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Transition liquid phase bonding of a Hastelloy X and the bond strength at 1173 K

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Transition liquid phase (TLP) bonding has been conducted on a Hastelloy X using amorphous foil filler metals. The inserted fillers were made by rapid solidification of a Ni–15% Cr alloy containing 3 to 5 mass% boron as a melting point depressant. The microstructural change with isothermal bonding time from 2.4 to 38.4 ks was examined in the bond regions by optical microscopy and electron probe microanalysis (EPMA). An assessment of the bond strength was made by stress rupture tests performed at 1173 K. It was found that rupture strain and rupture life increased with increasing bonding time. A bond efficiency greater than 80% was achieved when the bonding time was 38.4 ks.

1. Introduction

There is an increasing demand for high strength transition liquid phase (TLP) bonding of Ni base alloys. This is partly because of the greater demand for heat resistant alloys and their more diversified use and partly because it is generally difficult to apply a uniform bonding pressure to intricate-shaped thin parts. Since TLP bonding [1] or activated diffusion bonding [2] is a kind of brazing, bonding pressure is unnecessary in principle. This beneficial aspect of TLP bonding is especially favourable for bonding of complex-shaped thin parts where some deformation could occur by application of a bonding pressure.

In order to achieve high quality TLP bonding which is equivalent to solid diffusion bonding, a filler metal (interlayer) must melt at a temperature below the melting point or solid solution temperature of the base metals. A prerequisite for the filler metal is that the bond region must solidify at the bonding temperature and become chemically and microstructurally homogeneous with the base metal as is necessary for practical use.

To meet the above prerequisites, some amount of boron is usually added to the fillers to act as a melting point depressant. Another reason for boron being selected is its high diffusivity. This enables the bonds to be of an equivalent strength to the base metal during bonding time or post isothermal diffusion process. An addition of boron has these beneficial effects. However, too much boron in the fillers may produce unwanted stable phases in the bond region and excess boron content will make the bonding time longer.

The present paper reports the high temperature strength of a Hastelloy X bonded by the TLP bonding process. Amorphous fillers containing 3 to 5 mass% boron were produced by rapid solidification. The effects of the amount of boron and bonding time on the microstructure and bond strength were examined.

2. Experimental details

A Hastelloy X, bonded by the TLP process, is a solid solution strengthened Ni base superalloy. A solid solution treatment was carried out at 1463 K for 9 ks. The chemical composition of the base metal (Hastelloy X) is shown in Table I. The filler metals (B2, B3 and B5) were produced by rapid solidification, namely, molten metal in a high frequency induction furnace was sprayed through a quartz nozzle onto a cold copper roll rotating at high speed. The resultant film-like filler metals were about 20 μm in thickness and 10 mm in width. Inspection by X-ray back reflection, which was conducted on various parts of these filler metals, revealed diffuse ring patterns. This indicates that the films were nearly amorphous except for one portion in which a ring pattern showing the presence of minute crystals was observed. The chemical compositions of the filler metals are shown in Table II.

Taking into account the temperature for the solid solution treatment, TLP bonding was carried out at 1143 K in a vacuum brazing chamber at a pressure of 10^{-3} Pa. A time of 2.4, 4.8, 9.6, 19.2 or 38.4 ks was

TABLE I Chemical composition (mass %) and heat treatment of Hastelloy X

C	Si	Mn	Co	Cr	Mo	Fe	W	Ni
0.10	0.34	0.36	1.39	22.14	9.30	18.34	0.57	Bal.

Heat treatment: 1463 K for 9 ks followed by water quench.

TABLE II Chemical compositions of filler metals (mass %)

Alloy	Cr	B	Ni
B3	14.98	2.90	Bal.
B4	14.69	3.83	Bal.
B5	14.42	4.92	Bal.

