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Degradation in Diffusion-welded Joint of Dissimilar-metal Combination at Elevated Temperatures†

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Abstract

When a welded joint between dissimilar metals is held at elevated temperatures, the diffusion in the vicinity of bond interface causes a change in the microstructure of bond. In the present paper, such change in the microstructure of bond and its effect on the joint strength have been investigated for diffusion-welded joints of tough pitch copper (TPC) to nickel (Ni) and of TPC to iron (Fe).

For diffusion-welded joints of TPC to Ni held at temperatures above 650°C, many particles of nickel oxide and voids were observed in the TPC base metal adjacent to the bond interface. The formation of these nickel oxide and voids were reduced remarkably when a base metal of oxygen free high conductivity copper was used instead of the TPC base metal. This result suggests that the nickel oxide forms by the reaction of nickel with oxygen involved in the TPC base metal and the nickel oxide promotes the formation of voids; i.e., the nickel oxide is considered to act as a nucleation site of void due to the Kirkendall effect. The formation of these nickel oxide and voids is considered to cause the degradation in the joint strength. The degradation in the joint strength was observed when the joint was subjected to the holding for at least 15 min at temperatures above 570°C.

When a joint of TPC to Fe was held at temperatures above 770°C, many iron-oxide particles (or precipitates of iron) were observed in the TPC base metal adjacent to the bond interface. However, in contrast to the joint of TPC to Ni no void was observed in the joint of TPC to Fe. This result can be related to the fact that the amount of voids due to the Kirkendall effect is much less in the joint of TPC to Fe than in the joint of TPC to Ni. The degradation in the joint of TPC to Fe at elevated temperatures is considered to be very slight compared with that in the joint of TPC to Ni, since the formation of void is not observed in the joint of TPC to Fe.

KEY WORDS: (Diffusion Welding) (Copper) (Nickel) (Iron)

1. Introduction

When a welded joint of dissimilar-metal combination is used at elevated temperatures, the diffusion of atoms in the vicinity of bond interface causes a change in microstructure, which in many cases results in the degradation of the joint efficiency. The diffusion welding is a suitable method for joining the dissimilar-metal combination[1-3]. There have been many investigations[1-8] reported of the diffusion welding of dissimilar-metal combination. However, almost all of them were concerned with the joint efficiency and/or the microstructure of bond in the as-welded state; that is, for the diffusion welding between dissimilar metals there has been only a little amount of information about the change in the microstructure of bond which occurs in the course of holding the joint at elevated temperatures. The authors[9] have already shown

that the diffusion-welded joint of titanium to aluminum which forms an intermetallic compound decreases in the joint strength when held at temperatures above 600°C.

The present investigation aims at elucidating the change in the microstructure of bond at elevated temperatures and its effect on the joint efficiency for diffusion-welded joints of tough pitch copper (TPC) to nickel (Ni) and of TPC to iron (Fe). The dissimilar-metal combinations used are those which do not form an intermetallic compound; the binary system of copper and nickel forms a continuous series of solid solution, whereas that of copper and iron forms a wide miscibility gap.

2. Experimental Details

Base metals used in the present investigation are tough

pitch copper (TPC), commercially pure nickel (Ni), iron (Fe) and oxygen free high conductivity copper (OFHC), whose chemical compositions are shown in **Table 1**. The base metal of OFHC was applied for comparison with that of TPC. As shown in **Fig. 1**, the base metals of TPC and OFHC were round bars 20 mm in diameter and 37 mm in length. The base metals of Ni and Fe were disks 20 mm in diameter and 3 mm in length. The diffusion welding was carried out with these base metals arranged as shown in Fig. 1. This arrangement of the base metals was employed in order to compensate the thermoelectric power which was generated at the interface of dissimilar metals on the electric resistance measurement described later.

