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Evaluation of transient liquid phase bonding between nickel-based superalloys

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Induction brazing of Inconel 718 to Inconel X-750 using Ni-7Cr-3Fe-3.2B-4.5Si (wt.-%) foil as brazing filler metal was investigated in this paper. Brazing was conducted at the temperature range 1373–1473 K for 0–300 s in a flow argon environment. Both interfacial microstructures and mechanical properties of brazed joints were investigated to evaluate joint quality. The optical and scanning electron microscopic results indicate that good wetting existed between the brazing alloy and both Inconel 718 and Inconel X-750. Microstructures at joint interfaces of all samples show distinct multilayered structures that were mainly formed by isothermal solidification and following solid-state interdiffusion during joining. The diffusion of boron and silicon from brazing filler metal into base metal at the brazing temperature is the main controlling factor pertaining to the microstructural evolution of the joint interface. The element area distribution of Cr, Fe, Si, Ni and Ti was examined by energy dispersive X-ray analysis. It was found that silicon and chromium remain in the center of brazed region and form brittle eutectic phases; boron distribution is uniform across joint area as it readily diffuses from brazing filler metal into base metal. The influence of heating cycle on the microstructures of base material and holding time on the mechanical properties of brazed joint were also investigated.

1. Introduction

Inconel 718 and Inconel X-750 are austenitic superalloys containing precipitation strengthened phases such as MC (NbC, TiC), and $M_{23}C_6$ type ($Cr_{23}C_6$) carbides [1, 2]. They have found wide applications in automobile, aerospace, and nuclear industries based on their mechanical properties, oxidation and corrosion resistance [3, 4]. However, complex component of these superalloys can only be acquired with difficulty using conventional mechanical processing. To utilize the full capability of these alloys, joining techniques yielding high-integrity bonds are required.

Welding, diffusion-bonding and brazing are mainly three joining techniques that have been commonly applied in industry [5, 6]. The applications of traditional fusion welding processes is impractical for Inconel 718/Inconel X-750 because the formation of brittle Nb-Ni compounds and differential expansion coefficient between the base metals contribute to cracking problems in the fusion and heat affected zone (HAZ) of welded region. Although solid-state bonds with rather good joint properties can be acquired by diffusion bonding, the long processing time, high external pressure, and the high corresponding operation/installation costs may render this joining technique useless for practical applications, especially for large-sized workpieces. On the other hand, brazing has also been considered as a useful technique for joining nickel-based superalloys. The principal disadvantage of this method is that the service temperature of a brazed components is constrained to a temperature below the melting point of its filler metal, which is much lower than the usual application temperature of superalloys. Furthermore, the formation of intermetallic phases along the centerline of brazed joint greatly reduces its joint strength.

However, transient liquid phase (TLP) brazing [7–11] is the preferred joining method for nickel- and cobalt-based superalloys, because it has the advantage of not requiring the rather high pressure needed in typical solid state diffusion bonding process and is highly tolerant to the presence of a faying surface oxide layer. As an effective joining process, it can offer many advantages over other joining processes such as relatively low joining temperature, the possibility of making of many joints at one pass, dissimilar-material joining, joining complex structure joining, high precision and better joint properties. Therefore, TLP brazing process is ideal when joining materials are inherently susceptible to hot cracking or post-weld heat-treatment cracking problems.

In TLP brazing, the filler metal is placed between the base metal surfaces and the joining operation is carried out at a temperature between the liquidus of the filler metal and the solidus of the base metal. On holding at the brazing temperature, solute elements in the filler metal diffuse into the base metal and dissolution occurs

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TABLE I Nominal compositions (wt.-%) of base metals and filler metal

Materials	Chemical composition (wt.-%)											
	Ni	Cr	Fe	Mo	Mn	Cu	Si	B	C	Nb	Al	Ti
Inconel 718	Bal.	19.0	18.5	3.0	0.2	0.30	0.2	—	0.04	5.1	0.5	0.9
Inconel X-750	Bal.	15.5	7.0	—	0.5	0.50	0.2	—	0.04	1.0	0.7	2.5
AMS 4777	Bal.	7.0	3.0	—	—	—	4.5	3.2	0.06	—	—	—

prior to isothermal solidification. While the joints are held at the brazing temperature, interdiffusion between the liquid and the base material occurs, leading eventually to isothermal solidification. Maintaining the joint at the brazing temperature after isothermal solidification will homogenize the joint with chemical composition and microstructure closely equivalent to those of the base material [12].

