The Effect of Base Metal Cr Content on the Transient Liquid Phase Bonding Process for Joining ODS Superalloys

R. K. Saha, Tahir I. Khan
Department of Mechanical and Manufacturing Engineering
University of Calgary, Calgary, Alberta, Canada
tkhan@ucalgary.ca

Abstract

The transient liquid phase (TLP) bonding of a nickel based oxide dispersion strengthened (ODS) alloy was performed using a Ni-P interlayer. The diffusion of Cr from the parent metal into the joint region was investigated and its affect on isothermal solidification of the joint was determined. Metallurgical observations suggested the isothermal solidification process is not only controlled by the loss of P from the liquid interlayer, but the diffusion of Cr into the liquid interlayer helps to accelerate the solidification process.

Keywords: Diffusion Bonding, Transient Liquid Phase, Isothermal Solidification, Microstructure, Alloying Element, Interlayer

Introduction

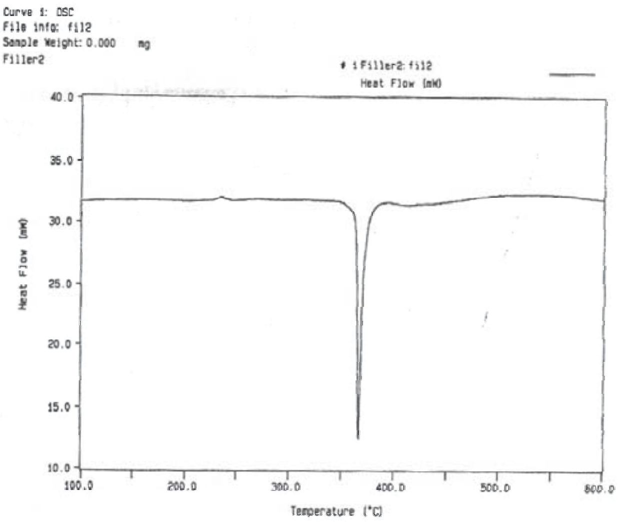
Transient liquid phase (TLP) bonding holds promise as a technique for joining superalloys and the process has been described in detail in earlier work [1]. In brief, this is a technique which uses an interlayer of specific composition containing melting point depressants (e.g. B, Si or P) between the two surfaces to be joined. At the bonding temperature, the interlayer melts filling the gap between the joining surfaces with a thin liquid layer. While the parts are held at the bonding temperature, diffusion of alloying elements occurs between the interlayer and base metal. As the melting point depressants diffuse from the interlayer and into the adjacent parent metal, the interfaces of the joint suffer some dissolution resulting in parent metal melt-back. As diffusion continues, a gradual change in the composition of the joint leads to isothermal solidification of the joint producing a metallurgical bond. By permitting the specimens to remain at the bonding temperature for longer times beyond that required for isothermal solidification to occur, the bond can be homogenized. After homogenization the joint microstructure generally resembles that of the base metal. A detailed description of the TLP bonding mechanism using the phase diagrams for binary systems has been given by Tuah-Poku *et al* [2].

An important parameter in the TLP bonding process is the composition of the interlayer which can determine the final chemical properties of the joint, but also the time required for isothermal solidification. Consequently, in previous research on TLP bonding processes, more attention was given to the outward diffusion of elements from the interlayer, especially the melting point depressants within the joint region [3]. However, it is known that some alloying element present in the base metal can also make a significant difference on the isothermal solidification process which in turn affects the final joint quality. Nakagawa et al. [4] reported that the chromium (Cr) content in the parent alloy or in the interlayer can result in a relatively longer time to complete the TLP bonding process. Although considerable research has been carried out on the effect of interlayer composition on TLP bonding behavior, to date little effort has been devoted to the study of the effect of base metal composition on isothermal solidification behavior during the joining process. Therefore, the intention of this study was to investigate the effect of base metal alloying elements, such as Cr on the isothermal solidification stage of TLP bonding a nickel based oxide dispersion strengthened alloy (MA 758).

