Transient Liquid-Phase Bonding Using Coated Metal Powders

Ni-20Cr and 304L stainless steel powders coated with a melting point depressant, Ni-10P, were used as the interlayers to produce large root opening 304 stainless steel joints

BY W. D. ZHUANG AND T. W. EAGAR

ABSTRACT. Powder particles coated with a small amount of melting point depressant (MPD) reveal different sintering behavior in comparison to an uncoated powder mixture of the same composition. Interlayers consisting of the coated powder particles were used in the transient liquid-phase (TLP) bonding process. The coating material and the thickness of the deposit are important parameters that influence shrinkage. The amount of MPD was controlled such that the volume fraction of the liquid was very small but existed at all contacts, thus improving densification of the interlayer. Ni-20Cr and 304L stainless steel powders coated with Ni-10P were applied to join 304 stainless steels. Fully dense joints with mechanical properties comparable to those of the base metals were obtained with Ni-20Cr powder interlayers, whereas joints with 304L stainless steel powder interlayers showed inferior mechanical properties due to residual porosity in the joints.

Introduction

The transient liquid-phase (TLP) bonding process offers a unique way to join materials to yield high strength and ductility (Refs. 1–3). In this process, a layer of melting point depressant (MPD) is placed between the faying surfaces. The MPD forms a liquid layer at the bonding temperature and eventually diffuses into the base material and causes the joint to solidify isothermally. The kinetics of the process is controlled by solid-state diffusion of the MPD into the base material to be joined. The TLP bonding process can be divided into four stages, namely, dissolution, liquid widening, isothermal solidification and homogenization, as described by Tuah-Poku, *et al.* (Ref. 4).

Probably the first report on the TLP bonding process was by Lynch, *et al.* (Ref. 5), who prepared interface-free titanium joints using a nickel-copper interlayer. A series of micrographs in their paper showed the progressive dissolution of the interlayer and the eventual formation of a joint that was "effectively just a grain boundary." Following this work, the TLP process has been applied successfully to a number of material systems. One such fruitful application is in the aerospace industry. Hoppin and Berry (Ref. 6), working at the Aircraft Engine Group at General Electric, developed activated diffusion bonding for joining superalloys such as René 80. The Nor-Ti-Bond process, described by Wu (Ref. 7), was developed at Northrop by Wells and Mikus (Refs. 8, 9) to join titanium structures. At Pratt and Whitney, Duvall, *et al.* (Ref. 10), joined superalloy Udimet 700 using a nickel-cobalt interlayer and a process patented by Owczarski, *et al.* (Ref. 11). Nickel-boron eutectic was also used as the MPD for joining a similar alloy (Ref. 12). Niemann and Garrett (Refs. 13, 14) of MacDonald Douglas developed eutectic bonding for joining boron-fiber-reinforced aluminum matrix composites with a copper interlayer. Titanium-aluminum joints were made in a similar way. Liquid interface diffusion (LID) bonding was developed at Rohr Industries to bond honeycomb sandwich structures using copper-nickel interlayers (Ref. 15).

KEY WORDS

Coated Powder Interlayer
Large Root Opening Joining
Melting Point Depressant
Ni-10P
Ni-20Cr
Transient Liquid Phase
304L Stainless Steel

Despite the success of the TLP process in producing quality joints for a variety of materials, its use is restricted to small clearance joints (within tens of micrometers) to avoid intermetallic compound formation and/or unacceptably long isothermal solidification times. The time for isothermal solidification of a TLP joint can be expressed as (Ref. 4)

$$t_{is} = \left(\frac{C_i}{C_L}\right)^2 \frac{W_0{}^2}{16\beta^2 D_S} \qquad (1)$$

where W_0 is the initial thickness of the filler metal, C_i is the initial MPD concentration in the filler metal and C_L is the saturation concentration of the MPD in the liquid. D_S is the diffusivity of the MPD in the base metal, and β is a dimensionless parameter that is determined by the solidus and liquidus compositions at the bonding temperature ($0 < \beta < 1$). It is apparent that thinner filler materials require much less time for solidification.

