

High Temperature Brazing for Stainless Steel SUS 316 L with Pd-Cu Brazing Filler Metals*

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Abstract

Newly developed palladium-containing brazing filler metals, 30 Pd-60 Cu-10 Co, 30 Pd-60 Cu-10 Ni, and 30 Pd-50 Cu-10 Ni-10 Co (all in wt-%), were studied for use in brazing stainless steel SUS 316 L at high temperatures. Palladium-containing filler metals were selected because of their ductility, oxidation resistance, relatively high melting points, and lower cost than gold-based brazing filler metals. Wettability, microstructures and reactions were studied with experimental brazed joints between base and filler metal. Joint strengths, tensile strengths at room and elevated temperatures of 473K to 1073K and creep rupture strengths at 673K to 1073K, were investigated. The joints brazed with the 30 Pd-50 Cu-10 Ni-10 Co filler metal had superior tensile strengths at elevated temperature and creep rupture strengths. The joints with this filler metal had tensile strengths of 327 MPa to 429 MPa at temperatures of 293K to 873K.

Key Words: Palladium-containing brazing filler metals, Stainless steel SUS 316 L, Tensile testing, Joint strength, High temperature strength, Creep rupture strength

Introduction

Brazing filler metals for high temperature brazing are nickel-based, gold-based, and palladium-containing ones selected, because of their high strength at elevated temperatures, and excellent corrosion and oxidation resistance properties. Compared with nickel-based filler metals, the gold-based and palladium-containing noble filler metals exhibit lower hardness, better ductility, less tendency towards intergranular penetration, and no intermetallics, but are more expensive.

Many reports of studies on joints brazed with nickel-based and gold-based filler metals have been published (Refs. 1-9), but reports of joints brazed with palladium-containing filler metals have been relatively few. In this research, a study was made to develop palladium-containing filler metals with strength properties superior to those of nickel-based and gold-based filler metals, and of lower cost than gold-based filler metals.

This study was undertaken to develop three types of palladium-containing filler metals, 30 Pd-60 Cu-10 Co, 30 Pd-60 Cu-10 Ni, and 30 Pd-50 Cu-10 Ni-10 Co. The metallurgical and mechanical properties of stainless steel SUS316L joints brazed with these filler metals were investigated using a resistance-heated brazing vacuum furnace with particular attention paid to high-temperature strength. The microstructure and mechanical properties of the joints will be discussed and the most suitable filler metal for high temperature brazing will be decided in this study.

Base and Filler Metals

The base metal used in this research is a commercially available austenitic stainless steel SUS 316 L (ASTM: S31603). The chemical compositions and mechanical properties of the base metal are shown in Table 1 and Table 2 respectively. The newly developed palladium-containing filler metals are 30 Pd-60 Cu-10 Co, 30 Pd-60 Cu-10 Ni, and 30 Pd-50 Cu-10 Ni-10 Co (all compositions are given in wt-%). The compositions of the brazing filler metals were decided using ternary phase diagrams (Ref. 10), to determine their capability to be made into foil by rolling. Nickel-based filler metal Ni-Cr-W-Fe-B-Si was used for comparison with the joint properties of these palladium-

Table 1 Chemical compositions of base metal SUS 316 L

C	Si	Mn	P	S	Cr	Ni	Mo	Fe
0.017	0.49	1.37	0.032	0.002	16.85	12.26	2.03	Bal

Table 2 Mechanical properties of base metal SUS 316 L

Yield strength (0.2% Offset)	314 (MPa)
Tensile strength	539 (MPa)
Elongation	55 (%)
Hardness	75 (HRB)

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Table 3 Chemical compositions of brazing filler metals

	Pd	Cu	Co	Ni	Cr	W	Fe	B	Si
30Pd-60Cu-10Co	30.0	60.0	10.0	----	----	----	----	----	----
30Pd-60Cu-10Ni	30.0	60.0	----	10.0	----	----	----	----	----
30Pd-50Cu-10Ni-10Co	30.0	50.0	10.0	10.0	----	----	----	----	----
Ni-Cr-W-Fe-B-Si	----	----	----	Bal.	11.0	7.6	4.3	2.2	1.5

Table 4 Solidus and liquidus temperatures of brazing filler metals

Filler metal	Solidus	Liquidus
30Pd-60Cu-10Co	1363 (K)	1373 (K)
30Pd-60Cu-10Ni	1412	1423
30Pd-50Cu-10Ni-10Co	1403	1433
Ni-Cr-W-Fe-B-Si	1283	1438

containing filler metals. The chemical compositions and the solidus and liquidus of the brazing filler metals are shown in Table 3 and Table 4 respectively.