Experimental procedure

In this study, an oxide dispersion strengthened nickel alloy, Inconel MA 758, was bonded using a Ni-P interlayer (91.72 wt% Ni and 8.28 wt% P). The samples used for bonding trials were cut from an extruded bar which was received in the recrystallized and elongated grained state with the composition of 30 wt% Cr, 0.05 wt% C, 0.3 wt% Al, 0.5 wt% Ti, 0.6 wt% Y_2O_3, 1.0 wt% Fe and 67.55 wt% Ni. The Ni-P interlayer was received as an amorphous 50 μm thick foil in which the P acts as the melting point depressant for this filler meal. The DSC and DTA traces for the Ni-P interlayer are shown in figure 1 and 2. Only one crystallized phase, Ni_3P, was detected, which has a melting temperature of 885 ºC. Accordingly, a bonding temperature of 1000ºC was chosen. The bonding conditions were based on earlier work [3], and TLP bonding was performed in a vacuum chamber using induction heating with an optimum bonding time of 30 minutes. A minimum bonding pressure was applied to provide intimate contact between the joining surfaces. For metallographic characterization, samples were sectioned transversely through the joint region, polished and etched using a solution of 1 part H_2O_2, 1 part concentrated HCl and 4 parts H_2O. The joint region was examined using light and scanning electron microscopes. Energy dispersive x-ray spectroscopy (EDX) was used to perform compositional analysis across the bonded interface.

Corresponding Author: Tahir I. Khan



TEMP 1: 40.0 c TIME1: 0.0 MIN RATE: 30.0 c/min
TEMP 2: 700.0 c

Fig. 1. Crystallization of the Ni-P interlayer (DSC trace)

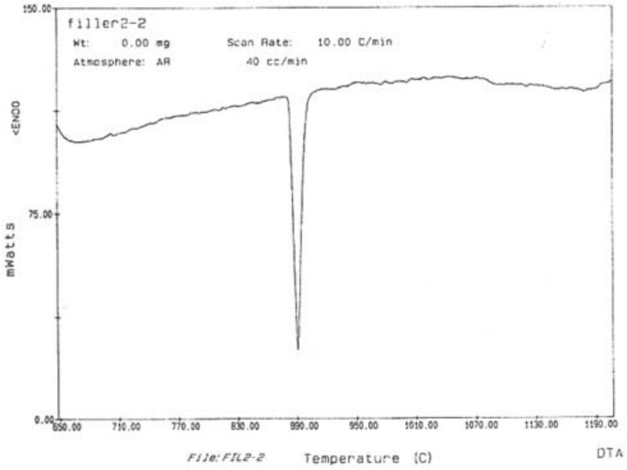


Fig. 2. DTA trace showing the melting point for the Ni-P interlayer

Results and discussion

The micrograph in figure 3 shows the microstructure of a TLP bond made using a Ni-P interlayer at 1000ºC with a hold time of 30 minutes. In agreement with earlier research [3, 4] no residual eutectic structure was found within the joint region and this suggested that isothermal solidification of the bond was completed. Some liquid penetration along grain boundaries in the parent alloy and adjacent to the joint was seen. This was attributed to the high rate of grain boundary diffusion compared to that of volume diffusion. Tuah-Poku *et al.* suggested that liquid penetration along grain boundary regions increased the effective solid/liquid interfacial area and the amount of phosphorus diffusion into the parent alloy [2]. As a result, this penetration accelerated the isothermal solidification of the liquid interlayer.