2. Experimental procedure

TLP brazing was conducted in induction furnace under one atmosphere of argon. Nickel-based brazing alloy, AMS 4777, with thickness of 100 μm was selected as the filler metal. The nominal chemical compositions of base metals and filler metal were given in Table I. The contacting surfaces of base metals were polished using 1000 grade emery paper to remove the surface oxide and were then ultrasonically cleaned in an acetone both prior to TLP brazing. All TLP brazing trials were performed at 1423 K.

3. Results and discussion

Once the brazing temperature is reached, the filler metal melts, the liquid, by capillary action, fills the joint clearance and eliminates potential voids at the interface between two parts. The capillary force entails an interaction between the base metal and the filler metal. At first, there are heterogeneous chemical reactions at the solid-liquid interface, whereby atoms accumulate at the interface of the solid and liquid metal. The process is known as wetting. Secondly, some dissolution of the base metal does occur due to interdiffusion between filler metal and base metals. When the composition of the boron and/or silicon in the molten brazing filler metal reaches equilibrium in the interface at the brazing temperature, the dissolution of the solid base metal would stop. Usually, this stage will take a few seconds [2]. Diffusion of boron and/or silicon from the molten brazing filler metal into the base metal would come next. Therefore, when the diffusion of boron and/or silicon from the brazing filler metal to the base metal was observed, the dissolution reaction had finished. There is concentrated equilibrium behind a thin limit zone and, it appeared that the isothermal solidification had started. Nickel, chromium and iron migrated rapidly during progression of the dissolution reaction, but this reaction is finished within a few minutes. After the dissolution reaction was completed, it appears that that the isothermal solidification of nickel, chromium and iron in the base metal slows down because the process is controlled by element diffusion in solid state. However, the boron rapidly diffused into the base metal, and therefore, the eutectic structure disappeared and the elemental distribution of chromium, iron and silicon became more uniform.

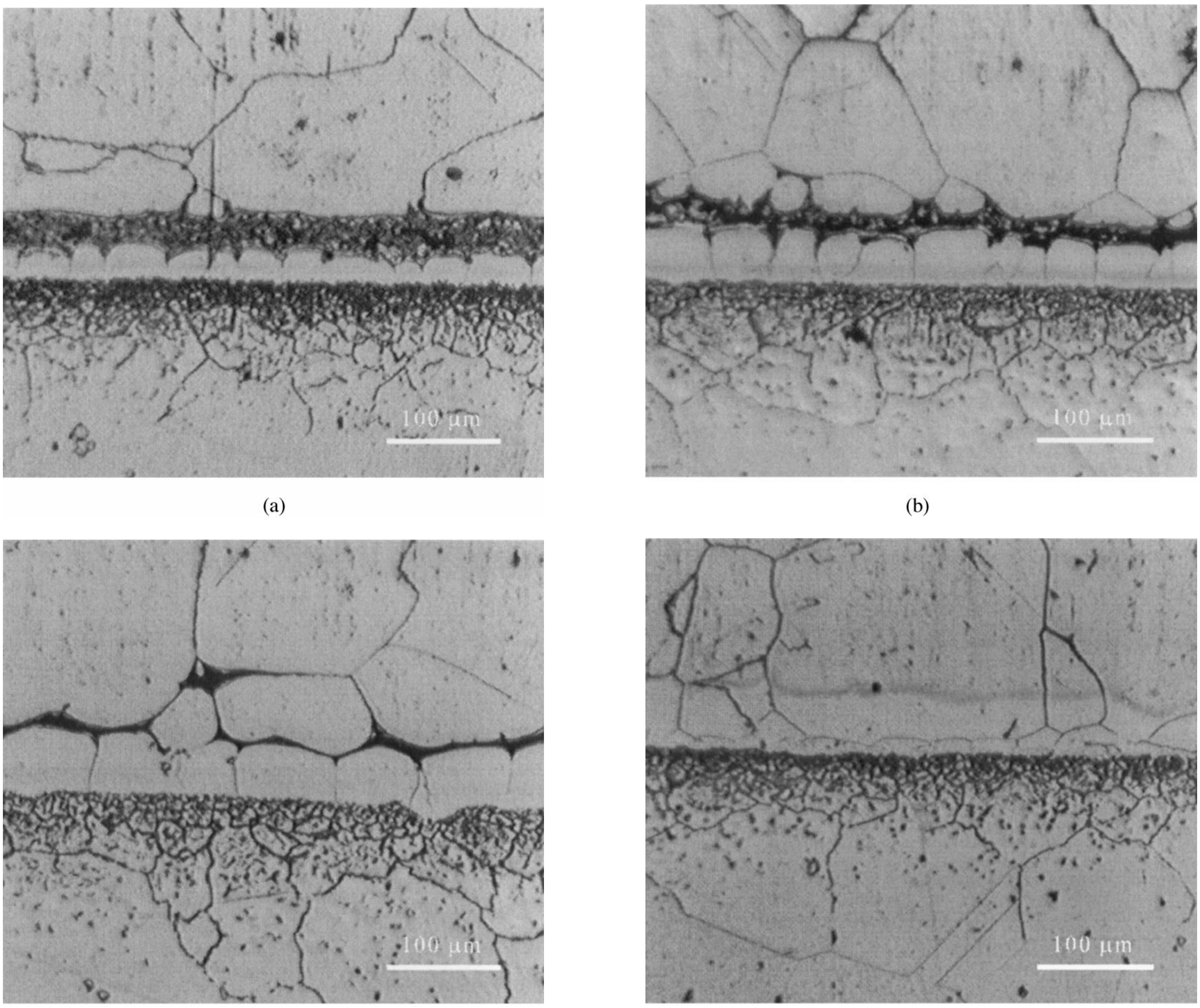
3.1. Typical microstructure of brazed joint