Table. 1 Chemical compositions of the base metals of tough pitch copper (TPC), oxygen free high conductivity copper (OFHC), commercially pure nickel (Ni), and iron (Fe).
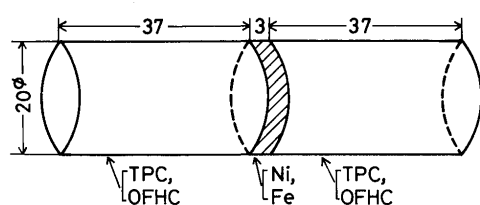
TPC

Fe	Ni	S	Ag	0	Pb	Cu
7	5	19	11	415	4	Bal.

OFHC

Fe	Ni	S	Ag	0	Pb	Sn	Cu
3	1	6	3	8	1	1	Bal.

Ni

Fe	Cu	Pb	Co	Mn	Si	C	S	Ni
400	30	10	100	1300	300	140	5	Bal.

Fe

| C	Si	Mn	P	S	Fe
90	800	2500	160	100	Bal.

Fig. 1 Arrangement of base metals for the diffusion welding.

The faying surface of the base metal was finished by grinding on emery paper of 1500 grade, and subsequently degreased by washing in acetone just before the welding. The diffusion welding was carried out using a similar apparatus as that reported in a previous paper[6]; the bond zone was heated with radiant-resistance heater of molybdenum foil 0.1 mm thick; the welding pressure was applied with hydraulic press. The temperature of the bond zone was monitored with CA thermocouple percussion-welded in the vicinity of the bond interface, and controlled to an accuracy of ±1°C. The welding was carried out in a vacuum of 2 ∼ 5 x 10⁻⁴ Torr.

Figure 2 shows a specimen to examine the change in the microstructure of bond which occurs in the course of holding at elevated temperatures; the specimen, which was cut from a joint shown in Fig. 1, was held at elevated temperatures in a vacuum of 10⁻⁵ Torr. The holding temperature was controlled to an accuracy of ±0.5°C by using a PR thermocouple percussion-welded to the specimen. The change in the microstructure at elevated temperatures was investigated with scanning electron microscope and electric resistance measurement across the bond interface.

The electric resistance measurement was carried out with the specimen kept in a vacuum furnace at elevated temperatures. **Figure 3** schematically shows the circuit for the electric resistance measurement. That is, a direct
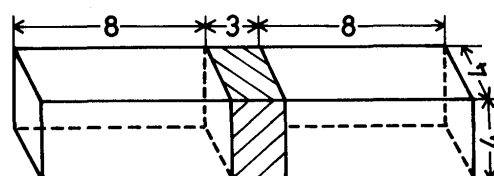
Fig. 2 Dimensions of the specimen.
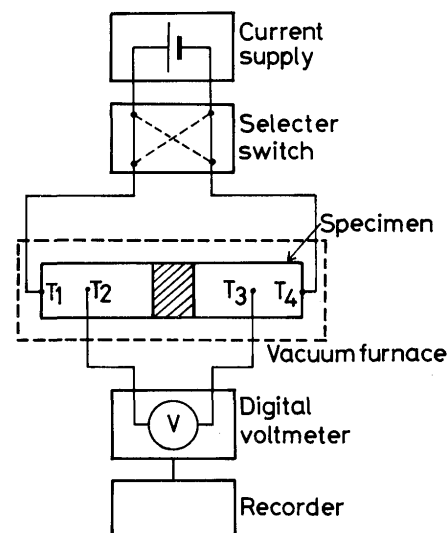
Fig. 3 Circuit for the electric resistance measurement of bond.

current of 100.00 ± 0.01 mA flowed between terminal T_1 and terminal T_4, and the potential difference which was generated between T_2 and T_3 by the current was measured to an accuracy of ±10 nV. Then the electric resistance R between T_2 and T_3 can be estimated from the current and the potential difference by using Ohm's law. In order to compensate the effect of thermoelectric power in the circuit on the electric resistance measurement, the direction of the current was reversed, and the mean value

of the potential difference for each direction was employed in estimating the electric resistance R. The increment of the electric resistance per a unit area ΔR is defined as;

$$\Delta R = \{R(t) - R(0)\} \cdot S/2, \qquad (1)$$

where $R(t)$ is the electric resistance R at a holding time t, $R(0)$ the initial value of $R(t)$, and S the cross-sectional area of the specimen. In the present paper, the variation of the electric resistance in the course of the high-temperature holding was expressed in terms of the electric resistance increment ΔR.

