Influence of process parameters on microstructure of transient liquid phase bonded Inconel 738LC superalloy with Amdry DF-3 interlayer

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Abstract

The effect of bonding temperature and time on microstructure of diffusion brazed joint of nickel base superalloy Inconel 738LC using Amdry DF-3 filler, alloy was investigated. It was observed that the formation of eutectic microconstituents, within the joint regions, was significantly influenced by the brazing temperature and time. A deviation from the conventional transient liquid phase (TLP) bonding diffusion models was observed in samples brazed above 1175 °C. The rate of isothermal solidification was substantially reduced at this brazing temperature, and also resulted in the formation of a centerline eutectic microconstituent, which was different from that observed at lower bonding temperatures. It is suggested that a probable factor contributing to the change in isothermal solidification rate and the formation of a different type of eutectic microconstituent from that observed at lower temperatures, is the considerable enrichment of the liquated insert with Ti atoms from the base alloy matrix, since they normally exhibit a lower solidification partition coefficient in nickel based alloys.

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1. Introduction

Aero and industrial gas turbines are generally designed within the limits of current technology for the best possible efficiency and output, and as such, complex shaped parts made of suitable materials are needed to withstand the stringent demands encountered in these engines. Inconel 738LC, which is a gamma prime precipitation strengthened nickel based superalloy, is extensively used in hot sections of aero-engines and land based power generation turbines due to its excellent elevated temperature strength and hot corrosion resistance [1]. Typical applications include stator parts such as nozzle guide vane, blades and integral wheels [1,2]. This alloy, like other gamma prime precipitation strengthened superalloys that contain a substantial amount of Ti and Al, is very difficult to weld because of its high susceptibility to heat affected zone (HAZ) cracking. Transient liquid phase (TLP) bonding [3], also known as diffusion brazing, has evolved into an attractive alternate joining technique for difficult to weld alloys like Inconel 738 superalloy due to its technological and economic advantages. Nevertheless, proper optimization of various process parameters, and an appropriate choice of filler metal, is paramount to achieving sound and reliable joints for high temperature applications in this and other γ′ precipitation hardened superalloys. Formation of eutectic constituents and/or other second phase particles in a continuously distributed fashion either along the central region of the joint or at the base metal–braze interface are often found to be deleterious to the properties of superalloy brazements [4].

Several models of TLP bonding, most of which are based on binary alloy systems, have been developed and proposed to explain the process controlling mechanisms. However, the phase relationships that are actually encountered in commercial joining of complex multicomponent alloys do not always lend themselves to an extrapolation of the binary analysis. Therefore, an explicit consideration of diffusion brazing parameters, namely temperature and time, which directly influence microstructural evolution during bonding, is imperative to producing joints with properties consistent with that of the base multicomponent alloy. It was thus the objective of the present research work to investigate the influence of diffusion bonding temperature and time, on microstructure of diffusion brazed Inconel 738 superalloy using commercial brazing filler alloy Amdry DF-3.
2. Experimental procedures

The base alloy used in this study was as-cast Inconel 738LC in the form of 0.6 cm × 2.5 cm × 15.0 cm plates supplied by Hitchiner Manufacturing Co., Inc. The chemical composition of the alloy was (wt%) 0.11C, 15.84Cr, 8.5Co, 2.48W, 1.88Mo, 0.92Nb, 0.07Fe, 3.46Al, 3.47Ti, 1.69Ta, 0.04Zr, 0.012B and balance nickel. 0.6 × 1.5 × 0.6 coupons were used in the as-received without any pre-braze heat treatment. TLP bonding (diffusion brazing) was performed using 75/H9262 m thick amorphous foil of Amdry DF-3 filler alloy with a composition of (wt%) 3B, 20Cr, 20Co, 3Ta, 0.05La and balance nickel. The specimens were vacuum diffusion brazed in a LABVAC II brazing furnace under a vacuum of 10^{-4} Torr to 10^{-5} Torr at 1120 °C and 1160 °C for 30 min, 290 min, 420 min, and at 1175–1225 °C for 290 min, 420 min and 720 min. The brazing cycle employed is shown in Fig. 1.