In metallographic terms a "good" joint can be classified as one free from precipitate, free from eutectic phases, free from porosity and with composition and microstructure as similar as possible to that of the parent alloy. Since joint properties are strongly dependent on microstructures, it is important that the microstructure evolution in the joint interface can be elucidated.

The optical micrographs of the TLP joint interfaces for the Inconel 718/Inconel X-750 with AMS 4777 interlayer in test specimens processed at 1423 K for various brazing times are shown in Fig. 1. It is clearly seen that bond microstructures mainly consisted of nickel solid solution and eutectic phases. Intermetallic compounds are formed along the centerline of a joint if the part was cooled down before the isothermal solidification finished. Therefore, it is quite clear that the amount of the brittle phases formed in the joint decreased with increasing brazing. The bond microstructures showed little change with hold time, but the thickness of the eutectic phases was seen to decrease noticeably. Initially, this decrease in thickness was considered that filler metal diffuse into the parent metals. Shorter bonding times resulted in the formation of borides of nickel, iron and chromium along filler metal interface. Fig. 1a displays that large amount of eutectic phases remains in the centerline of bond after holding 10 min at TLP brazing temperature. When brazing time was prolonged to 30 min, the amount of eutectic phase in the brazed joint continued to decrease and solid nickel solution grew into the liquid phase. If brazing was processed for 60 min, there still existed somewhat eutectic phase in the bond region. Further increase in brazing time to 120 min resulted in complete isothermal solidification; the intermetallic phases had completely dissolved into solid solution and the brazed joint merely consisted of nickel solid solution as shown in Fig. 1d.

It is evident that some erosion happens in the side of Inconel X-750 base metal (upper part of Fig. 1). Grain boundaries adjacent to joint interface are wetted by liquid filler metal. Liquation penetration can be observed at the distance of 300 μm away from bonds. This can be directly associated with diffusion of boron.

The photomicrographs indicate clearly that a layer of nickel solution grows from Inconel 718 into the molten liquid, and precipitates form at the side of Inconel 718 base metal. The precipitates appear to have segregated both along grain boundaries and inside grain



(a) (b)

(c) (d)

Figure 1 Typical microstructures of brazement made with nickel-based brazing foil at 1423 K for various times: (a) 10 min, (b) 30 min, (c) 60 min, and (d) 120 min.

immediately adjacent to the liquid/Inconel 718 interface. However, at the distance of 200 μm away from interface precipitates only occur along the grain boundaries. This phenomenon will be discussed in detail later. Examination of the typical morphology of brazed joint during solidification at 1423 K shown in Fig. 1a and b, reveals the following: the layer adjacent to the side of Inconel 718 contains grains separated by deep grain grooves. Between two grains of lateral similar size the grooves are oriented perpendicular to the layer (parallel to the direction of the layer growth), but if a large grain neighbors a small one, the grain boundary grooves between them is oriented toward the smaller grain. The interface between grains and the liquid is smoothly curved (not faceted), and a negative curvature of the interface is observed.