Brazing and Experimental Procedures

Figure 1 shows the schematic of a brazed joint. The surfaces to be brazed were polished in direction z with #400 emery paper, and the base metals and filler metals were ultrasonically-washed in acetone. For brazing, two base metals were butted lengthwise as shown in Fig. 1 and set in a brazing jig after the brazing filler metal was inserted between them. Brazing was conducted in a resistance-heated vacuum furnace with vacuum ranging from 2.6×10^{-3} to 6.7×10^{-3} Pa. Table 5 shows the brazing conditions for each brazing filler metal.

After brazing, excess brazing filler metal around the brazed area was removed and the microstructure at the brazed joint was examined by an optical microscope. The content distribution of the brazed joint was then investigated with an electron microprobe

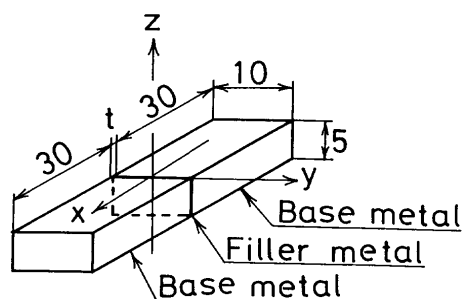


Fig. 1 Dimensions of brazed specimen

Table 5 Brazing conditions

Filler metal	Brazing temp.	Brazing time	Pressure
30Pd-60Cu-10Co	1403 (K)	5 (min)	2.6×10^{-3} 5 6.7×10^{-3} (Pa)
30Pd-60Cu-10Ni	1443		
30Pd-50Cu-10Ni-10Co	1453		
Ni-Cr-W-Fe-B-Si	1448		

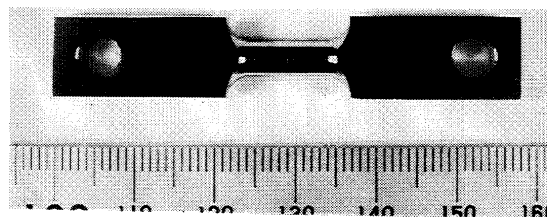


Fig. 2 Tensile test specimen for elevated temperature and creep rupture test specimen

analyzer (EPMA). The microhardness distribution of the brazed joint was also studied using a micro-Vickers hardness meter at a load of 25 g. Furthermore, to investigate the joint strength, tensile tests were carried out using an Instron material test machine at room temperature and elevated temperatures of 473K, 673K, 873K, and 1073K. Creep rupture tests of brazed joints were performed at 673K, 873K, and 1073K, and the time to failure was recorded. Brazed pieces were used as tensile specimens for the room temperature tests, and those with brazed central parts machine-ground to rods of 4.5 mm in diameter and about 10 mm in length, as shown in Fig. 2, were used as the tensile test pieces for the high temperature tests.

Results and Discussion

Wetting Behavior

Wetting tests were performed by putting 0.1 g of brazing filler metal on a 25 mm square SUS 316 L plate in the vacuum furnace for holding times of 5 min, 10 min, and 20 min at each brazing temperature. The spread areas of brazing filler metal were recorded as a function of holding time. The results of the wetting