To apply the TLP bonding process to large root opening joints (100 µm and above), which is frequently required in manufacturing, several efforts have been made. Interlayers with mixtures of base powders and MPDs were studied by Nakao, *et al.* (Ref. 16), and MacDonald, *et al.* (Ref. 17). Infiltration of liquid MPDs into the base metal powder interlayers was also investigated previously by the present authors (Refs. 18, 19). Nakao used a 250-µm-thick IN-100 powder sheet and a 44-µm-thick filler MPD metal M8F-80 (Ni-15Cr-4B) to join superalloy MM007. It was found that the time for isothermal solidification can be reduced by more than 2 orders of magnitude as compared with joints without the powder sheet. MacDonald applied interlayers consisting of mixtures of titanium and Ti-15Cu-15Ni powders to join Ti-6Al-4V. To eliminate the residual pores in the interlayer, a relatively large amount of MPD (Ti-15Cu-15Ni) has to be used (over 30

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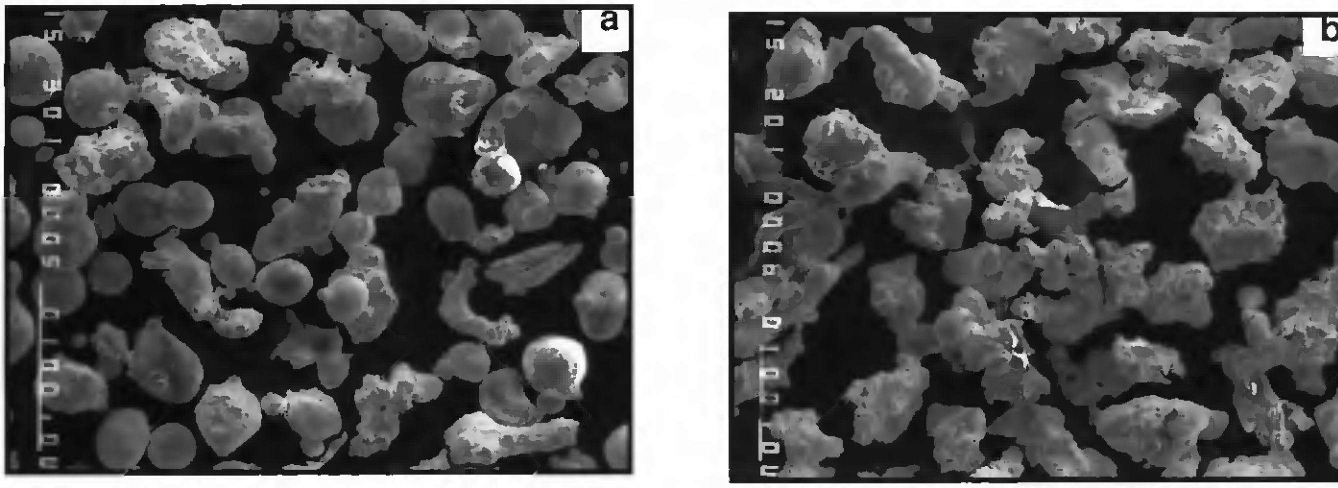

Fig. 1 — Features of the as-received metal powders. A — Ni-20Cr alloy powders; B — 304L stainless steel powders.