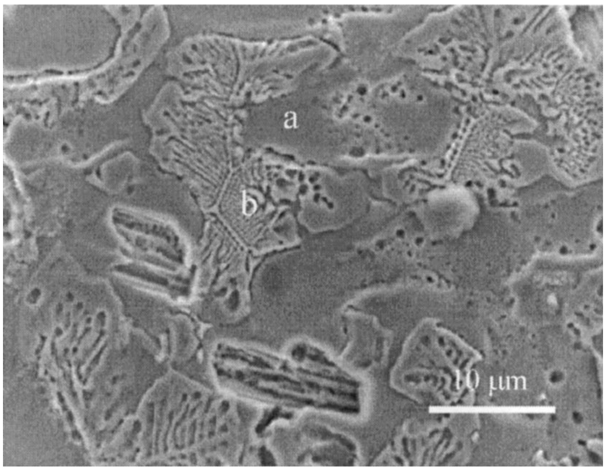


Figure 2 Intermetallic phases remain in the braze center: (a) is *g* nickel and (b) intermetallic.

However, as a result of the very large negative enthalpy of mixing (ΔH_m) between boron and silicon, and nickel and chromium, intermetallic compounds are often observed at brazed joints. The presence of these intermetallic compounds greatly deteriorates the joint properties and limits the success of direct joining of these two materials [13]. Intermetallic compounds mainly formed at the braze center and were seen more clearly using SEM as shown in Fig. 2. The formation of intermetallics based on silicon and boron would be expected at the center of the braze on cooling after a short hold time at the brazing temperature. EDS assisted indicated that these intermetallics were silicides

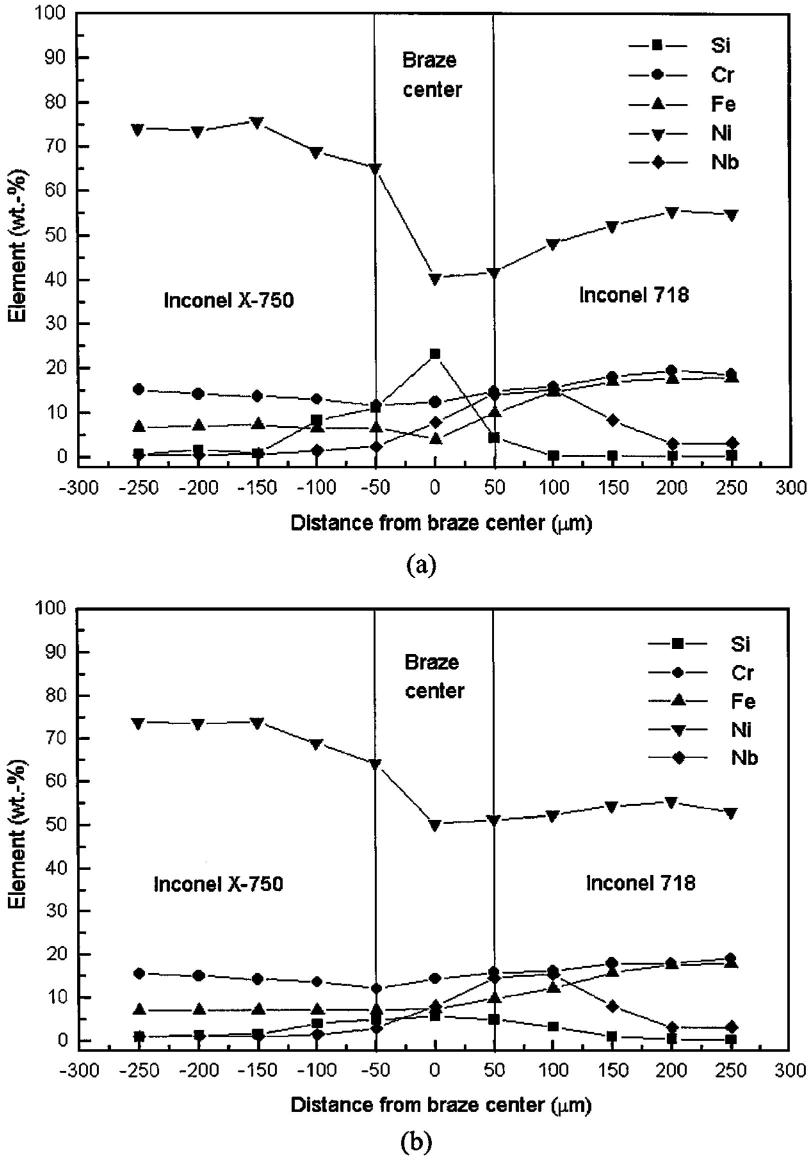


Figure 3 EDS composition profiles taken across bonds made using AMS 4777: (a) holding at 1423 K for 10 min, and (b) holding at 1423 K for 120 min.

and borides based on nickel, iron and chromium. According to the Ni-B-Si phase diagram [14], the intermetallic compounds were mixed with Ni_3B, Ni_3Si and Ni_5Si_2 at a temperature of 1423 K.