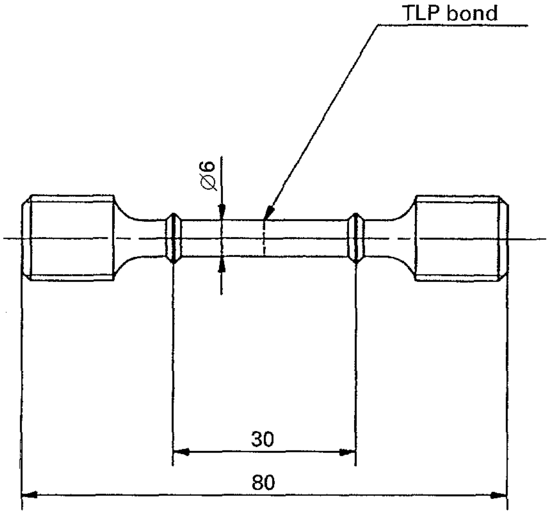

Figure 1 Microstructure of a brazed Hastelloy X.

allotted for the diffusion process. In principle, bonding pressure is unnecessary for TLP bonding. However, Nakahashi *et al.* [3] recently reported the remarkable effect of bonding pressure on high temperature bond strength. Therefore, in the present study a bonding pressure of 3.92 MPa was employed after referring to the preliminary study on the effect of bonding pressure.

Prior to machining of creep specimens, the top surfaces of a pair of bars were polished with 600# emery paper and degreased and then a butt joint was formed using various filler metals. In the following discussion, a creep specimen bonded with a filler metal of B3, B4 or B5 will be designated simply as a B3, B4 or B5 specimen. The dimension of a creep specimen is shown in Fig. 1. Creep tests were conducted at a temperature of 1173 K in a stress range of 29.4 to 53.9 MPa. Creep elongation was automatically recorded using a differential transformer with 0.01 mm precision. The melting point of B3 and B5 fillers is 1413 K and that of B4 is 1323 K.

3. Results and discussion

3.1. Microstructural change with bonding time

Fig. 2 shows the microstructural changes with bonding time for B3, B4 and B5 specimens. Close