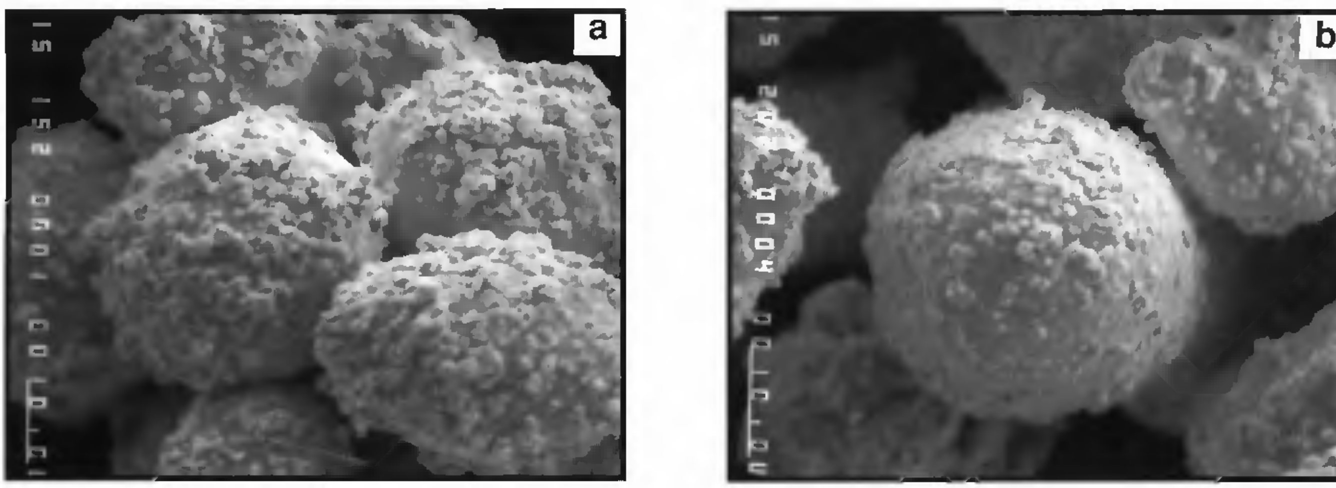

Fig. 2 — Ni-10 wt-% P electrolessly coated base metal powders. A — Ni-20Cr alloy powders with 16 wt-% coating; B — 304L stainless steel powders with 16.3% coating.

vol-%). Unfortunately, the resulting joints showed impaired mechanical properties due to excessive brittle intermetallic phases such as Ti_2Cu and TiNi. The present authors found that infiltrating the powder interlayer of the base metal with MPD was capable of producing fully dense joints while allowing much less of the MPD to be used. Large root opening copper joints were successfully made by infiltrating a copper powder interlayer with silver-copper eutectic alloy. However, the infiltration process is difficult to apply to material systems where there is a strong interaction between the powder particles and the liquid. The fast kinetics of chemical reaction can easily block the paths for infiltration as in the case of joining titanium alloys. Also, the process parameters have to be controlled carefully to produce mechanically sound joints.

In this paper, metal particles coated with MPDs are used as interlayers for large root opening TLP bonding. The amount of MPD is controlled by the coating thickness. In contrast to localized shrinkage induced by the liquid in the case of mixed powder interlayers, which usually needs more than 30 vol-% of liquid to fully densify the joint, full densification is obtainable with much less MPD for the coated powder interlayers, because the liquid is present at all powder-powder contact points, which leads to uniform shrinkage of the interlayer. Decreasing the amount of MPDs in the joint allows much shorter isothermal solidification time and makes it possible to produce strong and tough large root opening joints. Meanwhile, the coated powders can be used just like homogeneous particles, greatly simplifying the joining procedure. In this study, Ni-20Cr and 304L stainless steel powders with Ni-10 wt-% P coatings were used to join 304 stainless steels. Mechanical properties of the resulting joints were evaluated.

Experimental Procedures

Commercial Ni-20Cr (all compositions are in weight percent) and 304L stainless steel powders with particle sizes less than 44 µm (–325 mesh) were used. Figure 1 shows the general features of the powders. They were activated in a palladium-containing solution and electrolessly coated with Ni-10P in an aqueous solution. The phosphorus content was maintained by adjusting the pH value of the solution (Ref. 20). The coated powders were rinsed and dried. The amount of the deposit was determined by the weight gain of the powders.

Powder compacts of 10-mm diameter were made in a cylindrical die. The compression pressure was 350 MPa (51 ksi). The finished compacts were ready to be used as interlayers to join 304 stainless steels.

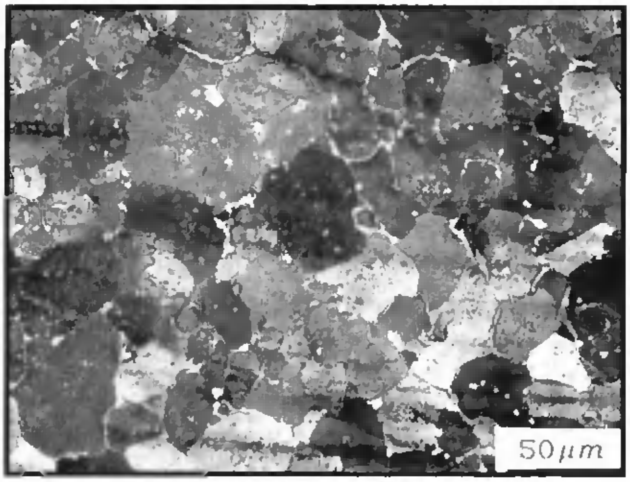

Fig. 3 — Microstructure of the Ni-20Cr powder interlayer after bonding at 1000°C for 1 h in vacuum. The overall phosphorus content in the joint is about 1.6 wt-%. Phosphides are uniformly distributed in the interlayer.

