Influence of brazing clearance on microstructure and properties of W-Cu/1Cr18Ni9 steel brazed joint with Ni-Cr-Si-B amorphous filler metal

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Abstract

W-Cu composite and 1Cr18Ni9 steel were brazed with Ni-based amorphous filler metal in a vacuum furnace. Four-point bending strength, fracture morphology, microstructure and elements distribution were studied by a series of standard methods. Then the influence of brazing clearance on microstructure and properties of the brazed joint were analyzed and discussed. Results indicated that, within the scope of the selected parameters, bending strength of the brazed joint increased significantly as reducing the brazing clearance. While the brazing clearance controlled to be 20 μm, the four-point bending strength performed the largest value, up to 333 MPa. The joint fracture showed a typical ductile fracture characteristics. Microstructure of the brazing seam was mainly composed of γ-Ni solid solution, W-Ni solid solution, CrB, and a part of eutectic phases of Ni_3Si and Ni_3B. The microstructure of brazed joint with brazing clearance 20 μm was more uniform and more stable. Microstructure and element of brazed joint almost transformed into single-phase solid solution. Meanwhile, micro holes disappeared and a number of brittle phases reduced, which provided well plastic reserves for the brazed joint.

K e y   w o r d s: W-Cu composite, brazing clearance, microstructure, fracture, bending strength

1. Introduction

W-Cu composite has excellent performance of tungsten and copper metal, not only showing high thermal and electrical conductivity and low coefficient of thermal expansion but also behaving features of favorable high-temperature strength, thermal shock resistance, erosion resistance, and dimensional stability [1–3]. W-Cu composite is a promising material in serve condition of high temperature and good thermal stability, usually being used in high-temperature components, heat sink materials and electrical contact materials [4–6].

While W-Cu composite and stainless steel being welded, it is easy to show the phenomenon of phase brittleness and stress concentration in the obtained joint, due to great difference existed in characteristics of physics and chemistry. All the above problems existed lead to increasing of fracture tendency and reducing mechanical properties of the brazed joint of W-Cu and stainless steel. Now considerable interest has been generated in brazing of W metal and W-Cu composite with amorphous filler metals [7–12]. Researchers from the Moscow Institute of Engineering and Physics have researched the bonding mechanism of dissimilar materials in brazing W alloy and steel with amorphous filler metals of Ni-based, Ti-based and Fe-based [13]. Amorphous filler metals showed a good promising property in improving the performance of the brazed joint.

In this study, W-Cu composite and stainless steel were brazed in a vacuum furnace with amorphous Ni-based filler metal. Ni-brazed amorphous filler metal exhibited excellent wettability and spreadability on the surface of stainless steel and hard alloy. Moreover, the brazed joint with Ni-brazed filler metal performed high strength, good toughness and features of oxidative stability, corrosion resistance and low-temperature toughness. The Ni-based amorphous filler

Table 1. Chemical compositions of 1Cr18Ni9 stainless steel and Ni-Cr-Si-B amorphous filler metal

	Chemical compositions (wt.%)										
Material	C	Cr	Si	Mn	S	P	Ti	Co	B	Ni	Fe
1Cr18Ni9	0.018	17.50	0.59	0.97	0.0043	0.0278	0.13	0.14	–	9.27	Bal.
NiCrSiB		6–8	4 5	0.04	–	0.02	0.05	0.10	2.75–3.5	Bal.	2.5–3.5

metal has great potential in brazing W-Cu composite and stainless steel, to obtain dissimilar joint with a good performance at high temperature. The results provided the necessary theoretical foundation for preparation and application of composite component of W-Cu composite.

2. Materials and methods of experiment

Materials used in this study were W-Cu composite (W55-Cu45, wt.%) and 1Cr18Ni9 stainless steel, which were machined into cubes with dimensions of 20 mm × 20 mm × 5 mm. Amorphous foils of Ni-Cr--Si-B alloy were adopted as brazing filler metal, with melting range of 971–999 °C. Chemical compositions of 1Cr18Ni9 stainless steel and Ni-Cr-Si-B amorphous filler metal are shown in Table 1.

Before being brazed, the oxide and grease on the surface of W-Cu composite and 1Cr18Ni9 should be strictly cleared. The surface to be brazed should be smooth with a certain roughness. Ni-Cr-Si-B amorphous foils were prepared with dimensions of 20 mm × 5 mm × 0.02 mm.