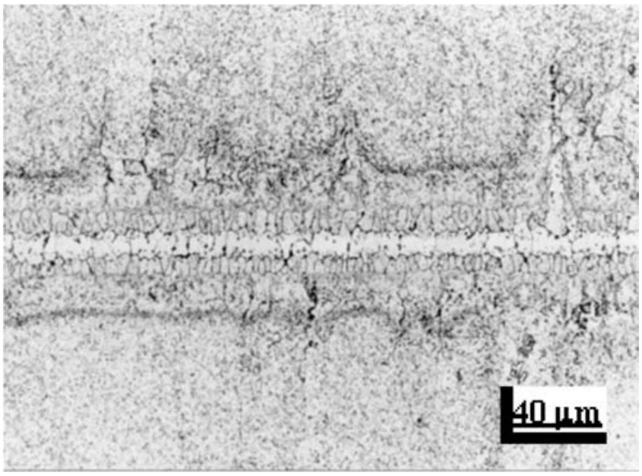


Fig. 3. Microstructure of bonds made using bonds made using a hold time of 30 min

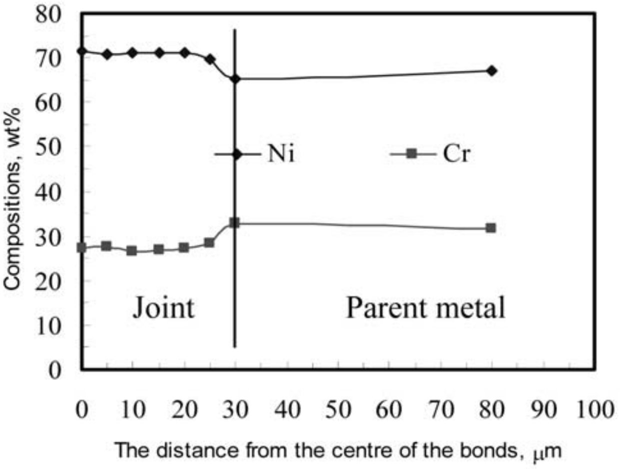


Fig. 4. Ni-Cr composition profiles of a hold time of 30 min

However, the results from this study show that the presence of Cr within the parent alloy shortened the time to complete the bonding process rather than prolong it. This is not consistent with the findings of Nakagawa *et al.* [4]. They found that the presence of Cr in the interlayer or in the base metal would prolong the time required for complete dissolution by considering the diffusion of solute in the liquid layer. The time to complete the bonding process is in order of a few hours. Firstly, after dissolution was complete, the Cr content of the bond would increase due to the rapid diffusion of Cr from the parent alloy in to the liquid interlayer. The Cr content within the MA 758 parent alloy is 30 wt% and this concentration is high enough to raise the solidus temperature of the Ni-P liquid interlayer, which in turn accelerates the isothermal solidification process. Furthermore, the diffusion of phosphorous from the interlayer in to the parent alloy would lower the content of phosphorous within the interlayer. This in turn will also raise the solidus temperature of the liquid interlayer. Such a change would make the dissolution and solidification processes much faster than otherwise observed during the TLP bonding processes. Furthermore, when Nakagawa *et al.* [4]

predicted the prolonged time for complete dissolution during TLP process, grain boundary diffusion and penetration was not considered as a contributing factor in their work. However, grain boundary diffusion can accelerate the time for completion of dissolution and isothermal solidification. Furthermore, in the present work an induction heating was used, and the presence of a magnetic flux at the joint region can result in a stirring effect and hence, accelerate the homogenization process.

The Ni-Cr composition profiles for the TLP bond for a hold time of 30 minutes is shown in Fig. 4. A higher concentration of Ni (~71.2wt %) and a lower concentration of Cr (~27.2wt %) was detected within the bond region when compared to that within the parent alloy. The concentration for both elements was uniform across the joint region and a sudden change in the content of Ni and Cr at the joint interface was observed. The high concentration distribution of Cr across the joint region further supports the discussion given above.

