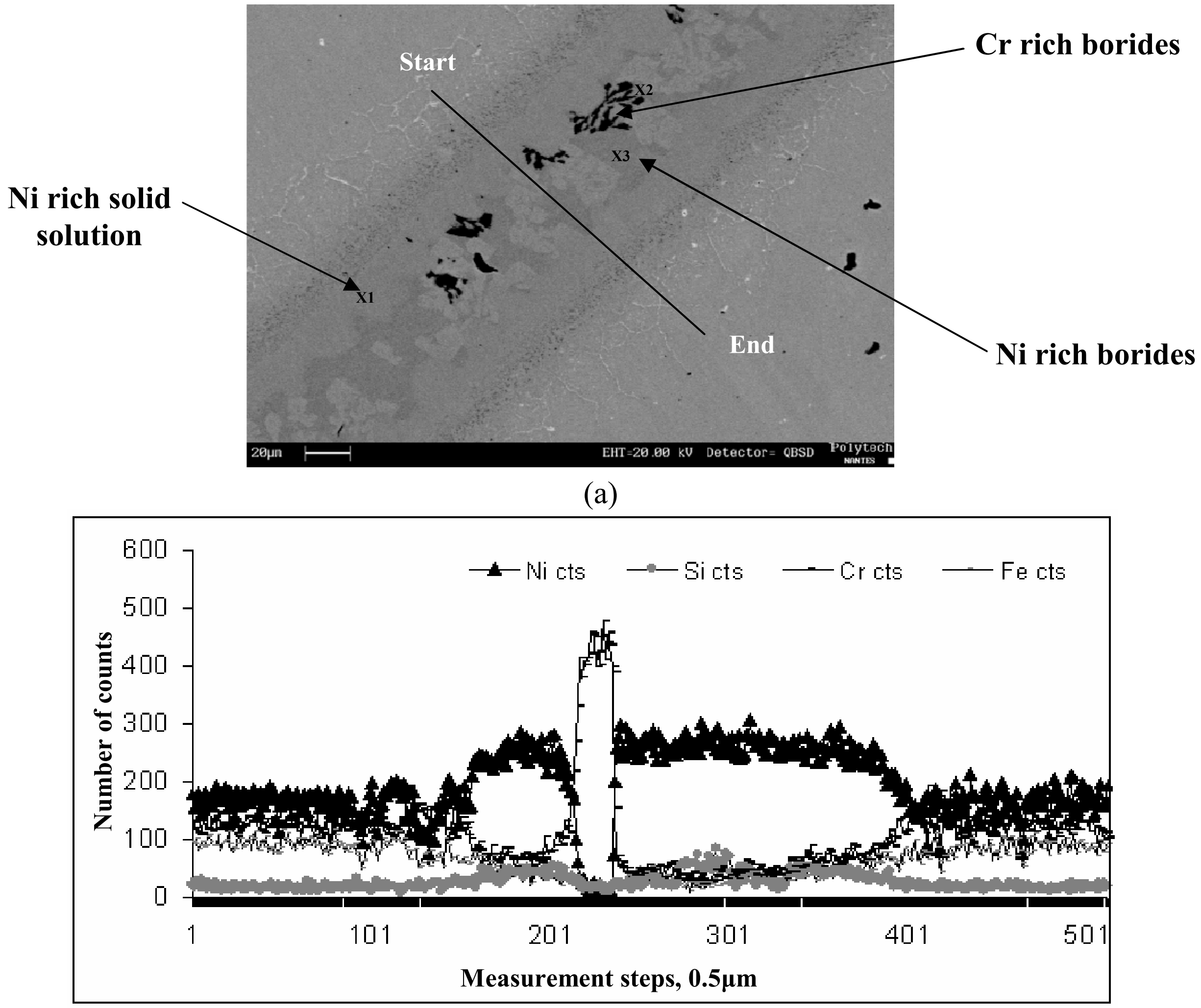
The samples were nicro-blasted and then acid cleaned. To prevent the oxide build-up, the base alloy was pre-plated with very thin layer of nickel (nickel flash) and subsequently vacuum brazed at a vacuum pressure of 106.6 Pa in an argon atmosphere according to the matrix shown in Table 1. The brazed samples were prepared metallographically and studied under the 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	90
1358			30	50		70	90
1394	10	20	30	50			90

Microstructures of the Joint

SEM micrograph of the Inconel 625/BNi-2 joint brazed at 1325K for 10 minutes 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.



(a)

(b)

Fig. 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 steps (0.5 µm).