3.2. EDS profile across the bonds

EDS composition profiles as shown in Fig. 3a taken across the joints, indicated that after a short hold of 10 min, the braze center is rich in silicon and niobium, and only at a distance of about 150 μm away from the interface (within Inconel X-750 parent metal) did the concentration of silicon begin to decrease. Nickel is deplete in the braze center and is much lower than the concentration of filler metal (80 wt.-% Ni), because the base metal dissolved in the molten filler metal. The EDS analysis indicated 3.99 wt.-% iron, 2.90 wt.-% niobium, and 12.37 wt.-% chromium in the bond. It means that the dissolution of both Inconel 718 and Inconel X-750 does occur during TLP brazing operation. The concentration of iron is slightly higher than that in filler metal, while the concentration of chromium is much higher than that in the filler metal. It can be explained by that chromium has similar characteristics of nickel and its diffusivity is higher than that of iron. Niobium appeared in the brazed center due to the dissolution of Inconel 718. It is also observed that the degree of niobium at immediately adjacent to bond is relatively higher than that in Inconel 718 base metal. This is because niobium carbides segregated in the vicinity of liquid/Inconel 718 interface due to boron diffusion. It was also observed that a concentration of boron in the eutectic structure. Therefore, it seems that this eutectic structure consists of boron-chromium and boron-nickel intermetallic compounds. It would also appear that the boron did not completely diffuse from the brazed joint to the base metal, at the brazing temperature of 1423 K for 10 minutes. Both titanium and molybdenum were detected in the braze as a result of a substantial base metal dissolution in the liquid filler metal and affinity of base metal molybdenum to the boron and silicon.

Maintaining the specimen at TLP brazing temperature, the composition of the filler metal changed with time, because the boron and silicon in the filler metal diffused away from the joint, and into nickel matrix and grain boundaries of the nickel-based superalloys. This causes the intermetallic phases that formed at the centerline of the specimen joints, to decrease and the volume of the nickel solid solution to increase. Increasing the hold time to 120 min further homogenized the composition of the joint, especially with respect to nickel, iron and silicon. The matching X-ray maps show a very uniform elemental distribution in the joint including silicon, iron, chromium and nickel as shown in Fig. 3b. The nickel profile spread out with increasing hold time and this corresponded to the increase in thickness of nickel solid solution. The level of nickel increased to 50.3 wt.-% corresponding to Inconel 718 base metal, and iron increased to 7.30 wt.-% conforming to Inconel X-750 base metal. It is clear that the bond region is more homogeneous and the levels of boron and silicon have been considerably reduced. The EDS analysis indicated 5.57 wt.-% silicon remained in the joint region. This can explain that there are no eutectic phases left in braze center because the limit solubility of silicon in nickel is approximately 6 wt.-% at TLP brazing temperature. The large amount of intermetallic compounds (probably silicides and borides) at the braze center seem to have gone into solution but smaller and more irregular precipitates (likely to be the stable niobium and chromium carbide) are still visible on the side of Inconel 718 base metal.

3.3. Boron segregation

The commonly used melting-point depressants boron, silicon and phosphorus are an essential part of TLP brazing, because they can diffuse rapidly out of the liquid filler metal during brazing process. The loss of melting point depressants from the filler metal raises the melting temperature of the subsequent joint to a level similar to that of the parent metal.

Boron in Inconel X-750 probably causes the liquation of a small amount of Inconel X-750 as shown in upper part of Fig. 1. On the other hand, when boron diffuse into Inconel 718 (lower part of Fig. 1), it induced the niobium and chromium carbide precipitates in the base metal as shown in Fig. 4. Boron is a major melting point depressant for nickel-based alloys and can diffuse readily in the solid state at elevated temperature due to it's small atomic size ($R_B = 0.0079$ nm). Since Inconel X-750 base metal which contains 70 wt.-% nickel, has relatively higher concentration of nickel than Inconel 718 (50 wt.-% nickel). When boron diffuse along grain boundaries, large amount of boron agglomerates at the grain boundaries. Continued boron diffusion into base metal would rapidly result in a substrate boron in excess of the boron solubility in nickel (0.3 at.-% at the Ni-B eutectic temperature of 1366–1368 K [15]. Therefore, the grain boundaries become binary systems. When bond is cooled down before molten filler metal completely solidified, Liquation penetration of Inconel X-750 is expected to be observed because TLP brazing temperature is higher than eutectic temperature.