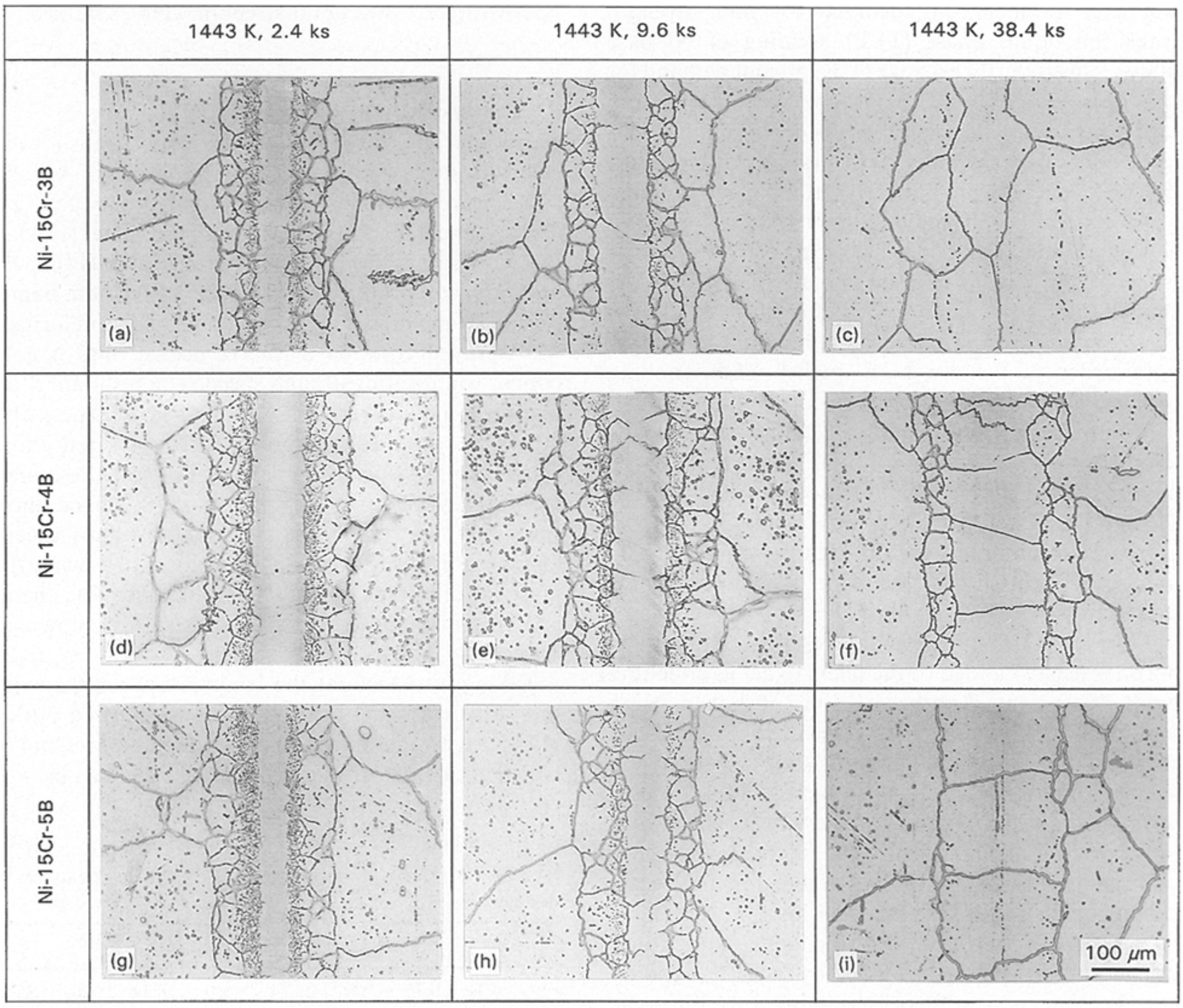

Figure 2 Shape and dimension of a creep rupture specimen.

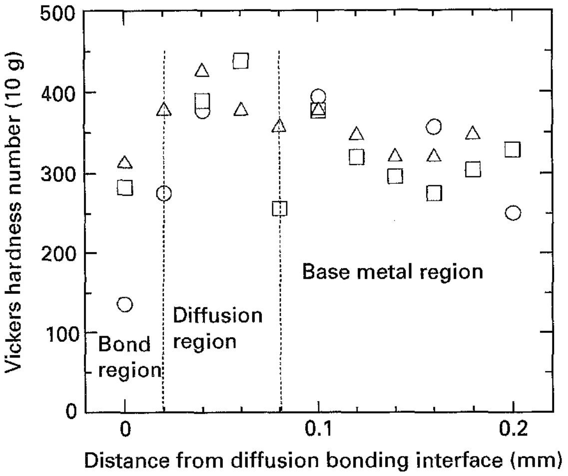

Figure 3 Vickers hardness distribution of the TLP bond (holding time: 9.6 ks). Key: ○ B3; □ B4; △ B5.

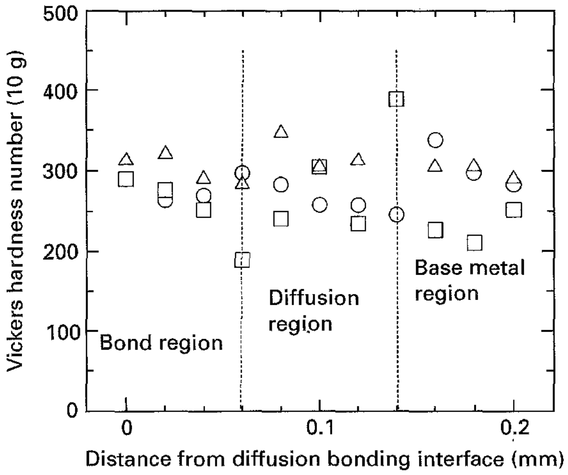

Figure 4 Vickers hardness distribution of the TLP bond (holding time: 38.4 ks). Key: ○ B3; □ B4; △ B5.

inspection shows that there are three different regions, namely (i) a bond region, (ii) a diffusion region and (iii) a base metal region for which there is no need for a comment. The bond region lies between two diffusion regions composed of relatively fine grains. The bond region is composed of a once-melted and re-solidified filler metal at least in the early stage of TLP bonding. This was ascertained by the fact that the presence of Fe was not detected in the bond region with short bonding times. The reason why the diffusion region is a fine-grained structure is not clear, however, it can be explained in the following way. Since the filler metal contains concentrated boron as a melting point depressant, it temporarily melts and resolidifies as the boron content decreases by diffusion into the base metal. Probably the fine-grained structure was produced by the action of stress occurring due to the isothermal phase transformation from liquid to solid. It is seen that the diffusion region becomes thinner and the corresponding bond region becomes thicker as the isothermal bonding time increases. However, the total width of the diffusion layers plus the bond region remains almost unchanged.