Experimental material prepared was put into the special fixture with a sequence of W-Cu composite, brazing foil and stainless steel. The brazing processes were carried out in a vacuum furnace with process parameters of brazing temperature 1060 °C, holding time 30 min and vacuum level superior to 4×10^{-3} Pa. In the vacuum brazing process, a stationary pressure of 40 kPa was imposed on the surface of 1Cr18Ni9 steel, for making the filler metal infiltrate and spread better in the gap of the base metals. Two special struts with certain size were placed between W-Cu composite and 1Cr18Ni9 steel to control the brazing clearance accurately. After brazing, the metallographic samples with dimensions of 10 mm × 5 mm × 5 mm were cut out via electric discharge cutting machine. Then the prepared samples were corroded with the blend solution of $FeCl_3 : HCl : H_2O = 5$ g : 10 ml : 100 ml. Microstructure and morphology of brazed interface were observed and analyzed using scanning electron microscope (JSM-6480). Then, distribution of elements in characteristic point and interface region were analyzed using energy disperse spectroscopy of INCA (EDS). Final four-point bending strength was tested by mechanical properties testing machine (CMT5205) with a special fixture.
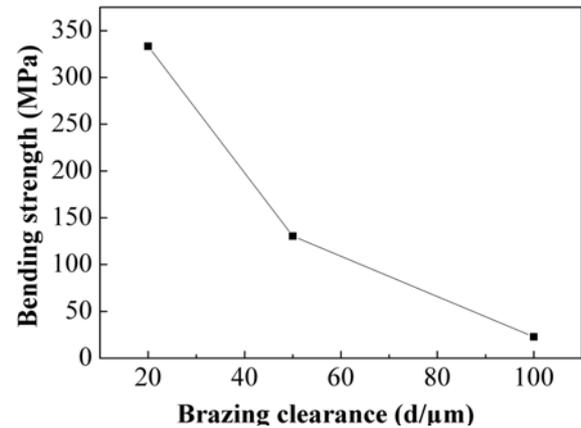

Fig. 1. Effect of the brazing clearance on bending strength of the brazed joint with Ni-based filler metal.

3. Results and discussion

3.1. Influence of brazing clearance on fracture feature of the joint

Four-point bending strength of the brazed joint of W-Cu composite and 1Cr18Ni9 steel with different clearance was tested at room temperature. The samples were prepared with dimensions of 40 mm × 5 mm × 0.02 mm. Results are shown in Fig. 1. As it can be seen, the four-point bending strength was significantly influenced by brazing clearance. As the brazing clearance increased at the same brazing temperature, the bending strength of Ni-based brazed joint showed a decreased tendency. The maximum bending strength of the brazed joint was 333 MPa with brazing clearance 20 μm. Fracture of the brazed joint showed the characteristic of ductile fracture, located at the side of W-Cu composite. While the brazing clearance being raised to 50 μm, bending strength of the joint decreased and the fracture located on the interface of stainless steel and brazing seam with the characteristic of cleavage fracture. The minimum bending strength of the brazed joint was showed in the joint with brazing clearance 100 μm. The fracture was located in the center of the brazing seam and showed characteristic of brittle fracture.

The fracture morphology of Ni-based filler metal with brazing clearance 20 μm is shown in Fig. 2. The characteristic of the brazing seam and W-Cu composite existed in the fracture interface. A mass of inten-
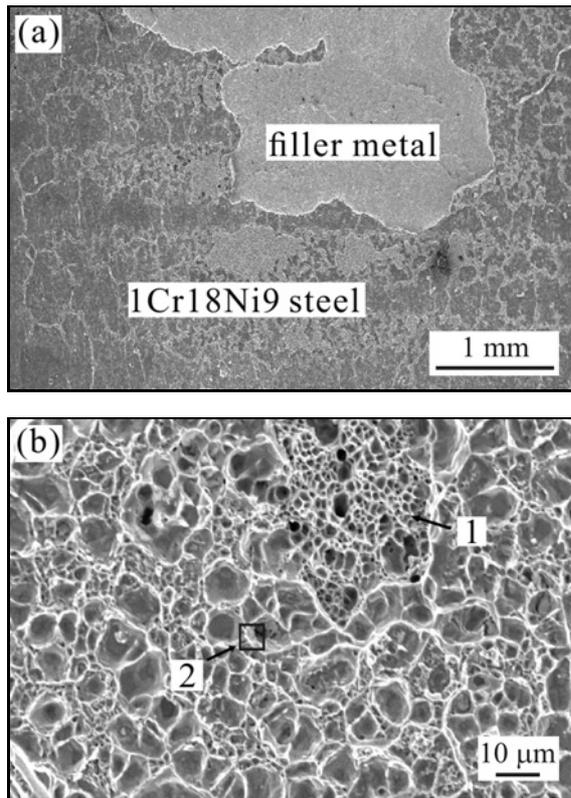
Fig. 2. Fracture morphology of W-Cu/1Cr18Ni9 steel brazed joint with clearance 20 μm thick: (a) fracture morphology at low magnification; (b) microfracture morphology.