Brazed samples were sectioned perpendicular to the joint by electro-discharge machining (EDM) due to the brittle nature of the brazement, and prepared by standard metallographic techniques for microstructural examination by optical and scanning electron microscopy (SEM). Cross sections of brazed joints were etched electrolytically in 12 ml H₃PO₄ + 40 ml HNO₃ + 48 ml H₂SO₄ solution at 6 V for 5 s. Preliminary general assessment of the joint microstructure was performed by an inverted-reflected light microscope equipped with a CLEMEX Vision 3.0 image analyzer. Further scanning electron microscopic microstructural examination, using secondary and backscatter electron imaging modes, and compositional analysis of brazement were conducted on a JEOL 5900-LV scanning electron microscope, equipped with a thin window Oxford energy dispersive spectrometer (EDS) system equipped with INCA software. Microhardness of the centerline region of the brazed joints was determined by a Leitz microhardness tester at 100 g load.

3. Results and discussions

3.1. Effect of diffusion time on brazement microstructure

To study the effect of holding time on the brazement microstructure, brazing was done at 1120 °C and 1160 °C for 30 min, 290 min and 420 min. The microstructure of the samples brazed at these temperatures, as obtained by SEM operated in secondary electron mode, is shown in Figs. 2 and 3. The microstructure of the joint produced after 30 min holding time at these temperatures showed the occurrence of a continuous centerline eutectic constituent. The average eutectic thickness, after this holding time, decreased from 20.3 μm in the 1120 °C sample to 14.9 μm in the 1160 °C sample. EDS compositional analysis of the three main phases observed within the eutectic, given in
Table 1. Composition of eutectic constituents formed in the first regime of isothermal solidification

<table>
<thead>
<tr>
<th>Element</th>
<th>Cr-rich boride (at.%)</th>
<th>Ni-rich boride (at.%)</th>
<th>Gamma (γ) (at.%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni</td>
<td>4.87</td>
<td>62.35</td>
<td>57.63</td>
</tr>
<tr>
<td>Cr</td>
<td>86.09</td>
<td>10.41</td>
<td>18.64</td>
</tr>
<tr>
<td>Co</td>
<td>7.86</td>
<td>19.96</td>
<td>17.99</td>
</tr>
<tr>
<td>Al</td>
<td>–</td>
<td>–</td>
<td>1.42</td>
</tr>
<tr>
<td>Ti</td>
<td>–</td>
<td>3.42</td>
<td>1.86</td>
</tr>
<tr>
<td>Nb</td>
<td>–</td>
<td>0.62</td>
<td>0.30</td>
</tr>
<tr>
<td>Ta</td>
<td>–</td>
<td>2.77</td>
<td>1.29</td>
</tr>
<tr>
<td>W</td>
<td>0.54</td>
<td>0.47</td>
<td>0.39</td>
</tr>
<tr>
<td>Mo</td>
<td>0.64</td>
<td>–</td>
<td>0.48</td>
</tr>
</tbody>
</table>

Fig. 3. SEM micrographs of specimens brazed at 1160°C for (a) 30 min, (b) 290 min and (c) 420 min.

Table 1, suggested them to be nickel base solid solution phase, a nickel rich boride phase and a chromium rich boride phase. On increasing the holding time at the bonding temperatures there was a considerable reduction in the width of the eutectic constituent, and after 290 min and 420 min the centerline region of the samples contained only a discontinuous distribution of chromium rich boride particles (Figs. 2b and c, and 3b and c). Boron was detected in all the boron rich particles, as seen in the EDS spectra shown in Fig. 4, but could not be quantified because of the inability of the software to quantify light elements accurately. In addition, extensive precipitation of globular and acicular particles was observed in the brazed samples within the base alloy grains and intergranular regions adjacent to the joint interface, as shown in Fig. 5. In another study, the X-ray microanalysis of some of these particles by SEM and TEM suggested them to be Cr-rich boride/borocarbide [5]. Quantitatively, the number density of these particles was observed to decrease in a gradual fashion with increasing distance from the joint interface.

In the TLP diffusion bonding process, the interlayer alloy melts and reacts with the base metal, which involves base alloy dissolution, and forms a liquid phase. This phase rapidly attains equilibrium with the solid base metal. Subsequent to this, due to the inter-diffusion of alloying elements between the base metal and the liquid, the melting point of the interlayer liquid at the liquid–solid interface starts to increase and isothermal solidification of the liquid at the interface begins. As the melting point depressing solute diffuses continuously into the base metal, the volume of liquid that can be maintained at equilibrium decreases, causing solidification to proceed towards the center of the joint from the mating solid surfaces. Due to the absence of solute rejection at the solid–liquid interface during isothermal solidification under equilibrium the only solid phase which forms is the solid solution phase, and the formation of other phases is basically prevented [3]. However, in situations where sufficient holding time for complete isothermal solidification is not allowed, the residual liquated insert could transform during cooling from the bonding temperature into a eutectic-type solidification product.