Figure 5 shows a SEM micrograph of bond with a hold time of 30 minutes. In the joint region, there are two distinct regions: the joint-centre line and a cluster of small grains on either side of the bond interface. There are precipitates present along the grain boundaries. This was made clearer at higher magnifications, as shown in figure 6(a). EDX analysis shows that these precipitates were rich in phosphorous (P), see figure 6(b). It also shows that there was a higher Cr content and a lower Ni. The element mapping of figure 6(a), shown in figure 7, confirmed that the precipitates along the grain boundaries were Cr (Ni) phosphides. The formation of phosphides was found to occur during the dissolution and isothermal solidification stages because precipitates formed not only along the grain boundaries, but also within the parent alloy. Although the solubility of P in Cr is very limited (about 0 wt% at 1000ºC), the solubility of P in Ni-Cr is much higher (≥ 14 wt% at 1000ºC). Kokawa *et al.* [6] did not observe phosphide when studying the TLP bonding of Ni/Ni-4wt%Cr-10wt%P/Ni system. This suggested that a high Cr content (e.g. ~30 wt% as in this study) was necessary for the formation of phosphides. In order for such a high Cr content to build up within the liquid interlayer, a rapid diffusion of Cr from the parent alloy into the liquid interlayer is required. It is suggested that local equilibrium was not established across the solid/liquid interface, and Cr was free to diffuse from the parent alloy into the liquid interlayer immediately during melting of the parent alloy. The continuous diffusion of Cr results in a phosphorous concentration in excess of the solubility of phosphorous in Ni-Cr, which would than result in the formation of phosphides. The formation of phosphides reduces the concentration of phosphorous in the liquid interlayer at the joint and as a result can accelerate isothermal solidification of the joint. A similar observation was made by Blanc and Mevrel [7], who found that boron consumption as a result of boride formation accelerated the isothermal solidification process in TLP bonding of the Ni/Ni-Si-B/Ni system.

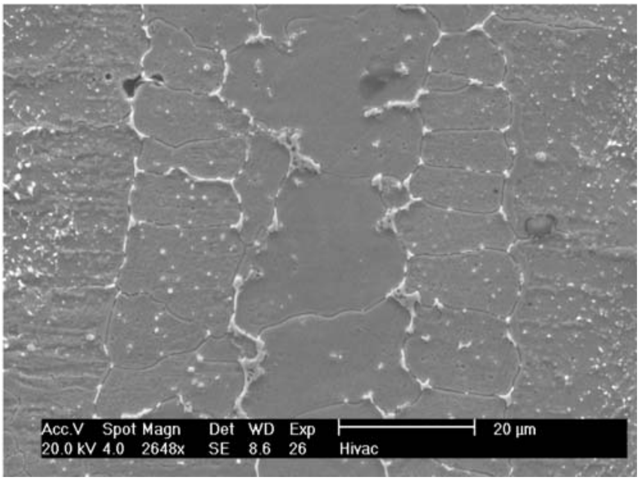


Fig. 5. SEM micrograph of bond with a hold time of 30 minutes.

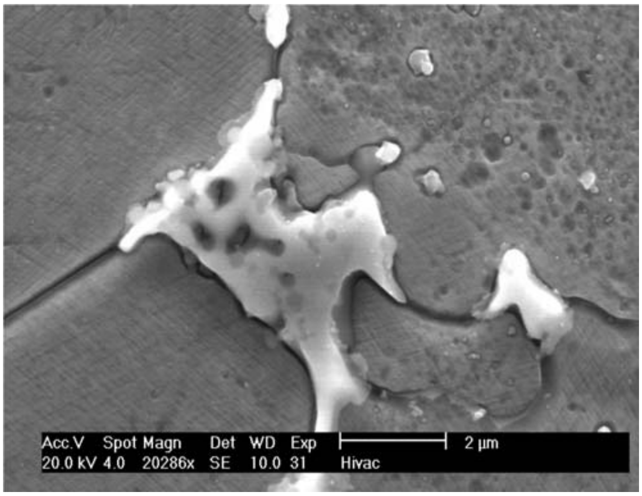


Fig. 6 (a). Precipitates along the grain boundary.