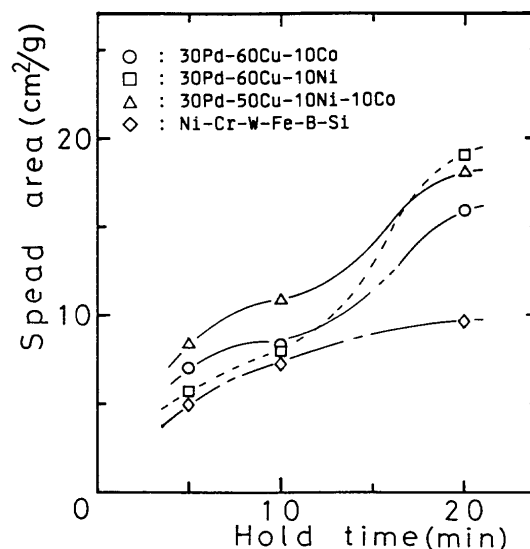


Fig. 3 Relation between spread area and hold time of each brazing filler metal

tests on SUS 316 L are shown in Fig. 3. Compared with the Ni-Cr-W-Fe-B-Si filler metal, all of the palladium-containing filler metals had better wettability.

Microstructure and Microhardness

Figure 4 shows the microstructure, electron microprobe traces, and microhardness of the joint brazed with the 30 Pd-60 Cu-10 Co filler metal. Figure 4a is the microstructure of a typical brazed joint. The base metal at the interface was dissolved by molten filler metal. The interface reaction area which etched dark, was identified using EPMA as the Co-rich phase. This fine Co-rich phase also appeared in the brazed zone. The microhardness pattern was measured through the brazed joint as shown in Fig. 4c. The interface reaction area was harder than the brazing zone because of the presence of a Co-rich phase at the interface.

Figure 5 shows the results about the brazed joint

with the 30 Pd-60 Cu-10 Ni filler metal. The microstructure of the joint brazed with 30Pd-60Cu-10Ni is shown in Fig. 5a. The grain boundaries of the base metal at the interface were dissolved by molten filler metal. This brazed joint shows a homogeneous structure at the brazed zone, with a darker area appearing at the central brazed zone. The result of electron microprobe analyses as shown in Fig. 5b shows that Cu content has increased in this area, and Fe content decreased. The hardness of the homogeneous structure near the interface reached 1.7 times the hardness of the base metal, because Fe had diffused into this zone.

Figure 6 shows the microstructure, electron microprobe analysis, and hardness of the joints brazed with the 30 Pd-50 Cu-10 Ni-10 Co filler metal. The microstructure of this joint presents a light microscopic homogeneous structure in the brazed zone, and the interface was not dissolved by molten brazing filler metal. The line traces by EPMA indicate that the Ni

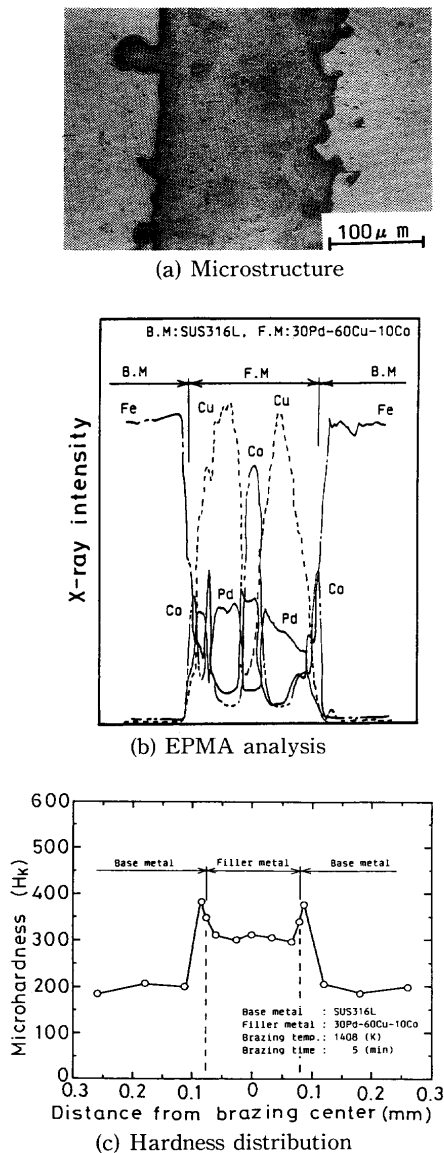


Fig. 4 Microstructure, EPMA analysis, and hardness distribution of brazed joint with 30 Pd-60 Cu-10 Co