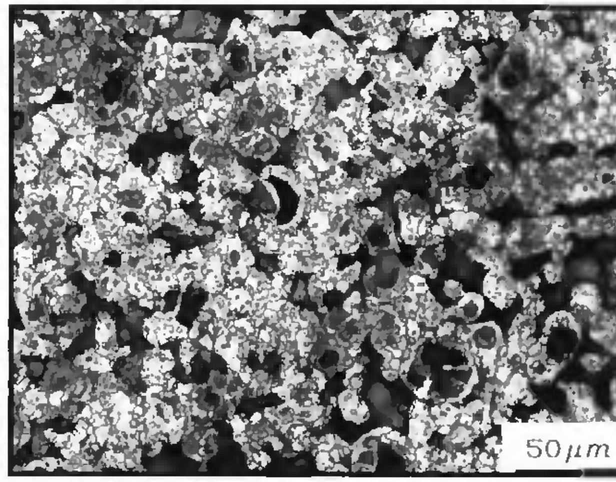

Fig. 4 — Microstructure of the 304L stainless steel powder interlayer after bonding at 1000°C for 1 h in vacuum. The overall phosphorus content in the joint is about 1.63 wt-%. Many residual pores indicate little shrinkage densification of the interlayer.

Joining was performed in a resistance-heated vacuum furnace with a vacuum level of better than 3 x 10^{-5} torr. Three joining temperatures 1000, 1055 and 1100°C (1832, 1931 and 2012°F) were chosen to make the joints. The average heating rate was determined to be about 20°C/min (68°F/min). At the bonding temperature, a compressive pressure of 0.29 MPa (42 psi) was applied to the joint. Butt joints for tensile tests were prepared in the same furnace. All the samples were air cooled to room temperature after joining with no further heat treatment.

After bonding, the joints were cut and polished for metallurgical examination in an optical microscope. Tensile tests of the joints were performed on an Instron test machine at room temperature, and the fracture surfaces were observed under a scanning electron microscope.

Results and Discussion

Microstructures

Figure 2 shows the features of coated Ni-20Cr and 304L stainless steel powders. In Fig. 1,16 and 16.3 wt-% Ni-10P was deposited for the powders illustrated, respectively. The corresponding overall phosphorus content in the interlayers made of these coated powders was about 1.6 wt-%. A phosphorus content from 0.7 to 2.3 wt-% was used to produce the joints. The microstructure of a joint with 3.8-mm-thick Ni-20Cr powder interlayer is shown in Fig. 3. The powder interlayer had fully densified after soaking at 1000°C for 1 h. Almost no remnants of transient liquid can be detected in the microstructure, indicating complete solidification of the joint. Some phosphides can be seen at the grain boundaries, as well as in the grains, due to the phosphorus in the joint. Heat treating to achieve diffusion of phosphorus into the base metal (SS304) is difficult because of the large joint clearance and, hence, the long diffusion distance.

A quite different microstructure was observed for the joints using 304L stainless steel powder interlayers, as illustrated in Fig. 4. After 1 h holding at 1000°C, the powder interlayer exhibits uniformly distributed fine phosphides. However, many residual pores can be found in the joint. The distribution of the pores in Fig. 4 suggests localized shrinkage of the particles in the interlayer. At first, it was suspected that poor wetting between the liquid and the base powder particles might be the cause, because surface oxides of the particles may retard wetting. To verify this, sessile drop tests were performed using compacted Ni-20Cr and 304L stainless steel powder substrates. Good wetting and flow of Ni-10 wt-% P on both substrates were observed in vacuum at 1000°C, therefore excluding the possible wetting problem.