Mathematical Modeling

For the unsteady state diffusion of a specie present in 2w thick region, into a semi-infinite substrate, solute distribution in the substrate is represented by [10]:

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

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_s. Substituting $C_{(y,t)} = C_s$ at y = 0 yields the following equation:

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

where, t_f is the time required to complete isothermal solidification. Using the experimental isothermal solidification times for two different holding temperatures, the activation energy and frequency factor for diffusion of solute atoms into the base alloy can be determined using the following equation:

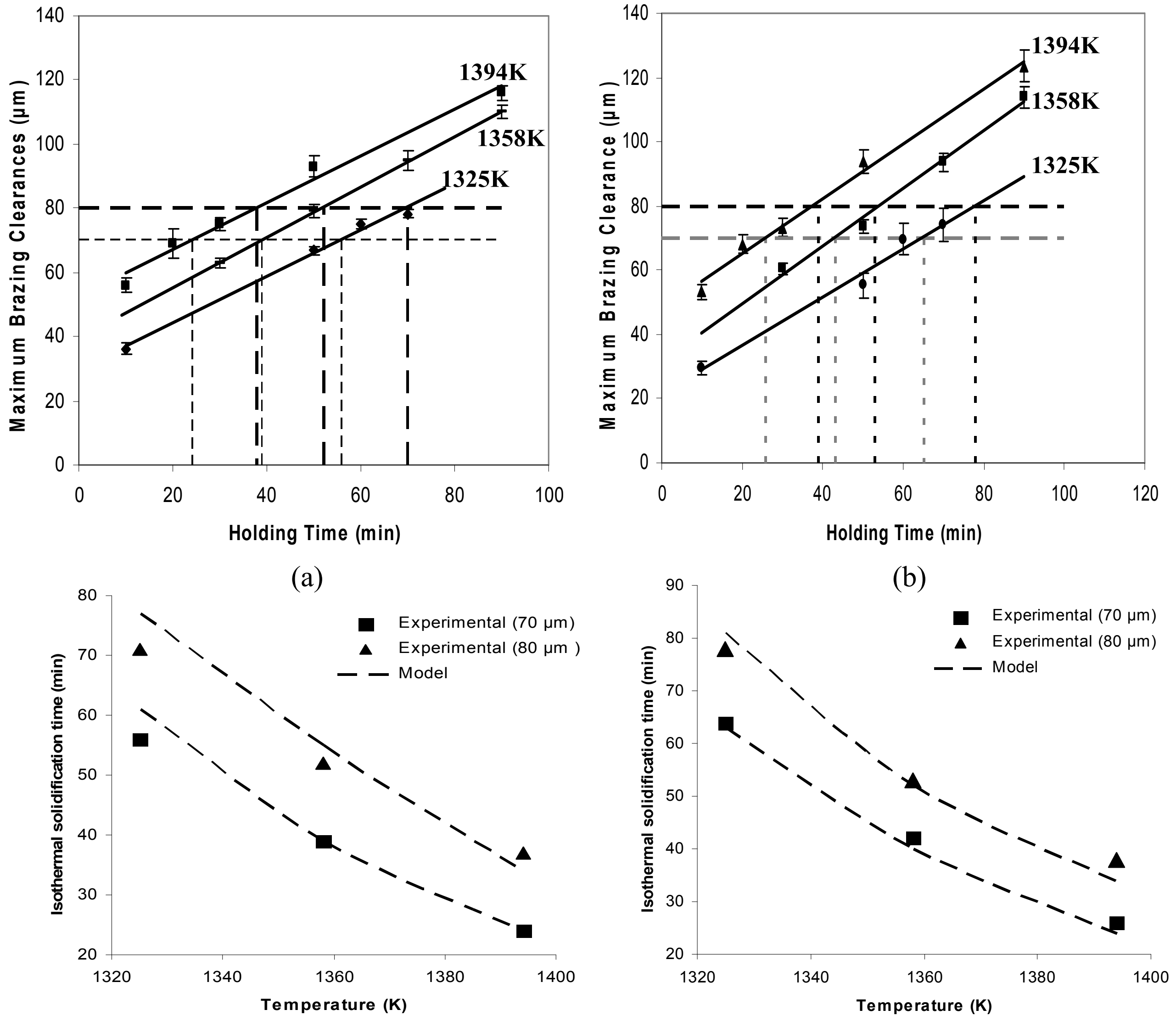
$$D = D_0 \exp\left(-\frac{Q}{RT} \right). \tag{3}$$

Experimental Verification of the Model

In the wedge gap brazed joint, a distinction is made between areas of free of brittle phase and brittle phase containing seam sections. The beginning of brittle phase stabilization marks the maximum brazing clearance (MBC) for the combination of base metals and filler alloy brazed at a particular temperature and holding time. Figure 3 (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.

For the Inconel 625/BNi-2 combination, the values of activation energy and frequency factor for diffusion of boron into the base alloy were found to be 211 kJ/mol and 0.033 m^2s^{-1}, respectively. Figure 3 (c) shows the comparison between the predicted and experimentally determined values of isothermal solidification times for the joints with initial gaps of 70 and 80μm, brazed at 1325, 1358 and 1394K. The predicted values were in good agreement with the experimental findings with an error of ±8%. These small deviations could be attributed to the following model assumptions: (i) The value of C_s was taken as 0.3 at% to simplify the variation of boron solubility in the multi-component melt, which is the maximum solubility of boron in the Ni-B binary system. An increase in boron solubility in the multi-component melt will result into overestimation of time requirement for isothermal solidification and vice versa. The predicted times for isothermal solidification at 1358 and 1394K were within the uncertainty limits of the experimental measurements; thus, 0.3 at% boron solubility, for a joint to be solidified isothermally, can be considered reasonable for this temperature range. However, the slight overestimations at 1325K suggest that the solubility limit might have increased a little bit at this temperature; (ii) The assumptions associated with Fick's second law might have contributed some errors to the calculations.