Ohsasa et al. [6], by use of Scheil simulations, studied the solidification behavior of residual liquid during TLP bonding of nickel with Ni–B–Cr ternary filler metal. They reported that ternary invariant centerline eutectic, consisting of Ni base FCC solid solution phase (γ), Ni boride (Ni3B) and Cr boride (CrB), formed at 997°C. Their simulation showed that during solidification of the residual liquid in a sample held at 1100°C, Ni-rich γ phase formed as the primary phase, followed by the eutectic reaction L → γ + Ni3B at 1042°C. Solidification was reported to be completed with a ternary eutectic reaction L → γ + Ni3B + CrB at 997°C. In addition, Gale and Wallach [7] in their work on TLP bonding of nickel using Ni–Si–B ternary filler alloy...
observed, with the aid of high temperature X-ray diffraction, a deposit of Ni + Ni3B eutectic mixture in the sample brazed at 1150 °C for 5 min. In the present work, the centerline eutectic constituent is believed to have formed by solidification reaction(s) during cooling due to an insufficient diffusion time for a complete isothermal solidification during holding at the brazing temperature. Formation of such a eutectic-type product with a continuously distributed morphology would be deleterious to the mechanical properties of brazed joints, as it would provide a low resistance path for crack initiation and/or propagation, as can be seen in Fig. 2a, and should therefore be avoided.

The isothermal solidification is considered to be complete when the concentration of the melting point depressant solute at the center of the liquated interlayer is reduced to the value of concentration of the melting point depressant solute at the solvus, \( C_s \). Gale and Wallach [8] and Ojo et al. [9] have shown that the time required to achieve a complete isothermal solidification can be predicted by the following equation:

\[
C_s - C_m = C_0 - C_m \left\{ \text{erf} \left( \frac{w}{\sqrt{4D t_f}} \right) \right\}
\]

where \( C_m \) is the initial solute concentration in the base metal, \( C_0 \) the initial solute concentration in the interlayer, \( w \) one half the initial thickness of the braze interlayer, \( D \) the diffusion coefficient of the solute in the substrate and \( t_f \) is the time required to achieve complete isothermal solidification.

Ojo et al. [9] also used an alternate approach based on a moving solid–liquid interface boundary model to reasonably estimate the value of \( t_f \) by the following expression:

\[
t_{1/2}^{1/2} = J \left( \frac{2b}{D^{1/2}} \right)
\]

where \( J \) is a constant and \( 2b \) is the maximum width of the liquid insert following homogenization of the liquid. According to Eqs. (1) and (2), it can be seen that the time required to obtain a sound joint devoid of centerline eutectic, by a complete isothermal solidification, is expected to be strongly dependent on the bonding temperature. An increase in diffusion brazing temperature is expected to significantly reduce \( t_f \) and produce a joint without the deleterious centerline eutectic. To investigate this hypothesis, diffusion brazing was done at higher temperatures, as described next.
3.2. Effect of diffusion temperature on brazement microstructures

Specimens with 75 µm gap interlayers were brazed at 1175 °C and 1190 °C for 290 min and 420 min, using the same brazing scheme described earlier. SEM micrographs of these brazed joints are shown in Figs. 6 and 7. It can be seen that an almost complete isothermal solidification was achieved in the sample brazed at 1175 °C for 420 min. This is as predicted by the TLP bonding model that an increase in the rate of isothermal solidification would occur with an increase in brazing temperature. Also, fine γ′ precipitate particles were observed in 1175 °C and 1190 °C specimens within the isothermally solidified austenitic matrix (Fig. 8). These would have formed by solid state precipitation reaction during cooling due to an increased concentration of Ti and Al in the interlayer liquid as an increased amount of Ti and Al would have diffused into the region.