3. Results and Discussion

3.1 Joint of tough pitch copper to nickel

Figure 4 shows tensile strength of joints of TPC to Ni as a function of welding temperature. The joint strength took a maximum value at the welding temperature of
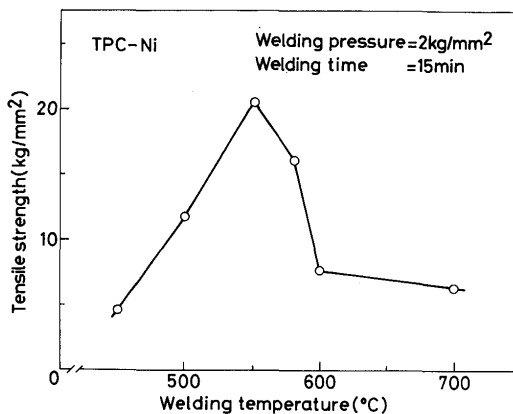

Fig. 4   Tensile strength of joints of TPC to Ni vs. welding temperature.

550°C, and above this temperature decreased with the rise of welding temperature. Generally, the strength of diffusion-welded joint increases with the rise of welding temperature, because the bond process is controlled chiefly by thermally activated process such as diffusion and creep deformation[1-3]. Therefore, the decrease in the joint strength with the rise of welding temperature above 550°C indicates that the joint strength was reduced by holding for at least 15 min (welding time = 15 min) at temperatures above 550°C.

In order to make clear the mechanism of the decrease in the joint strength with the rise of welding temperature, a joint welded at 550°C having the maximum strength was held at temperatures above 550°C, and the change in the

microstructure of bond was investigated with the electric resistance measurement. **Figure 5** shows the variation of the electric-resistance increment ΔR with holding time at temperatures from 650°C to 700°C. As shown in the figure, ΔR increased proportionally to the square root of holding time. In order to estimate the activation energy for the increase in ΔR, Arrhenius plots of holding time at which $\Delta R = 15$, 20 and 25 $p\Omega m^2$ are shown in **Fig. 6**. The activation energy was estimated to be $33 \sim 44$ kcal/mol increasing with the rise of ΔR. This result indicates that the increase in ΔR is due to a complex process controlled by plural factors. In the binary system of copper and nickel, the reported value of the activation energy for interdiffusion is $50 \sim 60$ kcal/mol[10] considerably higher than the activation energy obtained from Fig. 6

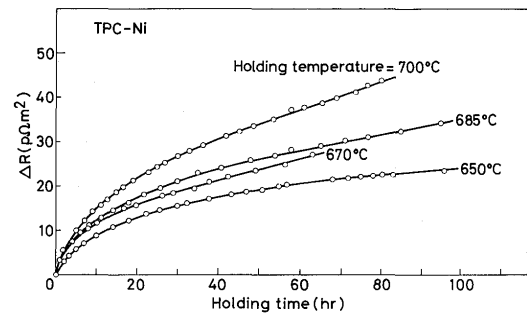Scanning electron micrographs of the microstructure of