The partial liquidus projection of the

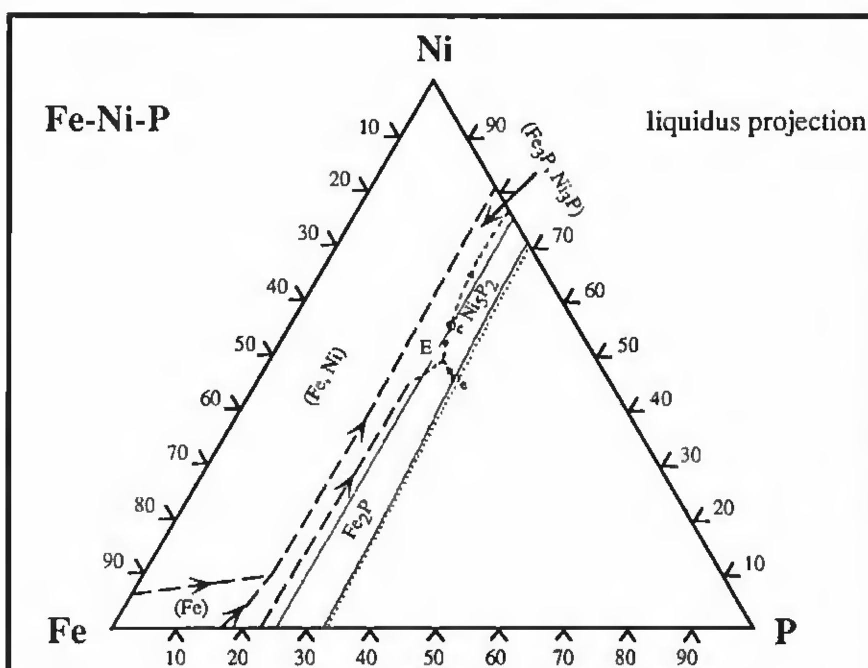

Fig. 5 — Liquidus projection of the iron-nickel-phosphorus ternary phase diagram (Ref. 21).

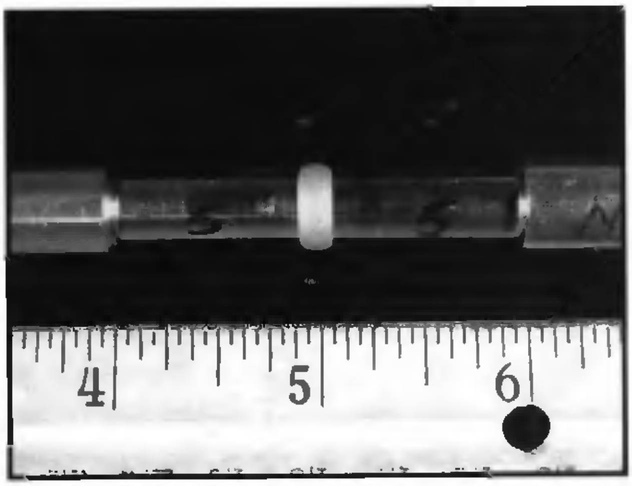

Fig. 6 — As-bonded tensile specimen using a Ni-20 wt-% Cr powder interlayer. The marker is in inches.

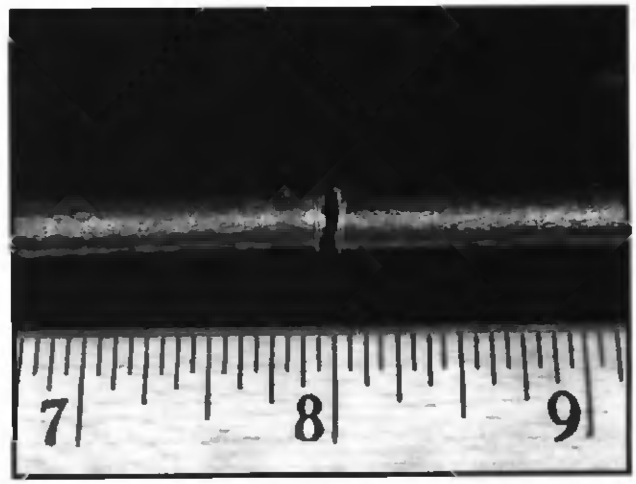

Fig. 7 — The specimen shown in Fig. 6 after machining and tensile testing. The marker is in inches.