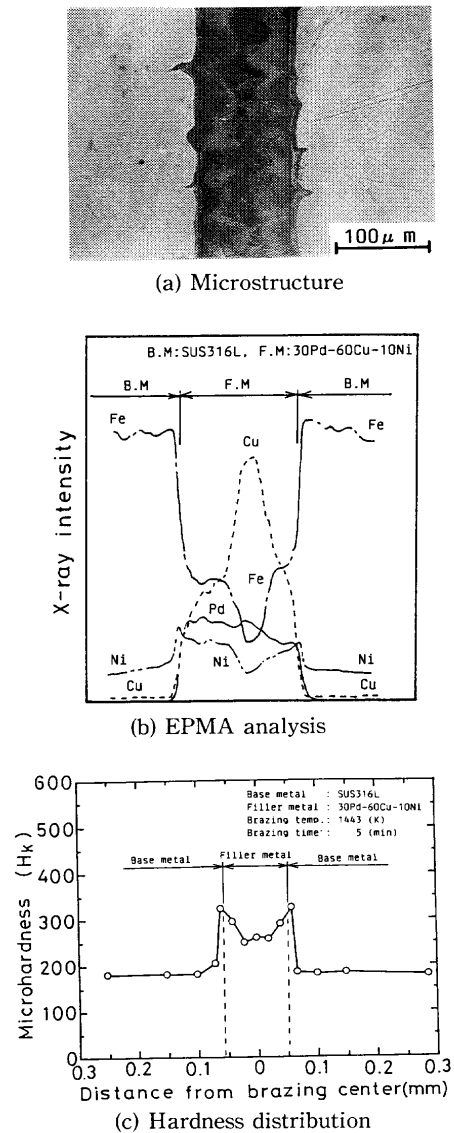


Fig. 5 Microstructure, EPMA analysis, and hardness distribution of brazed joint with 30 Pd-60 Cu-10 Ni

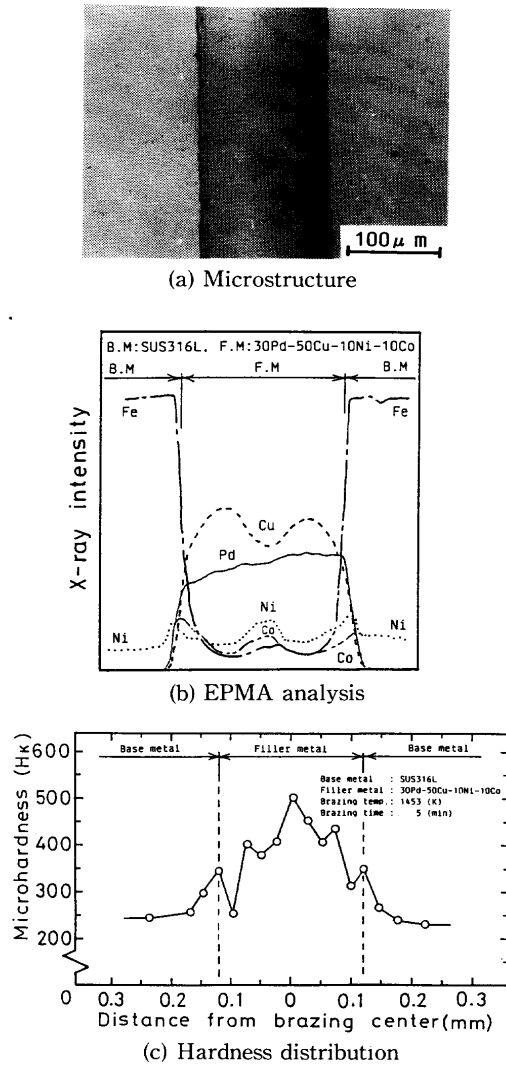


Fig. 6 Microstructure, EPMA analysis, and hardness distribution of brazed joint with 30 Pd-50 Cu-10 Ni-10 Co

and Co contents increased at the interface and the center of the brazed zone, but Cu slightly decreased at the center of the brazed zone. The microhardness increased at the interface and the center of the brazing zone, because of high Ni and Co contents at these areas.