Fig. 5   Variation of ΔR with holding time at temperatures from 650°C to 700°C for joints of TPC to Ni. The increment of the electric resistance of bond ΔR is given by eq. (1), where $R(t)$ is the resistance of bond at holding time t and S the cross sectional area of the specimen.
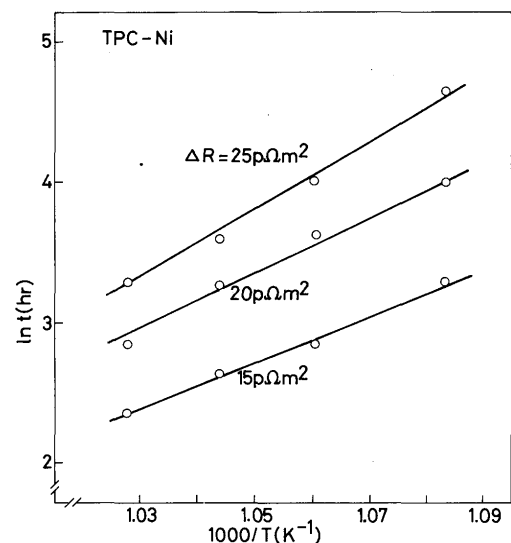

Fig. 6   Arrhenius plots of holding time at which $\Delta R = 25$, 20 and 15 $p\Omega m^2$. The activation energy estimated from the slope of lines are 33 kcal/mol for $\Delta R = 15$ $p\Omega m^2$, 39 kcal/mol for $\Delta R = 20$ $p\Omega m^2$ and 44 kcal/mol for $\Delta R = 25$ $p\Omega m^2$.

bond after holding at elevated temperatures are shown in Figs. 7(a) and (b). Many white particles and voids were observed in the TPC base metal adjacent to the bond interface. The number and volume of these white particles and voids were very small in the as-welded state, and increased with the increase in holding time at elevated temperatures. The increase in ΔR shown in Fig. 5 is considered to be due to the formation of these white particles and voids. In order to investigate the distribution of nickel in the bond zone, the intensity of the characteristic X-ray of nickel was analyzed along the white straight line in Fig. 7. As shown in the figure, the distribution curve of nickel had peaks at the white particles. The nickel content in the TPC was only 5 ppm as shown in Table 1, and the white particle was observed only in the TPC adjacent to the bond interface. These facts indicate that nickel atoms to form the white particles were supplied from the Ni base metal by diffusion. Nevertheless, the nickel concentration was higher at the white particle than in the matrix closer to the bond interface. On the basis of this result, the white particle is regarded as a stable nickel compound formed by reaction with a different kind of element. Copper and nickel form a continuous series of solid solution. Therefore, the white particle shown in Fig. 7 is considered to form by a reaction of nickel with impurity atoms contained by the TPC base metal.

Of all the impurities in the TPC the concentration of oxygen was the highest as shown in Table 1. In order to investigate the effect of oxygen on the change in the microstructure of bond at elevated temperatures, the base metal of OFHC was employed instead of the TPC base metal. As shown in **Fig. 8**, the increase in ΔR was much smaller for the joint of OFHC to Ni than for the joint of TPC to Ni. **Figure 9** shows a scanning electron micrograph of a joint of OFHC to Ni after holding for 100 hr at 700°C. The amount of white particles and voids was much smaller in the joint of OFHC to Ni than in the joint of TPC to Ni.

These results indicate that oxygen in the TPC is responsible for the increase in the electric resistance across the bond interface and the formation of white particles. Consequently, the white particle shown in Fig. 7 is considered to be nickel oxide forming by the reaction of nickel with oxygen in the TPC, because the affinity of nickel for oxygen is higher than that of copper. And the fact that the void also formed much more in the joint of TPC to Ni suggests that the nickel oxide promoted the formation of void. In fact, nickel oxides were observed inside some voids as shown in Fig. 7(b). The effect of the nickel oxide on the formation of void can be accounted for by the assumption that the oxide acts as a nucleation
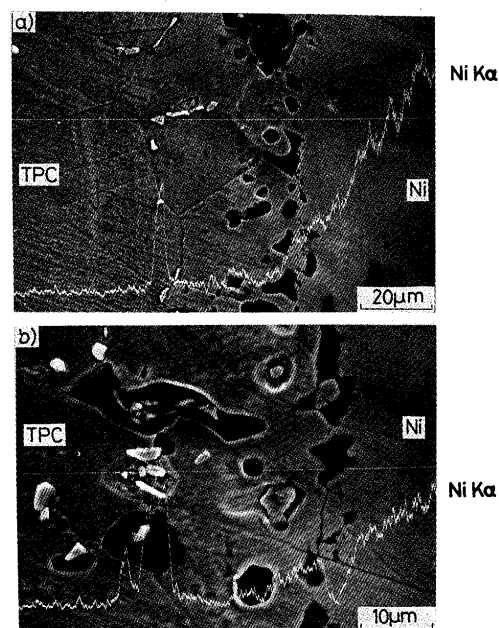