3.2. Hardness distribution in the bond region

Figs 3 and 4 show the hardness distribution in the bond regions for different bonding times. Comparison of these figures shows that the hardness is higher for a diffusion layer than that for base metal when bonding time is short. However, it decreases to the same level as the base metal values as the bonding time increases.

The increase of hardness in the diffusion layer is probably ascribable to the diffusion of Fe, Co, W, Mo and so on, from the base metal to the diffusion layer while the parent metal adjacent to the filler was melting temporarily as a result of the melting point depressant effect of boron. Among those elements which are not present in the fillers, Mo and W are most effective elements for solid solution strengthening of Ni [4–6]. In addition to this, there must be some contribution made by grain size refinement to the hardness increase.

3.3. Electron probe microanalysis of the bond region

The distribution of main elements composing the base metal and the fillers were investigated by electron probe X-ray microanalysis (EPMA). Figs 5, 6 and 7 are secondary electron images taken from the bond region of B3 specimens. Their bonding times are 2.4, 9.6 and 38.4 ks, respectively. It is seen that the longer the bonding time, the more the elements such as Ni, Fe and Cr diffuse into and are homogenized in the bond region.

Comparison of the area analysis shows that Mo and B are enriched along the grain boundaries when the bonding time is 9.6 ks (Fig. 6). However, it becomes indistinct when the bonding time is 38.4 ks. In practice, no segregation of boron is seen in Fig. 7.

3.4. Creep properties

Creep curves for B3 specimens are shown in Fig. 8 together with that for the parent metal. Testing was conducted at 1173 K and a stress of 49 MPa. The bonding time is shown in Fig. 8. Inspection of Fig. 8 shows that creep curves for bonded specimens approach that for the base metal as the bonding time increases. Figs 9 and 10 show the relation between bonding time and rupture time, and the relation between rupture strain and bonding time (logarithmic scale), respectively. It is seen that these relations are well expressed by a linear relation at least in the range investigated.

As shown in Fig. 8, the best condition for bonding was at 1143 K for 38.4 ks within the range of the present investigation. Based on this, the bonded specimens with different kinds of filler underwent creep at 1173 K at various stresses. The stress–rupture relations are shown in Fig. 11 together with the data for the parent metal. It is seen that each set of experimental data fails to fall on a well-behaved straight line. However, it can be safely said that a Hastelloy bonded