Figure 7 exhibits the microstructure, EPMA analysis, and hardness pattern of the joints brazed with the nickel-based filler metal Ni-Cr-W-Fe-B-Si. A darker phase can be seen at the interface, where the filler metal has diffused into the grain boundaries of the base metal. These areas show high Cr content by EPMA, therefore, boride precipitations appear at this zone. A fine spherical phase was exhibited in the brazed zone, and the Cr and W contents increased in this phase, indicating that boride $W_{3.2}Cr_{1.8}B_3$ had formed in this zone. The hardness of this zone reach 3.4 times the hardness of the base metal, because of the presence of the hard boride.

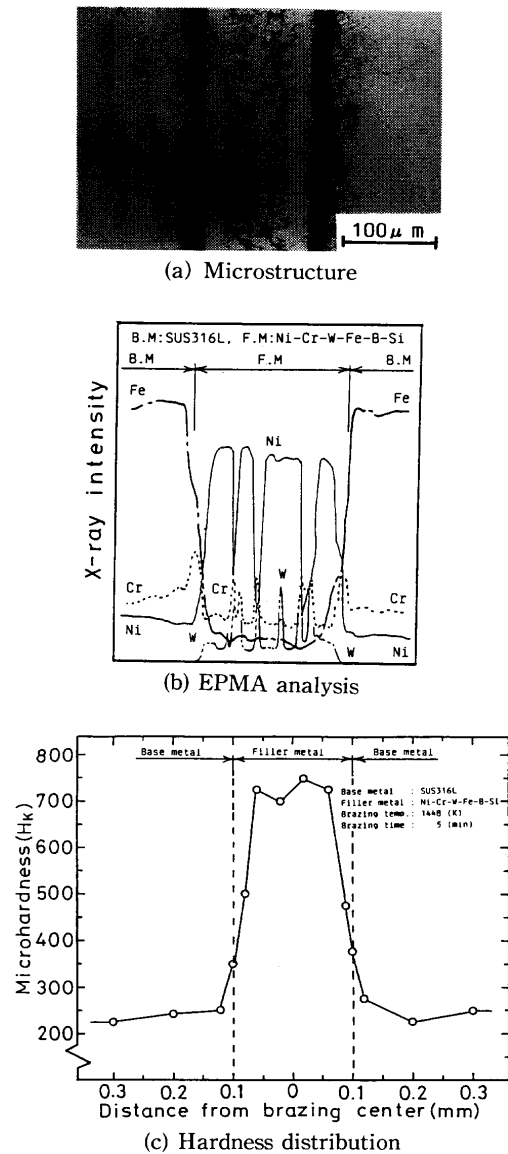


Fig. 7 Microstructure, EPMA analysis, and hardness distribution of brazed joint with Ni-Cr-W-Fe-B-Si

Mechanical Tests

Tensile Strengths of Joints at Room Temperature

The dependence of joint strengths on the brazing clearance was established by tensile strength/clearance graph -Fig. 8. Joints brazed with the 30 Pd-60 Cu-10 Co filler metal had the highest tensile strength of the tested filler metals. All the joints fractured at the brazed zone. The mean tensile strength of the clearances ranging from 0.07 to 0.15 mm was 536 MPa. Figure 9 shows the microstructure and SEM image of a joint having the maximum tensile strength of 548MPa at the clearance of 0.08 mm. From these results, it is seen that fracture occurred at the brazed zone, and the fractured surface had high plastic deformation. The joint strengths with 30 Pd-60 Cu-10 Ni and 30 Pd-50 Cu-10 Ni-10 Co filler metals are nearly equal at clearances, ranging from 0.05 to 0.22 mm.