Fig. 7   Scanning electron micrographs of the bond of TPC to Ni subjected to holding at 650°C for 96 hr. The intensity of nickel characteristic X-ray (Kα) analyzed along the white straight line is also shown.
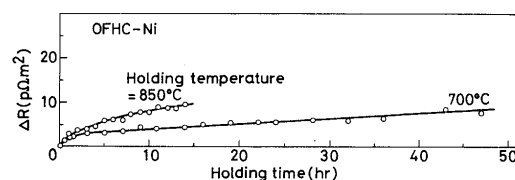

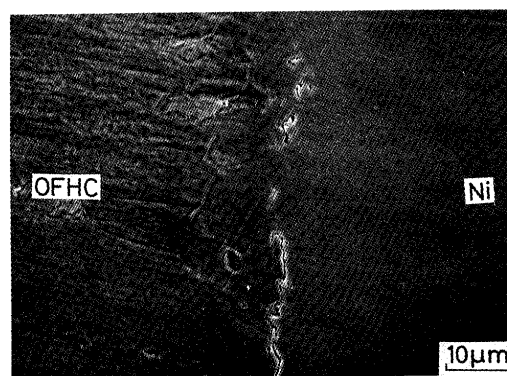**Fig. 8** ΔR vs. holding time for joints of OFHC to Ni.

Fig. 9   Scanning electron micrograph of the bond of OFHC to Ni held for 100 hr at 700°C.

site of void due to the Kirkendall effect or that the void is a trace of oxide removed by polishing for the preparation of metallographic examination.

If the void forms owing to the Kirkendall effect, a marker inserted between the faying surfaces will be displaced to the copper side in the high-temperature

holding. The displacement of marker was investigated using a joint in which molybdenum wire 20 μm in diameter was inserted as a marker. As shown in **Fig. 10**, the marker in a joint of TPC to Ni shifted to the TPC side with the increase in holding time. This indicates that vacancies flow more from Ni to TPC than from TPC to Ni and so voids due to the Kirkendall effect form in the TPC. Therefore, it is considered that the Kirkendall effect contributes more or less to the formation of voids in the course of the elevated-temperature holding.

Thus, when a joint of TPC to Ni was held at elevated temperatures, nickel oxide formed in the TPC by the reaction of nickel with oxygen in the TPC, and the oxide promoted the formation of voids due to the Kirkendall effect. The formation of these oxide and void is considered to reduce the joint strength. The oxide and void were also observed in the as-welded state as shown in **Fig. 11**. The decrease in the joint strength with the rise of welding temperature as shown in Fig. 4 is presumably attributed to the formation of these oxide and voids[11].

3.2 Joint of tough pitch copper to iron

Figure 12 shows tensile strength of joints of TPC to Fe as a function of welding temperature. The joint strength increased with the rise of welding temperature. Joints welded at temperatures above 600°C had a strength not less than that of the TPC. In contrast to the diffusion welding of TPC to Ni, no degradation in the joint strength with the rise of welding temperature was observed for the diffusion welding of TPC to Fe.

Fig. 12 Tensile strength of joints of TPC to Fe vs. welding temperature.