In Inconel 718, the formation of liquid has been attributed to the constitutional liquation of NbC particle, Laves phases, and possibly minority phases such as borides and silicides [16]. Carbide precipitates appear in Inconel 718 in the vicinity of joint interface during TLP brazing as shown in Fig. 4. This is directly associated with boron segregation. Because carbides appear close to joint interface where there is boron segregation. However, at the distance of 300 μm away from joint interface, no carbide occurred in this region where boron was also not detected. The role of boron in a nickel-based superalloy is believed to involve its segregation grain boundaries. The segregation of boron is believed to have one or more of the following effects: (i) increase grain boundary cohesion, (ii) reductions in grain boundary surface energy, (iii) lower grain boundary diffusion rates, and (iv) changes in γ' and/or $M_{23}C_6$ morphologies. This boron-induced carbide precipitation provided the pinning effect, which retard the grain growth. This can interpret that grain size adjacent to joint interface is smaller than that of far away from joint interface.

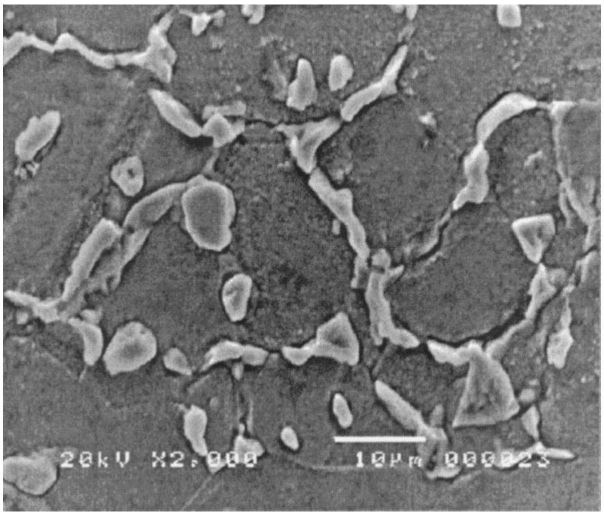


Figure 4 Carbide precipitate in Inconel 718 base metal.

It is important to point out that a moderate effect of nickel and chromium borides formation at grain boundaries of stainless steel on its mechanical properties is well known [17]. There is a special B-type 300-series of stainless steels for nuclear applications containing up 2.2 wt.-% boron. There are greater in boron content than in these brazements. In spite of such high boron concentration and extensive boride segregation, the yield and tensile strength of the B-type 300-series stainless steels are 205 and 515 MPa, correspondingly, i.e. only slightly lower than that of the conventional 300-series steel. Therefore, boron segregation does not seriously affect mechanical properties of TLP bonds.

3.4. Kinetics of TLP brazing

Because the actual brazing temperature was kept at 1423 K, liquid can be regard as a constitutional undercool even though there is no temperature gradient. The isothermal solidification process is controlled by the formation and growth of the nickel solid solution. In SEM and optical microscopy examinations and in XRD of the sample, nickel solid solution was the only phase detected during solidification. The growth kinetics of γ nickel was found to be time-dependent. At the beginning of the process, γ nickel grows parabolically. Because base metal is fine at the beginning of the process, large amount of grain boundaries provides diffusion path for boron and silicon.

In isothermal solidification of long duration a decrease in the growth kinetics of the layer is observed. There are reasons for these time-dependent kinetics. First, These superalloys possessed a very fine equiaxed grain size of about 10 μm. There are many grain boundary diffusion paths that promote diffusion of depressant elements from the interlayer into the base metal. This might be greatly beneficial for TLP brazing process. The atomic structure of the amorphous interlayer is a glassy state. At the TLP brazing temperature, the interlayer melted and the liquid zone was further widen by dissolution adjacent to the base metal. However, some grain boundaries disappeared because of grain growth in base metal as brazing time went on. This considerably reduces the contact area between liquid filer metal and base metals. Hence, kinetics of isothermal solidification decreases significantly. This is consistent with Kokawa's [18] results that liquid penetration at grain boundary region and the deviation from a planar liquid-solid interface profile accelerate the isothermal solidification process by increasing the rate of depressants diffusion into the base metal. This result is also supported by that the apparent mean diffusion rate of depressant in solid nickel decreased when the grain size of the nickel increased [19]. Secondly, lateral grain growth of the solution layer occurs simultaneously with the phase growth. As time elapses, the number of grooves per unit area of the solid/liquid interface decreases. Since the grooves are the source of Ni atom for the fast phase growth at the solid/liquid interface, the consequent decrease in their density results in a gradual decrease in the growth kinetics. The other reason is the decrease in the constitutional undercool with further increase in brazing time, since the concentrations of depressants continue to decrease due to diffusion into base metal.