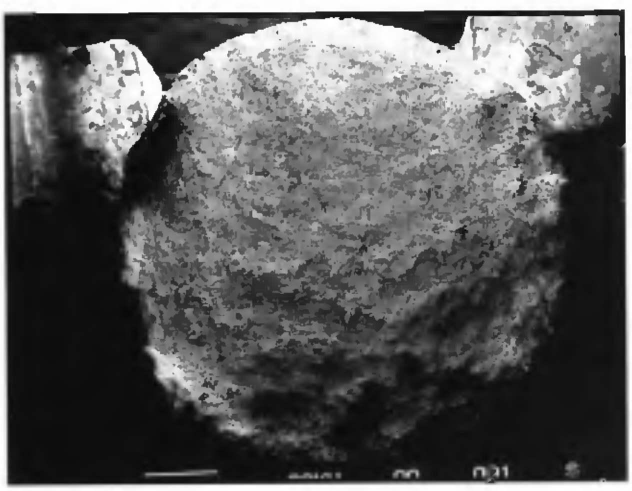

Fig. 8 — Fracture surface of the Ni-20Cr powder interlayer.

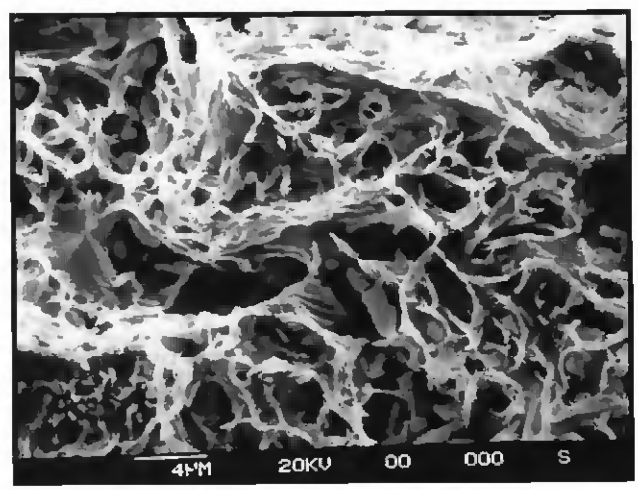

Fig. 9 — Fracture surface as shown in Fig. 8. Under higher magnification, considerable ductility of the joint is evident.

ternary nickel-iron-phosphorus system is given in Fig. 5 (Ref. 21), and it indicates an increase of the liquidus temperature as iron is introduced into the Ni-10P coating. The ternary eutectic temperature is about 950°C (1742°F), whereas the nickel-phosphorus binary eutectic temperature is only 870°C (1598°F). Interdiffusion between the Ni-10P coating and the 304L stainless steel powders during heating may increase the liquid incipient temperature. Thus, at the joining temperature, the liquid amount can be effectively reduced due to the diluted phosphorus concentration. Furthermore, because both nickel and phosphorus are soluble in the 304L stainless steel powders at the joining temperature, the liquid may disappear well before the powder particles can rearrange themselves and reach maximum shrinkage. As a result, a large number of residual pores remain in the interlayer. Elimination of such pores depends on further solid-state sintering, which is a slow process under the current bonding temperature. In contrast, because phosphorus is almost insoluble in Ni-20Cr powders, the liquid lasts much longer and fully dense joints can be produced. Increasing the joining temperature may help to increase the liquid amount and reduce pores in these stainless steel powder interlayers. However, such a beneficial effect may be offset by accelerated diffusion kinetics, which consumes the liquid more rapidly at the higher joining temperature. Indeed, this was observed in tensile testing where the tensile strength of the joints with 304L stainless steel powder interlayers showed no significant increase as the joining temperature was increased from 1000 to 1100°C.

Mechanical Properties

Figure 6 shows a typical as-bonded tensile test sample. The interlayer contained 1.6 wt-% P and was bonded at 1000°C for 1 h. Bulging of the powder interlayer can be seen in Fig. 6 because of the compressive load at the joining temperature. All the test samples were found to break in the interlayers when tensile tested. Considerable plastic deformation of the base metal is apparent during tensile testing when Ni-20Cr powder interlayers were used, as shown in Fig. 7. Figures 8 and 9 reveal the fracture surfaces of the sample shown in Fig. 7. The tensile strength as well as the strain was found to vary with the interlayer thickness, as illustrated in Fig. 10. When the interlayer thickness exceeds 1 mm, the tensile strength of the joint approaches that of the base metal. The strength of the joints with thinner interlayer thickness was not consistent in current experiments. One sample with 0.3-mm clearance was found to fail at the interlayer/base metal interface with about one-third of the area unbonded.