On increasing the brazing temperature to 1190 °C, a significant departure in the microstructure of the joint was observed to occur from that predicted by the conventional TLP models. Continuously distributed centerline eutectic constituents were observed to have formed in samples brazed for 290 min and 420 min at this temperature (Fig. 7). This implied that more residual liquid was present in these joints than that was present after the equivalent holding times at lower temperatures (1120 °C, 1160 °C and 1175 °C). Analytical SEM-EDS examination showed that in addition to Cr-rich boride particles, the centerline eutectic product in the 1190 °C brazed specimen contained Ni–Ti-rich intermetallic particles (Fig. 9, Table 2).
This suggests that significantly more enrichment of the residual liquid with Ti atoms occurred at 1190 °C. Fine γ′ eutectic constituents which are known to form due to the enrichment of the liquid by Ti [10], were also observed as part of the re-solidified product (Fig. 9). The average concentration of Ti in the center region of the joints brazed for 290 min at various temperatures is plotted in Fig. 10. It is seen from this plot that the rate of increase in Ti concentration in the centerline region of the joint increased considerably above 1175 °C. This temperature is close to the solvus temperature of Ti-rich γ′ precipitate particles, and their dissolution in the base metal would allow more Ti atoms to be available for diffusion into the brazed joint [10]. Correspondingly, the increase in concentration of Ti, which is one of the main constituent of the principal hardening phase of this alloy, is corroborated by the observed increase in hardness of the centerline region of the joint with an increase in temperature, as shown in Fig. 10.

In order to investigate further the effect of bonding temperature on the change in isothermal solidification rate, specimens were brazed at 1175 °C and 1225 °C for 12 h. As can be seen in Fig. 11(a and b) a complete isothermal solidification occurred in specimens brazed at 1175 °C for 12 h, while the same brazing time resulted in an incomplete isothermal solidification in

### Table 2

Composition of Ni–Ti-rich eutectic constituents formed in the second regime of isothermal solidification

<table>
<thead>
<tr>
<th>Element</th>
<th>Ni–Ti-rich phase (at.%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni</td>
<td>63.04</td>
</tr>
<tr>
<td>Co</td>
<td>5.11</td>
</tr>
<tr>
<td>Al</td>
<td>10.66</td>
</tr>
<tr>
<td>Ti</td>
<td>11.63</td>
</tr>
<tr>
<td>Nb</td>
<td>2.27</td>
</tr>
<tr>
<td>Ta</td>
<td>2.00</td>
</tr>
<tr>
<td>W</td>
<td>0.63</td>
</tr>
<tr>
<td>Mo</td>
<td>0.31</td>
</tr>
</tbody>
</table>

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Fig. 9. (a) SEM micrograph showing eutectic micro-constituent in a sample brazed at 1190 °C for 290 min and (b) EDS spectrum of Ni–Ti rich phase.

Fig. 10. Variation in concentration of Ti and microhardness of the centerline region of the brazed joints with the bonding temperature.

Fig. 11. SEM micrographs of specimens brazed for 720 min (12 h) at (a) 1175 °C and (b) 1225 °C.
1225 °C specimens. Again, this is a significant departure from conventional expectations that an increase in brazing temperature would produce more diffusion of the melting point depressant (B) into the base metal and thus results in a higher isothermal solidification rate. Sinclair et al. [11] have suggested that a deviation from the isothermal solidification rate predicted by the conventional TLP diffusion models, which are generally based on binary systems, can be encountered during TLP bonding of multicomponent alloys. It was noted that if the solubilities and/or diffusion coefficients of the two diffusing solutes capable of controlling the TLP bonding isothermal solidification process (normally melting point depressants) are very different, isothermal solidification stage can be divided into two parabolic regimes. The first being dominated by the “faster” solute, and the second by the slower of the two. In extreme cases, a complete isothermal solidification may not be realized in experimentally feasible times.

In the present work, diffusion of boron, which is essentially an interstitial atom in nickel and thus with a higher diffusivity compared with the substitutional solutes, is believed to be controlling the isothermal solidification process in the first regime. However, during brazing process, continual interdiffusion of elements between the liquid insert and the base metal would cause the composition of the liquid to be continually modified as solidification progresses. At 1190 °C a considerable enrichment of the liquated insert with the base alloy element Ti would occur due to an increased diffusion of this element caused by its high concentration gradient and increased diffusion coefficient with increased temperature. The concentration gradients of this element between the base alloy γ matrix and the liquated insert is expected to be very steep, since the latter is enriched with Ti atoms owing to the dissolution of γ′ (Ni3Al, Ti) at this temperature [12], while they are absent in the filler alloy. The solidification partition coefficient of Ti in nickel is less than 1 (one), therefore an increase in its concentration would result in a depression of solidification temperature [13]. Consequently, a considerable enrichment of this element in the liquated insert during diffusion brazing at 1190 °C could lead to the commencement of a second solidification regime, which is characterized by a slower isothermal solidification rate, as suggested by Sinclair et al. [11]. A similar deviation in isothermal solidification rate during brazing of Inconel 738LC at higher temperatures, but using a different filler alloy (Nickrobraz 150), has also been reported recently by Idowu et al. [14].