3.5. Evaluation of strength

The joint strength with joining temperature and time was evaluated by the shear strength test via an Instron-type tensile test machine using a special jig as shown in Fig. 5. Fig. 6 displays shear strength of Inconel 718/Inconel X-750 TLP bonds produced with AMS 4777 interlayer at 1423 K for various length of time. It can be seen that the shear strength of the TLP bonds increased with the TLP brazing time. Room-temperature testing bonds produced after 10 min with nickel-based foil gave a shear strength value of 329 MPa. As indicated in Fig. 1a, a large amount of brittle intermetallic phases formed at the centerline of the specimen joint at 1423 K for 10 min. The bond failed at the braze/parent metal interface. The fractograph in Fig. 7a shows the failure mode was by cleavage resulting in flat fracture surfaces. The location of brittle failure at the bond interface appeared to the location of silicide-boride precipitates that were seen to form in this region during brazing operation. These intermetallic compounds decreased the strength of a bonded joint and caused brittle fracture. The intermetallic phases continue to decrease when brazing time was extended to 30 and 60 min respectively. This is confirmed by the increase in shear strength with increasing brazing time at brazing temperature. The value of shear strength can achieve 350 and 385 MPa as shown in Fig. 6. Fractures occurred in the brazed zone in further tests with this brazing condition. However, TLP bond made at 1423 K for 120 min can obtain 550 MPa shear strength that is 60% of base metal strength. This is not surprising because a longer hold at brazing temperature results in a joint visibly free from eutectic phase. The intermetallics fully dissolved into solid solution at such brazing cycle. The fracture surface that indicates high plastic deformation in advance of fracture as shown in Fig. 7d. In this case, the bond strength of a joint with intermetallic-free phases was higher than that of a joint with brittle phases formed at the centerline of the specimen joints at 1423 K for 10, 30 and 60 min. Tear dimple fractographic features were observed.

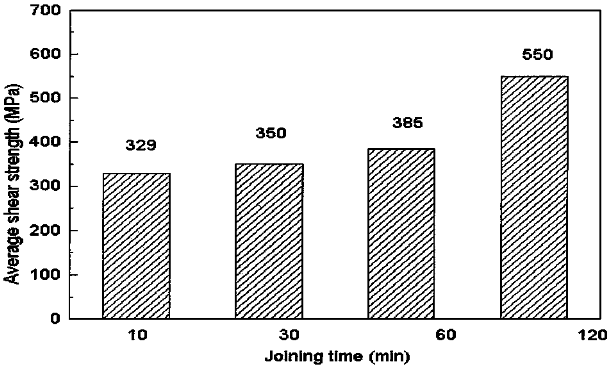


Figure 6 Average shear strength of Inconel 718/Inconel X-750 TLP bonds produced with AMS 4777 interlayer at 1423 K for various length of time.