This could be caused by improper alignment of the sample during joining.

Interlayers of stainless steel powders exhibited lower strength, and the strength decreased with increasing interlayer thickness, as depicted in Fig. 11. Obviously, residual pores impair the strength of these joints. More Ni-10P deposits on the 304L stainless steel powders will produce more liquid at the bonding temperature. Nonetheless, this showed no improvement in the tensile strength, as can be seen from Fig. 12. This may be caused by the trade-off between the amount of liquid and the amount of intermetallic compounds. Increasing the joining temperature improved tensile strength and strain, as shown in Fig. 13. However, the effect is more likely due to solid-state sintering of the interlayers rather than the increased amount of liquid, because the tensile strain showed better improvement than the tensile strength as the bonding temperature was raised.

Conclusions

Transient liquid-phase (TLP) bonding using coated metal powder interlayers was investigated. Ni-20Cr and 304L stainless steel powders coated with Ni-10P melting point depressants (MPDs) were used as the interlayers to produce large root opening joints. On the basis of the microstructures and tensile properties of the joints made, the following results were obtained:

1) A thick TLP joint can be produced by applying a powder interlayer with its individual particles coated with a layer of MPD. Such a configuration provides a way of using less MPD for a thick TLP joint. Uniform shrinkage of the interlayer due to liquid existing at all powder-powder contacts at the bonding temperature can lead to a fully dense, mechanically sound joint.

2) Tensile strength close to the base metal (SS304) strength is obtained by using powder interlayers of Ni-20 wt-% Cr coated with 16 wt-% Ni-10P. Joints fail in the powder interlayers due to phosphide formation in the joints. A considerable amount of plastic deformation is observed before joint failure. Microstructural examination reveals the full density of the interlayer.

3) Residual pores exist in the 304L stainless steel powder interlayers after joining that degrade the mechanical properties of the joints. The tensile strength decreased with increasing joint clearance and phosphorus content. Increasing the joining temperature improved mechanical properties due to enhanced solid-state sintering.

4) Interlayers with MPD-coated powder particles can produce TLP joints of virtually any thickness. However, successful joints depend upon careful consideration of the combination of the MPD with the powder particles; factors include interdiffusion, reaction and solubility effects.

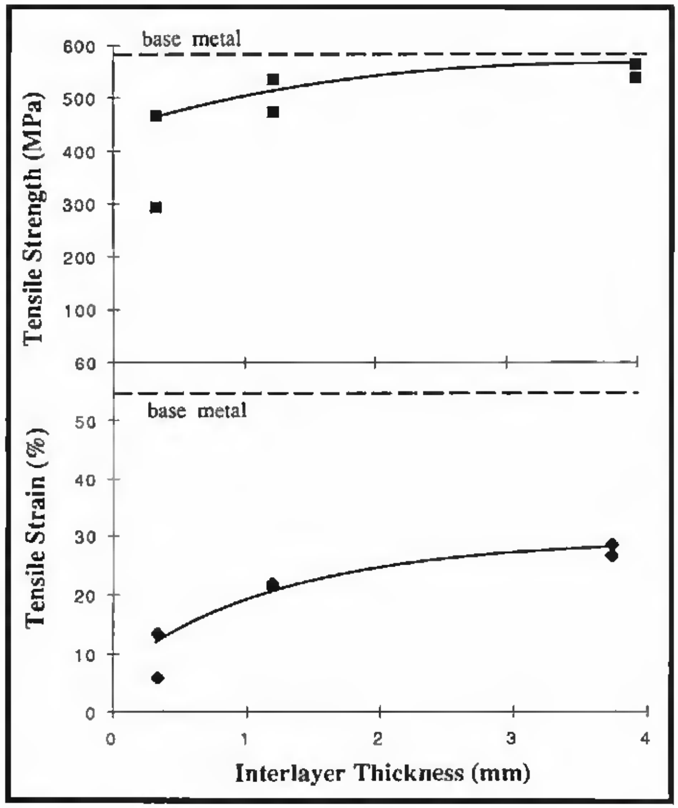

Fig. 10 — Mechanical properties of the joints with Ni-20Cr powder interlayers bonded at 1000°C for 1 h, showing tensile strength and strain as relating to interlayer thickness.