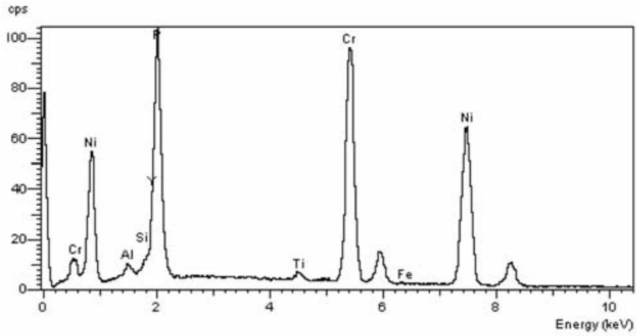


Fig. 6 (b). EDX spectrum taken from precipitates along the grain boundary.

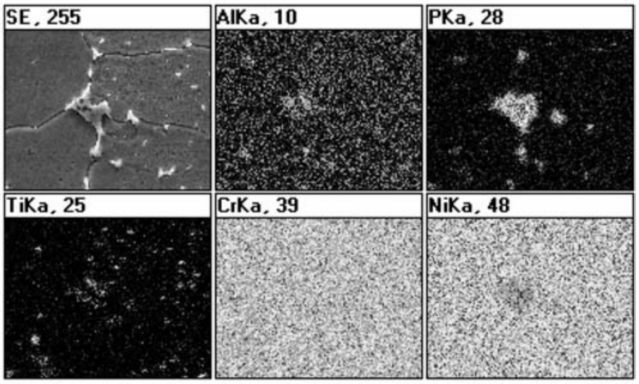


Fig.7. Elemental mapping of Al, P, Ti, Cr and Ni in figure 6(a)

Conclusion

From the result of this study it can be concluded that the composition of the parent alloy plays an important role in the solidification process during the TLP bonding of alloys. The MA 758 ODS alloy has a high Cr content and Ni-P could be a good interlayer to choose not only because it has a low bonding temperature, but the diffusion of Cr into the joint region can form a Ni-Cr-P liquid interlayer. It is thought that the diffusion of Cr into the joint region and the outward diffusion of phosphorous both contribute to the isothermal solidification process. The acceleration in the solidification process will also reduce the extent of dissolution of the parent alloy resulting in less disruption of the joint microstructure.

Acknowledgement

The authors wish to thank the Natural Sciences and Engineering Research Council of Canada for financial support for this research.

REFERENCES

1. T. I. Khan and S. Wei, "Application of diffusion bonding technology to join a Ni-based oxide dispersion strengthened (ODS) alloy", *Conf. Proc. 3rd International Conference on Advanced Manufacturing Technology*, 11-13 May 2004, Kuala Lumpur, pp. 991-996.
2. I. Tuah-Poku, M. Dollar and T.B. Massalski, "A study of the transient liquid phase diffusion bonding process applied to Ag/Cu/Ag sand-witch joint", *Metallurgical Transactions A*, Vol. 19A, 1988, pp. 675-686.
3. R. K. Saha, S. Wei and T. I. Khan, "A comparison of microstructural developments in TLP bonds made using ODS Ni-alloy", *Materials Science & Engineering A*, Vol. 406, 2005, pp. 319-327.
4. H. Nakagawa, C. H. Lee and T. H. North, "Modelling of base metal dissolution behavior during transient liquid phase brazing", *Metallurgical Transactions A*, Vol. 22A, 1991, pp. 543-555.
5. R. K. Saha and T. I. Khan, "Effect of bonding variables on TLP bonding of an ODS superalloy", *Journal of Materials Science*, Vol. 42, 2007, pp. 9187-9193.
6. H. Kokawa, C. H. Lee and T. H. North, "Effect of grain boundaries on isothermal solidification during Transient Liquid Phase Brazing", *Metallurgical Transactions A*, Vol. 22A, 1991, pp. 1627-1631.
7. A. Le Blanc and P. Mevrel, "Diffusion brazing study of DS247/BNi-3/Astroloy", *Conf. Proc. High temperature materials for power engineering*, ed. E. Bachelot et al., 1990, Dordrecht, The Netherlands, pp. 1451-1460.