Besides the influence of diffusion temperature on the formation of centerline eutectic, it was also observed, qualitatively, in the present work that an increase in temperature from 1120 °C to 1190 °C resulted in a decrease in the precipitation of chromium rich particles within the base alloy grains adjacent to the brazed joint (Figs. 2c, 3c and 6b). Analytical models of TLP bonding [15–17] treat the joining process to be consisting of a number of sequential discrete steps, namely, melting of the filler alloy, base metal dissolution, isothermal solidification and homogenization stages. It is assumed that due to orders of magnitude of difference in diffusivity of solute on the two sides of the solid–liquid interface, dissolution of the base metal into the liquid layer occurs at a much faster rate than the solid-state diffusion of the melting point depressant in the base metal. Hence, the interlayer alloy upon melting rapidly attains equilibrium with the solid base metal, which involves base alloy dissolution, following which the solid-state diffusion of melting point depressant element commences. Under this condition, boron rich second phase particles are not expected to form during the TLP bonding process, since it is assumed that solid-state solute diffusion in the base metal takes place under equilibrium condition subsequent to liquid–solid equilibration involving base metal dissolution. However, it has been suggested that the two processes occur simultaneously rather than sequentially [18], in which case the boride precipitation would be expected to occur within a region of the base alloy where solute solubility is exceeded as a result of substantial solid-state diffusion of the solute during base metal dissolution.


Under this condition, boride precipitation would be expected to occur within a region of the base alloy where solute solubility is exceeded as a result of substantial solid-state diffusion of the solute during base metal dissolution. Consequently, increase in the bonding temperature could reduce the time available for solid-state solute diffusion during the liquid–solid equilibration process. Perhaps this could be contributing to the observed decrease in the extent of precipitation of chromium rich particles with increase in brazing temperature in the base metal region adjacent to the joint. Considering the high chromium content of the particles, this would lead to a significant depletion of chromium around this region of the substrate, which may result in a decrease in the region’s corrosion resistance. A situation in which corrosion resistance depended essentially on chromium content in the intergranular regions controlled by Cr3B and CrB precipitation in superalloy has been reported [20]. Therefore, it is apparent that there is a need to optimize diffusion brazing parameters (temperature and time) for the Inconel 738 base metal—DF-3 filler alloy system to obtain a balance between the prevention of the formation of interface chromium rich particles with increase in temperature and a re-occurrence of the deleterious centerline eutectic at higher temperatures due to a change in the isothermal solidification rate.

4. Summary and conclusion

1. Diffusion based TLP bonding of Inconel 738 samples with Am dys DF-3 filler alloy and a gap width of 75 μm was done at 1120–1225 °C for various times.

2. Isothermal solidification of the liquated filler metal occurred during holding at the brazing temperatures, the extent of which increased with increase in time.

3. Complete isothermal solidification, which precluded the formation of centerline eutectic constituent, occurred within 12 h holding time at 1175 °C.

4. A deviation from predictions by TLP diffusion models was observed in samples brazed at 1190 °C and 1225 °C. An increased amount of liquid, and hence the amount of centerline eutectic, was present in the joints produced at these temperatures than that present after an equivalent holding time at lower temperatures (1120 °C, 1160 °C and 1175 °C).
5. Increased diffusion of Ti from the base metal into the liquated filler metal at 1190 °C and 1225 °C was suggested to be an important factor contributing to the observed deviation from the conventional expectation of an increase in isothermal solidification rate with increase in bonding temperature.

6. In addition to the formation of centerline eutectic product in brazed samples, precipitation of chromium rich particles was observed in the base metal regions adjacent to the joint interface. Increase in the diffusion brazing temperature resulted in a considerable decrease in the extent of the interface precipitation.

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References