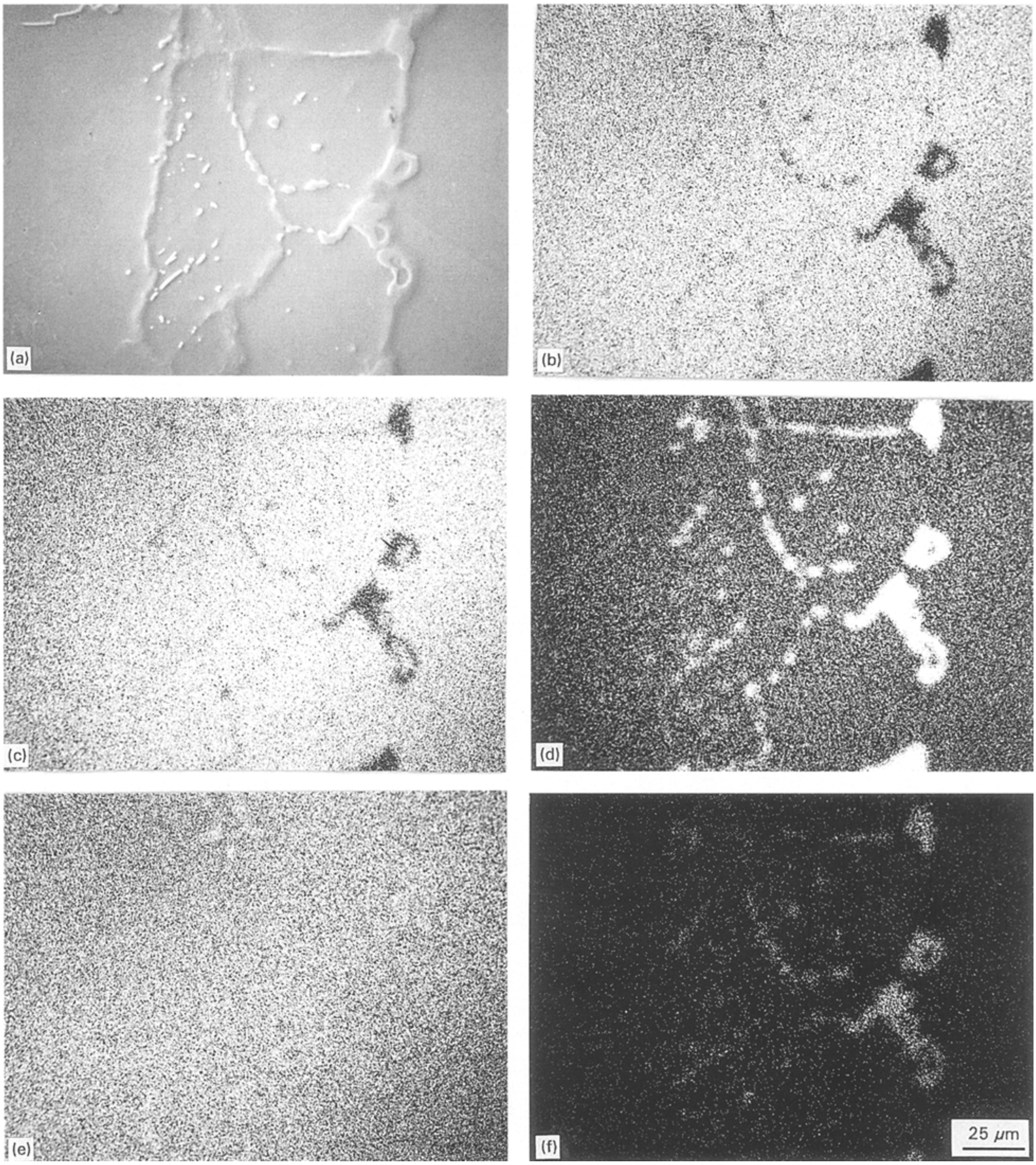

Figure 5 EPMA of the cross-sectional area for a brazed Hastelloy X, B3 (bonded at 1443 K for 2.4 ks). (a) SEM micrograph, (b) Ni, (c) Fe, (d) Mo, (e) Cr, (f) B.

by the TLP process can achieve a joint efficiency greater than 80% regardless of boron content. Close inspection of Fig. 11 shows that the filler of B3 is the most suitable for the present case. This tendency appears to become clearer as the applied stress decreases.

Fig. 11 shows the relation between rupture strain and rupture time for various bonded specimens. It is clear that rupture strain decreases with increasing boron in the filler metals. The fact that the bond efficiency decreases with boron content is probably due to the decrease in rupture strain with increasing boron [7]. In practice, the micrograph after rupture shows that creep rupture occurs near the bond region. Close inspection shows that there are some amounts of fine precipitates near the bond region although there were no precipitates before TLP bonding. Detail will be given elsewhere but these precipitates seem to deteriorate the creep elongation and eventually make the rupture life shorter.

4. Conclusions

TLP bonding was conducted on a Hastelloy X at 1443 K using amorphous fillers containing 3 to 5 mass% boron. The effects of boron content and bonding time on the microstructural changes and