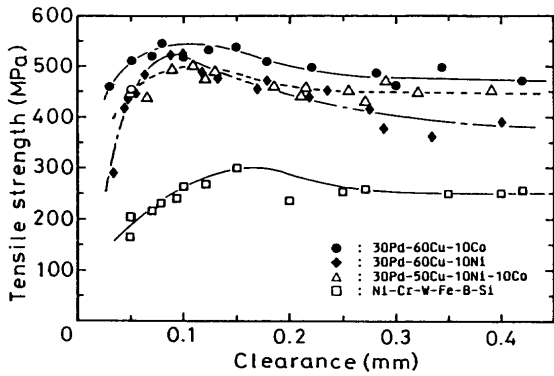


Fig. 8 Relation between tensile strength and joint clearance

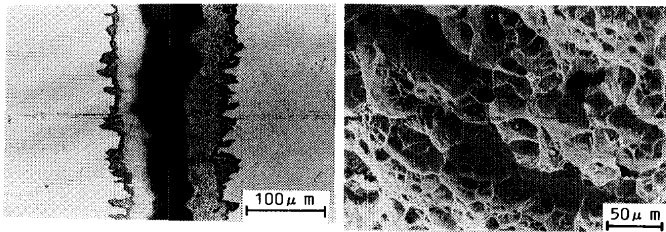


Fig. 9 Cross section and scanning electron micrograph of tensile test specimen brazed with 30 Pd-60 Cu-10 Co (a) Cross section (b) SEM

The tensile strengths of joints brazed with Pd-containing filler metals were about 1.7 times the strength of the Ni-Cr-W-Fe-B-Si filler metal joint. Considering a brittle-appearing fractured surface with poor deformation of the joint, a hard brittle boride seems to have formed at the joint brazed with Ni-Cr-W-Fe-B-Si filler metal.

Tensile Strengths of Joints at Elevated Temperature

Figure 10 represents the tensile strengths of the joints in relation test temperature. Joints brazed with 30 Pd-60 Cu-10 Co at 473K showed the highest tensile strength (488 MPa) of the tested filler metals. Joints brazed with 30 Pd-50 Cu-10 Ni-10 Co had the highest strength values ranging from 145 MPa to 462MPa at temperatures ranging from 673K to 1073K. The tensile strengths of joints brazed with Ni-Cr-W-Fe-B-Si filler metal gradually dropped at higher test temperatures, but these strengths were significantly lower than those of the Pd-containing filler metals at 673K or

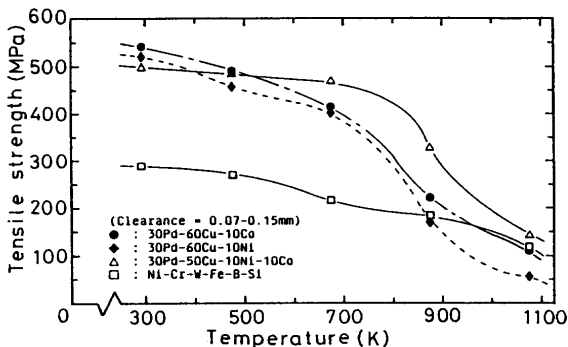


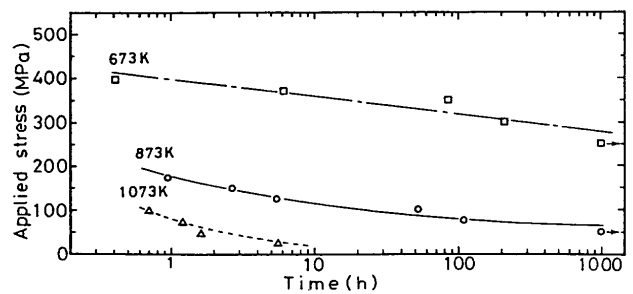
Fig. 10 Tensile strength of brazed joints at each temperature

below.

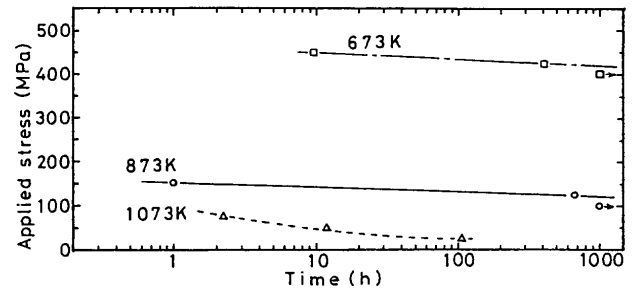
Joints brazed with 30 Pd-50 Cu-10 Ni-10 Co maintained a value approximately equal to the tensile strength at room temperature at 673K or below, and the tensile strengths at 673K or above had the maximum values in the filler metals used. This is because the 30 Pd-50 Cu-10 Ni-10 Co filler metal had a lower Cu content than the other Pd-containing filler metals, and its brazed joint did not form the high Cu content phase that appeared in the brazed zones of the 30 Pd-60 Cu-10 Co and 30 Pd-60 Ni-10 Co filler metals.