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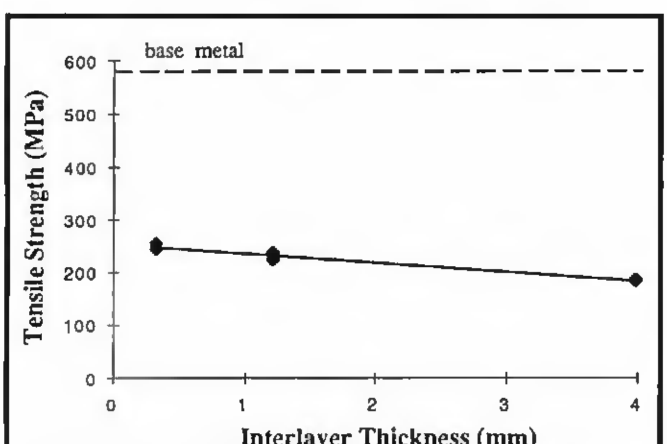

Fig. 11 — Tensile strength, as a function of interlayer thickness, of joints with 304L stainless steel powder interlayers bonded at 1000°C for 1 h.

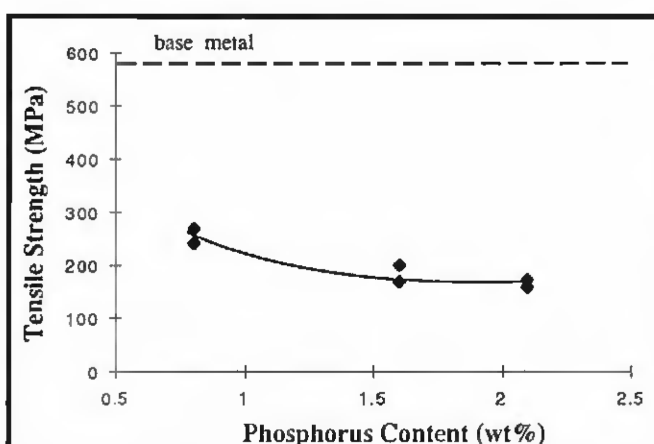

Fig. 12 — Tensile strength, as a function of phosphorus content, of joints with 304L stainless steel powder interlayers bonded at 1000°C for 1 h.

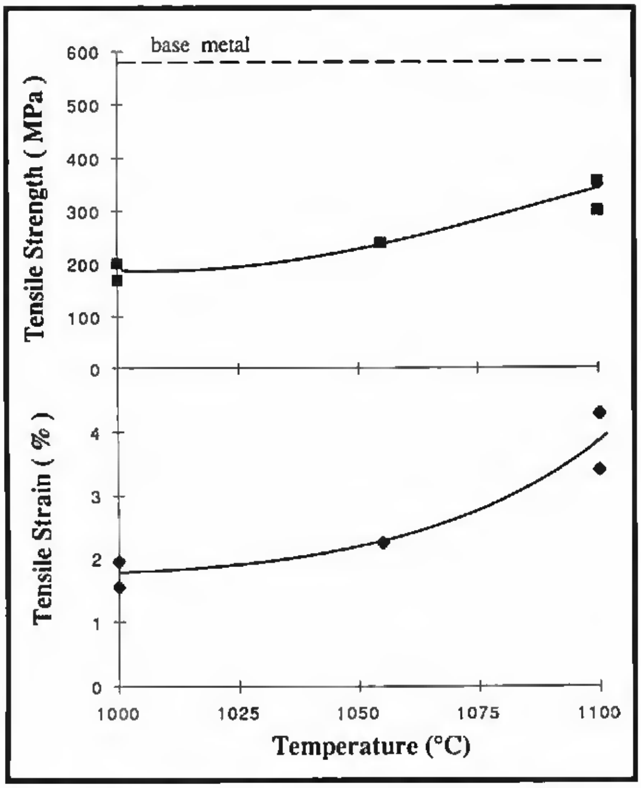

Fig. 13 — Mechanical properties of joints with 1.2-mm 304L stainless steel powder interlayers. Higher bonding temperature increases the joint strength and ductility.

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