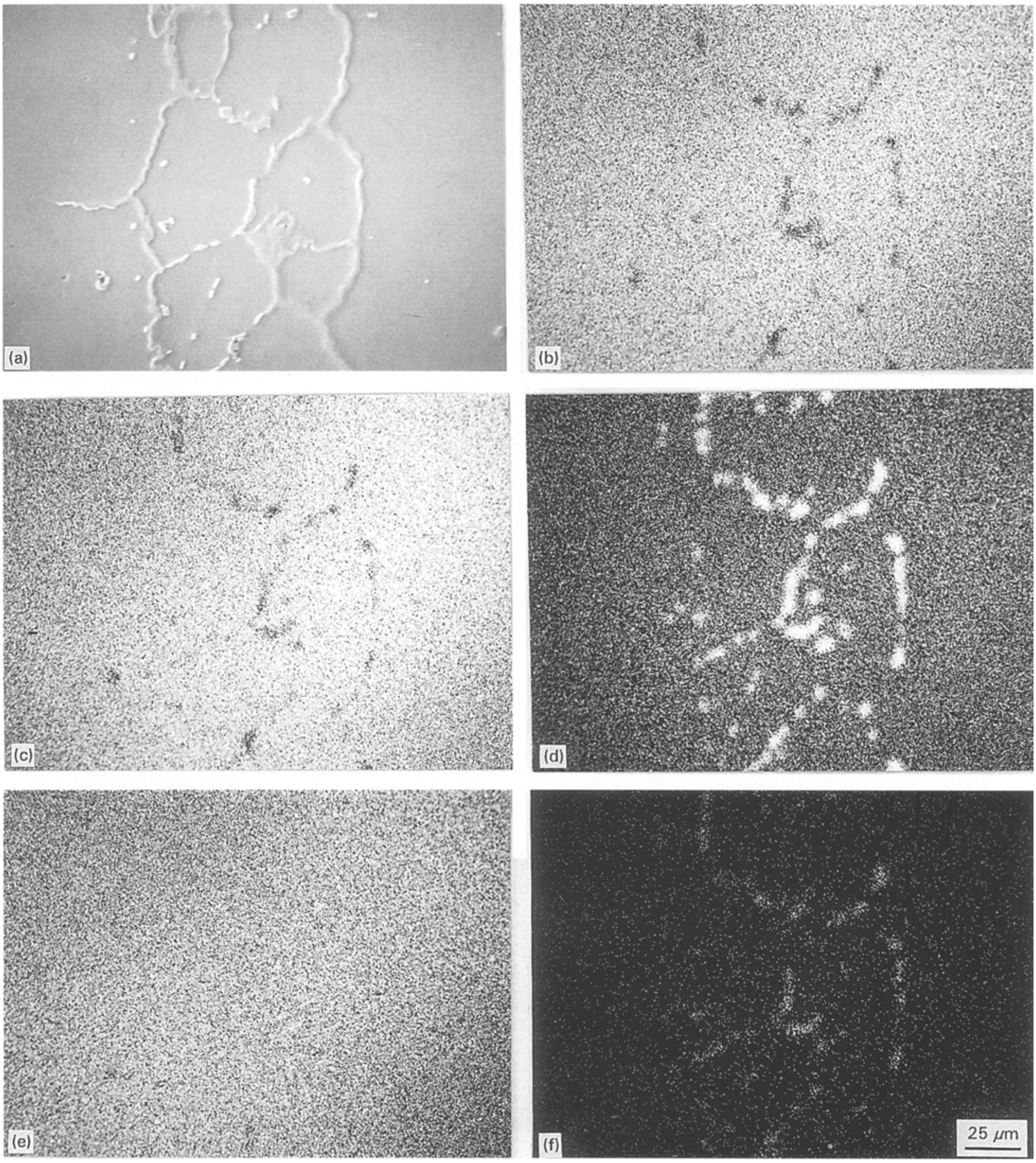

Figure 6 EPMA of the cross-sectional area for a brazed Hastelloy X, B3 (bonded at 1443 K for 9.6 ks) : (a) SEM micrograph, (b) Ni, (c) Fe, (d) Mo, (e) Cr, (f) B.

joint strength were studied. The creep rupture tests were carried out at 1173 K and a stress range from 29.4 to 53.9 MPa. The experimental results obtained are as follows:

1. Creep rupture strain of the base metal was about 50% at 1173 K and 49 MPa. On the other hand, the rupture strain of bonded specimens depends on the boron content and the bonding time.
2. When the bonding time is insufficient, three layers were observed near the bonded part. One is the bond region which lies in between two diffusion regions composed of relatively fine grains. Vickers hardness in the bond region is low during the early stages of bonding. The diffusion layers lie in between the bond region and the base metal region, and the Vickers hardness is higher than the other regions. However, interfaces between these regions become indistinct when the bonding time is sufficient.
3. Linear relations were obtained between bonding time and creep rupture time, and between rupture strain and the logarithm of bonding time.
4. Regardless of the selected filler metals, a bond efficiency greater than 80% was achieved when