Creep Rupture Test Results

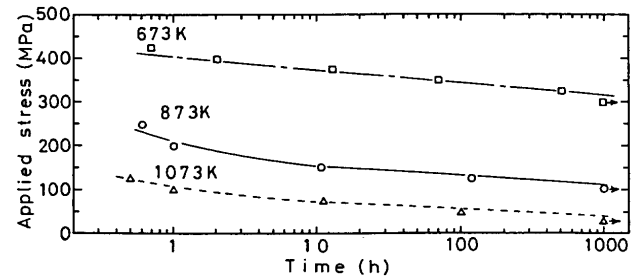
Figure 11 depicts the results of creep rupture tests in a temperature range of 673K to 1073K, and the rela-



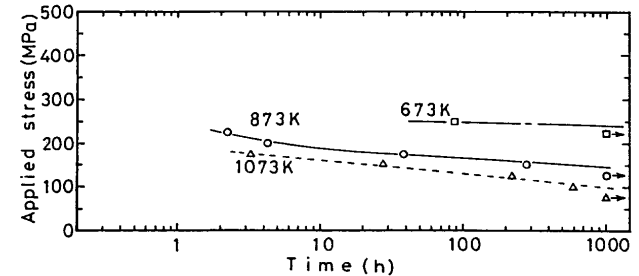
(a) 30 Pd-60 Cu-10 Co (Clearance: 0.08-0.12 mm)



(b) 30 Pd-60 Cu-10 Ni (Clearance: 0.09-0.12 mm)



(c) 30 Pd-50 Cu-10 Ni-10 Co (Clearance: 0.05-0.10 mm)



(d) Ni-Cr-W-Fe-B-Si (Clearance: 0.10-0.15 mm)

Fig. 11 Relation between applied stress and rupture time

tionships between applied stress and time to failure are shown in these figures. These tests continued until the time to failure reached 1000 hours. The arrows represent the time to failure of 1000 hours or over. Joints brazed with the 30 Pd-60 Cu-10 Ni filler metal had the highest creep rupture strength at 673K, registering a time to failure of 1000 hrs or over at 400 MPa applied stress. Joints brazed with the Ni-Cr-W-Fe-B-Si filler metal had the best creep rupture strengths with a time to failure of 1000 hrs or over at applied stresses of 125 MPa at 873K and of 75 MPa at 1075K respectively.

In the palladium-containing filler metals, joints brazed with the 30 Pd-50 Cu-10 Ni-10 Co filler metal had a longer time to failure at 873K and 1073K than joints brazed with the 30 Pd-60 Cu-10 Co and 30 Pd-60 Cu-10 Ni filler metals, because the 30 Pd-50 Cu-10 Ni-10 Co filler metal had a lower Cu content and a higher Ni content than the other Pd-containing filler metals.

Conclusions

Stainless steel SUS 316 L joints brazed with newly developed Pd-containing filler metals, 30 Pd-60 Cu-10 Co, 30 Pd-60 Cu-10 Ni, and 30 Pd-50 Cu-10 Ni-10 Co were investigated to determine the most suitable brazing filler metal and brazing conditions.

Joints brazed with the 30 Pd-60 Cu-10 Co filler metal had the highest tensile strength of the tested joints, and the mean tensile strength for joint clearances ranging from 0.07 to 0.15 mm was 536 MPa.

Joints brazed with the 30 Pd-60 Cu-10 Ni filler metal had the maximum tensile strength at 473K, and joints brazed with the 30 Pd-50 Cu-10 Ni-10 Co filler metal did at 673K, 873K and 1073K.

Joints brazed with the 30 Pd-60 Cu-10 Ni filler metal had the highest creep rupture strength at 673K, and joints brazed with the Ni-Cr-W-Fe-B-Si braze alloy did at 873K and 1073K.

It is concluded that 30 Pd-50 Cu-10 Ni-10 Co brazing filler metal is the most suitable filler metal for high temperature brazing of SUS 316 L.

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