Figure 13 shows the variation of ΔR with holding time at 700°C for a joint welded at 650°C. The broken line in the figure presents the variation of ΔR for a joint of TPC to Ni at the same holding temperature. As shown in Fig. 13, the increase in ΔR was much smaller in the joint of TPC to Fe than in the joint of TPC to Ni.

Figure 14 shows scanning electron micrographs of a joint in the as-welded state (a) and after holding for 98 hr at 800°C (b). As shown in Fig. 14(b), many white particles in which the concentration of iron was higher than the matrix were observed in the TPC adjacent to the bond interface. Since iron has a higher affinity for oxygen than
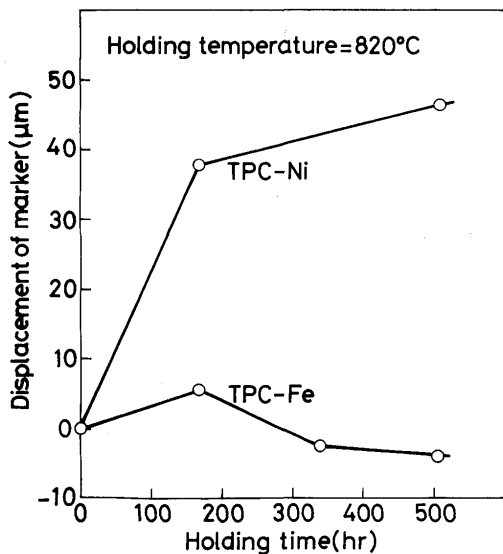

Fig. 10 Displacement of marker vs. holding time at 820°C for joints of TPC to Ni and of TPC to Fe. Displacement to copper side is defined as positive.
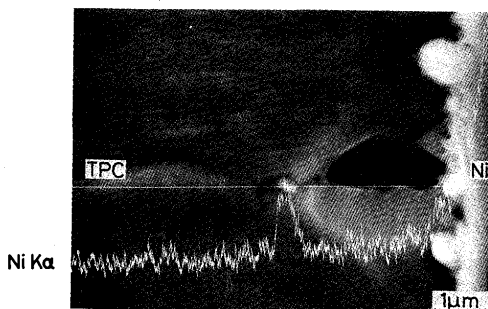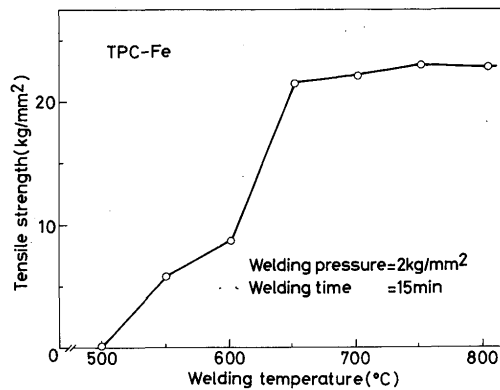

Fig. 11 Scanning electron micrograph of the bond of TPC to Ni in as-welded state ($T_W = 550°C$, $P_W = 2$ kg/mm^2, $t_W = 15$ min).
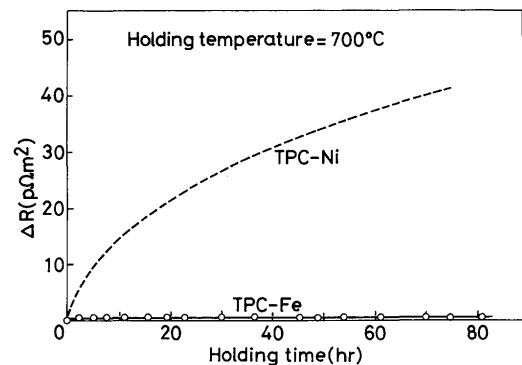