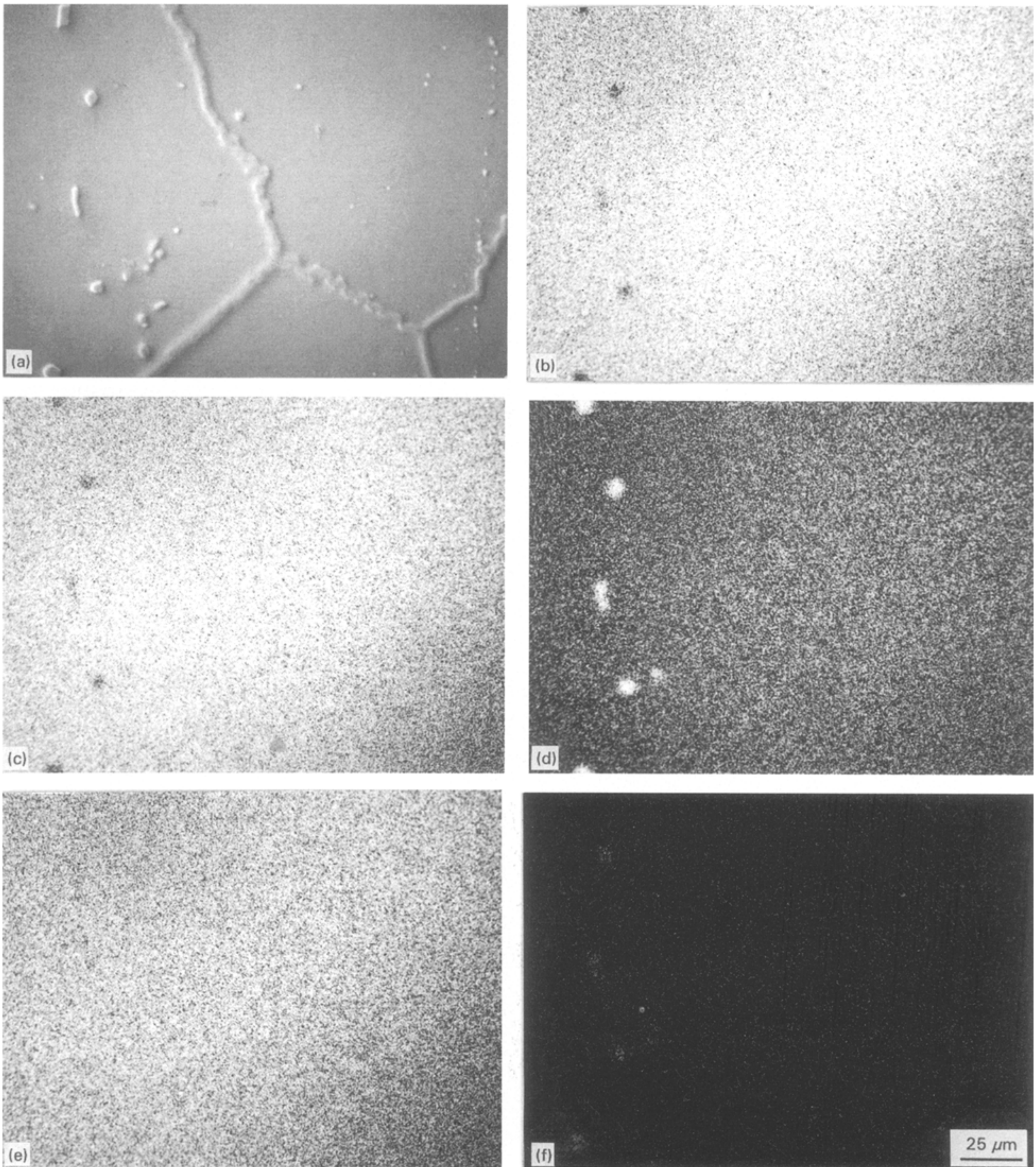

Figure 7 EPMA of the cross-sectional area for a brazed Hastelloy X, B3 (bonded at 1443 K for 38.4 ks) : (a) SEM micrograph, (b) Ni, (c) Fe, (d) Mo, (e) Cr, and (f) B.