Similar studies were carried out for Inconel 718 and BNi-2 combination. The values of activation energy and frequency factor were found to be 203 kJ/mol and 0.0159 m^2s^{-1}, respectively. These values were used to calculate the isothermal solidification times for an initial joint gap of 70 and 80 μm at 1325K, 1358K and 1394K bonding temperatures and compared with the experimentally determined values. Good agreement was observed as shown in Fig. 3(d).



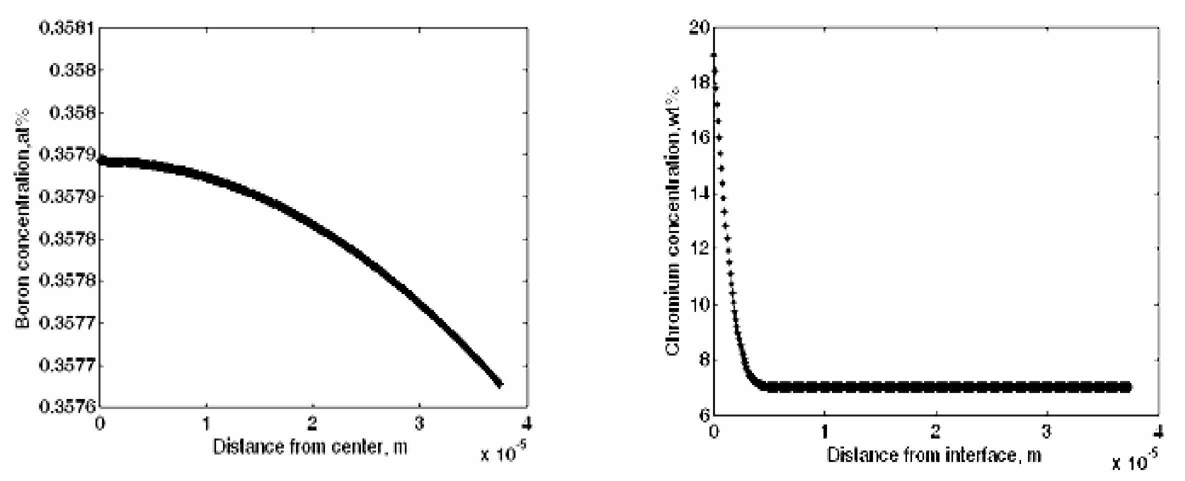
(a) (b)

(c) (d)

Fig. 3 Effect of holding time on the maximum brazing clearance of (a) Inconel 625/BNi-2 and (b) Inconel 718/BNi-2 combinations; comparison between model and experiements for (c) Inconel 625/BNi-2 and (d) Inconel 718/BNi-2.

Computational Thermodynamics

Considerable reduction of time required to complete isothermal solidification has been observed with the increase of brazing temperature and/or with the decrease of initial joint gap for both the combinations. However, when the material limits the temperature and the complex geometry does not allow the joint gap to be narrow enough to have a reduced isothermal solidification time, it is necessary to predict the extent of formation of brittle eutectic phases which are mainly borides with little amount of silicides [2]. Figure 4 shows the composition of boron and chromium in the Inconel 625/BNi-2 joint bonded at 1325K for 50 minutes. It is evident that boron composition is the highest at the center of the joint and gradually decreases towards the joint interface, and, therefore, solidification will proceed from the joint interface towards the center. However, there is a sharp concentration gradient of chromium just beside the interface, as shown in Fig. 4 (b), since the base metal has much higher concentration of chromium (19 wt%) than the filler alloy (7 wt%). Therefore, the solid solution that forms at the end of holding period, in the region close to the joint interface, has a significant higher concentration of chromium than that of the center and, it will result into a change of mechanical properties across the joint.



(a) (b)

Fig. 4 Composition of (a) boron and (b) chromium in the Inconel 625/BNi-2 joint bonded at 1325K for 50 minutes.

Summary and Conclusions

Diffusion modeling was used successfully to predict the isothermal solidification times for the Inconel 625/BNi-2 and Inconel 718/BNi-2 combinations. Predicted values were in excellent agreement with the experimentally determined values.

Experimental isothermal solidification times for both the combinations were obtained for a wide range of holding temperatures and joint gaps. These can be used to optimize the process parameters.

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. It is, thus, expected that these combinations of base and filler alloys will replace other currently used ones in many industrial applications.

The obtained activation energies and frequency factors for diffusion of boron can also be used to determine the isothermal solidification times during TLP bonding of Inconel 625 and 718 base alloy with any nickel based filler alloy that contains boron as melting point depressant.

The concentrations of the constitutive elements across the joint at the end of holding period, when the holding time was not long enough to complete isothermal solidification, were calculated. The solid solution that forms at the end of holding period, in the region close to the joint interface, has a significant higher concentration of chromium than that of the center.

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