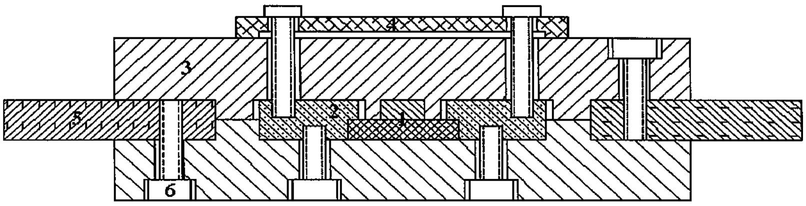
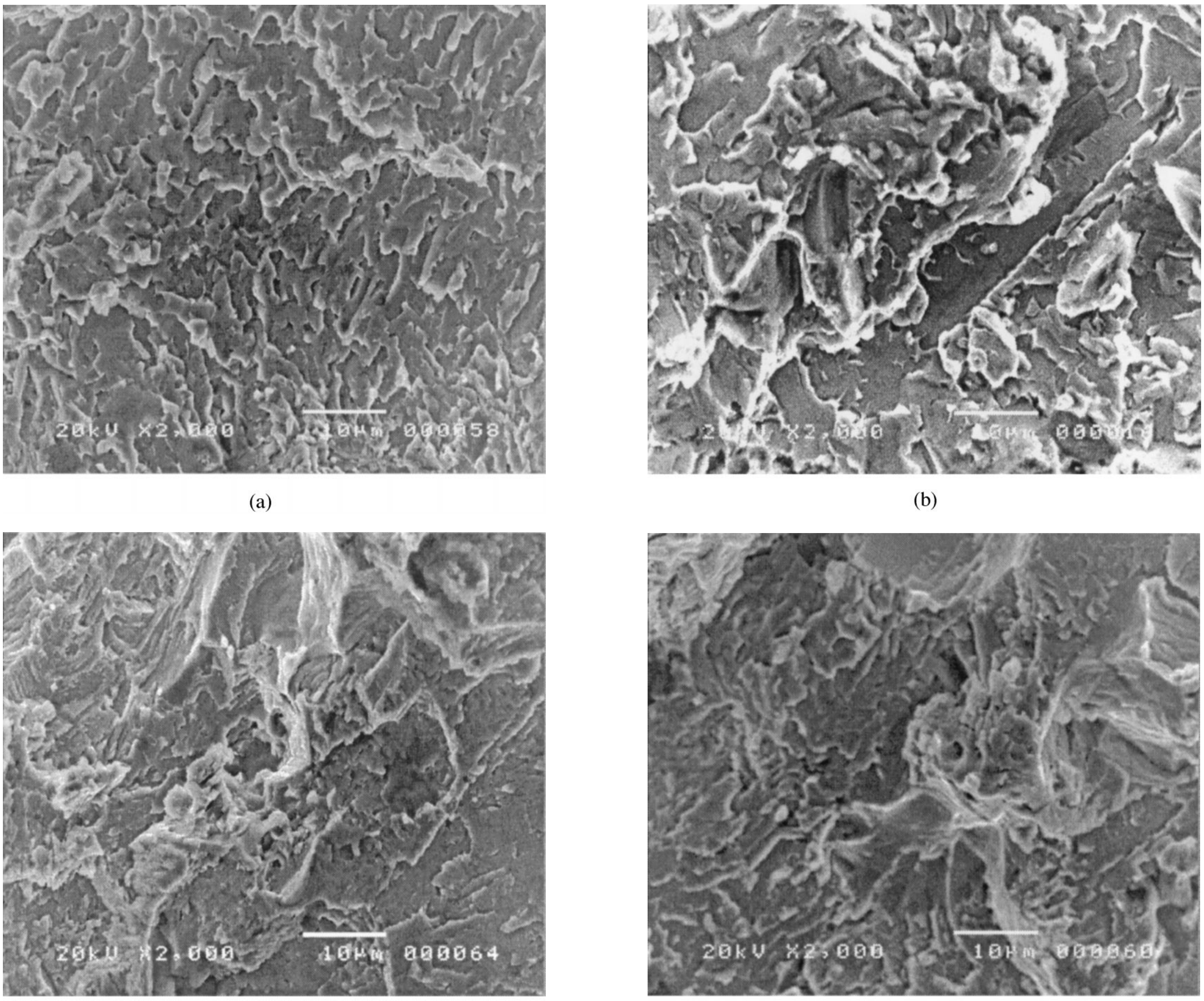


Figure 5 Shear-in-compression fixture developed for shear strength testing: (1) specimen, (2) internal clamp, (3) holder, (4) external clamp, (5) grip, and (6) screw.



(a) (b) (c) (d)

Figure 7 Fracture surface morphology of shear test specimens: (a) 10 min, (b) 30 min, (c) 60 min, and (d) 120 min.

4. Conclusion

A nickel-based foil containing the melting point depressant boron and silicon has been employed successfully as a TLP brazing interlayer for nickel-based superalloys Inconel 718 and Inconel X-750. The following conclusions were drawn from the present study:

Transient liquid phase (TLP) brazing of Inconel 718 to Inconel X-750 with AMS 4777 interlayer at 1423 K for 10, 30 and 60 min showed that Intermetallic phases formed in the centerline of braze seam. The thickness of brittle phases decreased with increasing brazing time. This results in the increase in shear strength of TLP bonds. Furthermore, it is also observed that niobium, titanium and molybdenum appear in the braze center due to the dissolution of base metals during TLP brazing second stage.

When TLP brazing was performed for 120 min, metallographic examination illustrated uniform chemical composition in the bond region and in the base metals. The shear strength can achieve 550 MPa at this brazing condition. The shear strength of the TLP bonds free from intermetallic phases indicated higher bond strength than that with brittle intermetallic phases. Fractograph displays ductile dimple rather than brittle fracture.

Boron has a strong tendency to segregate along grain boundaries. The segregation of boron is believed to increase grain boundary cohesion, reduce grain boundary surface energy, lower grain boundary diffusion rates, and change carbide morphologies. The diffusion of boron from the interlayer into the parent metal results in liquid penetration in Inconel X-750 and carbides precipitation in Inconel 718. However, boron segregation does not seriously affect the mechanical properties of TLP bond in this brazing condition. Kinetics of TLP brazing is time-dependent because effective diffusion path decrease and grooves disappear as grain grows during TLP brazing.

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