Fig. 13 ΔR vs. holding time at 700°C for the joint of TPC to Fe (—O—) and that of TPC to Ni (— — — —).
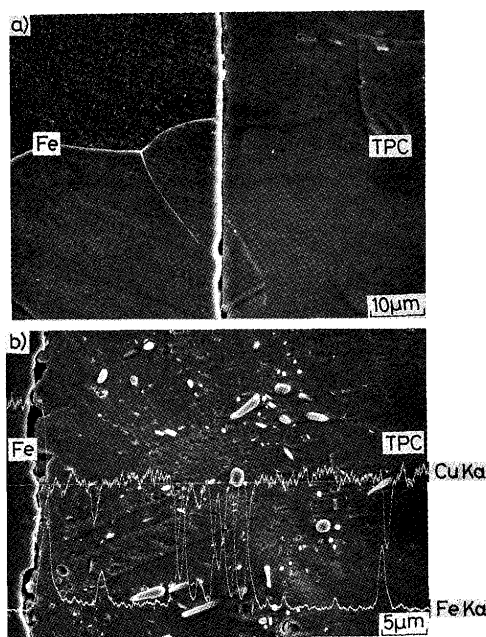
Fig. 14   Scanning electron micrographs of bonds of TPC to Fe in as-welded state(a) and after holding for 98 hr at 800°C(b).

copper, iron oxide is considered to form in the joint of TPC to Fe similarly to the nickel oxide in the joint of TPC to Ni. On the other hand, since the solubility of iron in copper decreases with the decrease in temperature[12], the precipitation of iron will occur in the TPC in the cooling process subsequent to the elevated-temperature holding. Therefore, the white particle in Fig. 14(b) is considered to consist of both iron oxide and iron precipitate, though the distinction between them is not possible.

On the other hand, no void was observed in the joint of TPC to Fe in contrast to the joint of TPC to Ni. This result is presumably related to the fact that the displacement of marker due to the Kirkendall effect was very small in the joint of TPC to Fe as shown in Fig. 10; for the amount of voids forming by the Kirkendall effect increases with the increase in the displacement of marker. The absence of void in the TPC is regarded as one main reason why the increase in ΔR was very small and the marked degradation in the joint efficiency was not observed in the joint of TPC to Fe.

4. Conclusion

When a welded joint of dissimilar-metal combination is used at elevated temperatures, the diffusion of atoms in the vicinity of bond interface causes a change in the microstructure, which in many cases results in the degradation of the joint efficiency. The present investigation is concerned with such change in the microstructure of diffusion-welded joints of TPC to Ni and of TPC to Fe with particular reference to its effect on the joint efficiency. The microstructure of bond was investigated with the electric resistance measurement across the bond interface and scanning electron microscope. Results obtained are summarized as follows:

(1) In the diffusion welding of TPC to Ni, the joint strength took a maximum value at the welding temperature of 550°C, and above this temperature decreased with the rise of welding temperature (the welding time was 15 min and the welding pressure 2 kg/mm^2).

(2) When the joint of TPC to Ni having the maximum strength was held at temperatures above 650°C, nickel-oxide particles and voids formed in the TPC adjacent to the bond interface. The formation of these nickel oxide and void is considered to reduce remarkably the joint strength.

(3) The amount of nickel oxides and voids forming in the TPC was reduced remarkably by using a base metal of OFHC instead of the TPC. On the basis of this result, the formation of nickel oxide is attributed to the reaction of nickel with oxygen involved in the TPC. The nickel oxide is considered to act as a nucleation site of void due to the Kirkendall effect and to promote the formation of the void.

(4) For the diffusion welding of TPC to Fe, the joint strength increased with the rise of welding temperature, and joints welded at temperatures above 650°C had a strength not less than that of the TPC base metal.

(5) When a joint of TPC to Fe welded at 650°C was held at temperatures above 700°C, many particles of iron oxide were observed in the TPC adjacent to the bond interface. And it seems that the particle consists partially of iron precipitate. However no void was observed in contrast to the joint of TPC to Ni. The absence of the void is explained by the fact that the amount of void forming by the Kirkendall effect was very small in the joint of TPC to Fe.

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