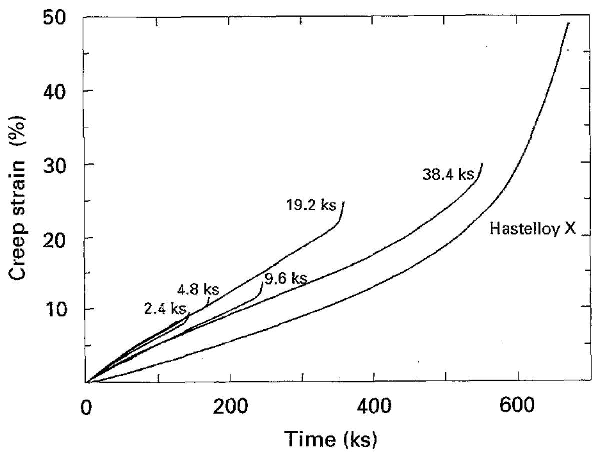

Figure 8 Creep curves of B3 specimens tested at 1173 K and 49 MPa.

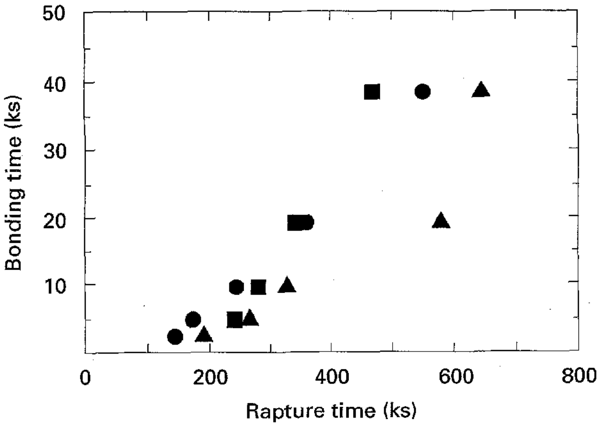

Figure 9 Bonding time versus time to rupture for B3, B4 and B5 specimens (tested at 1173 K and 49 MPa). Key: ■ B5; ▲ B4; ● B3.

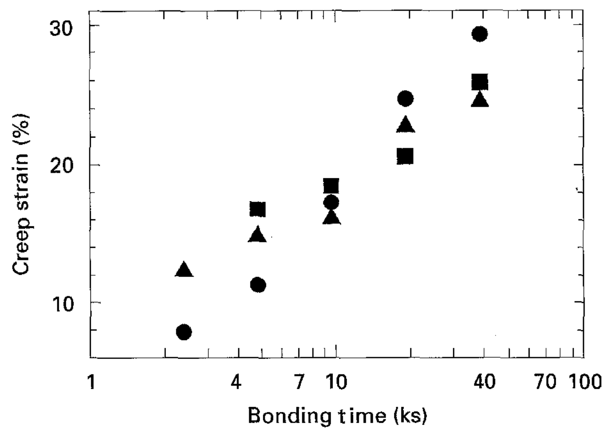

Figure 10 Change in creep rupture strain with bonding time for B3, B4 and B5 specimens (tested at 1173 K and 49 MPa). Key: ■ B5; ▲ B4; ● B3.

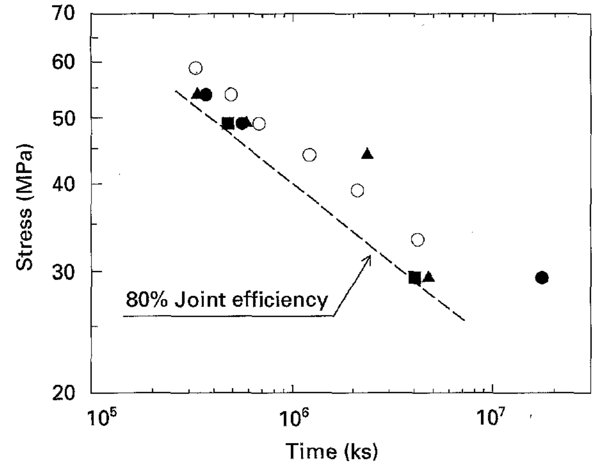

Figure 11 Stress versus time to rupture (bonded at 1443 K for 38.4 ks). Key: ■ B5; ▲ B4; ● B3; ○ Hastelloy X. Tested at 1173 K.

a bonding time of 38.4 ks was employed. However, B3 filler metal containing about 3 mass % boron appears to be better than the other fillers containing more than 3 mass % boron.

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