Fig. 3. The microstructure of W-Cu/1Cr18Ni9 steel brazed joint with clearance 100 μm: (a) microstructure of the brazed joint; (b) micromorphology and feature regions.

sive and net dimples and particles of the second phase (strengthening phase) existed. The obvious phenomena of slip, tensile and tear were shown. During the progress of the brazed joint being yielded a mass of interface free energy was released, with an obvious plastic feature that was a typical characteristic of ductile fracture.

With the help of the energy spectrum analysis, it can be sure that microstructure in the brazed seam region was almost γ-Ni(Cu) solid solution, without brittle compounds of B-Cr and Ni-W being found. Composition and phases of brazing seam were uniform and stable, so bending strength of the brazed joint with brazing clearance 20 μm was better than that of other brazing clearance.

3.2. Influence of brazing clearance on microstructure of brazing seam region

Elements of Cr, Si, and B in amorphous filler metal were diffused in the brazing process and reacted with elements of the base metals, leading to some brittle phases formed. So metallurgy between brazing filler metal and base metal was sensitive to the width of the brazing seam. The microstructure of the brazed
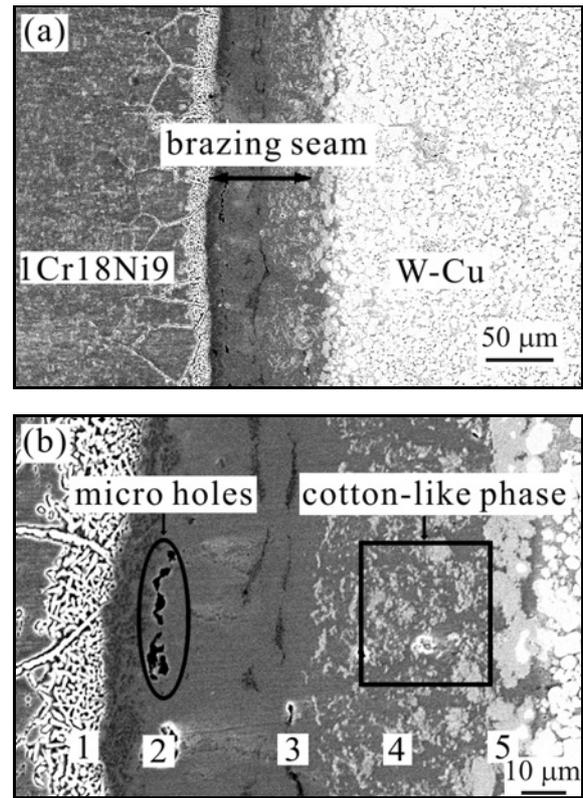
joint with Ni-based filler metal under three different brazing clearances of 100, 50 and 20 μm are shown in Figs. 3–5.

While the brazing clearance being controlled to be 100 μm thick, there was multilayer microstructure formed in the brazing seam as shown in Fig. 3a. The brazing seam region contained white and needle-like phase near the 1Cr18Ni9 steel side, dark-gray diffusion layer, interrupted and striped phases in the center of the brazing seam and gray and cotton-like layer near W-Cu composite side. Some micro holes existed in the brazing seam of stainless steel side. Part of B element in the filler metal diffused to the brazing interface then reacted with Cr element diffused from the stainless steel, forming the compound of CrB phase. While the temperature was being decreased, CrB phase precipitated in the form of supersaturated boride that was white and needle-like phases near stainless steel side as shown to be region 1 in Fig. 3b.

In addition to CrB phase, dark-gray and rod-like phases appeared following stainless steel interface, which were a eutectic phase of Ni_3Si and Ni_3B, according to the content of elements in Table 2. Referring to binary phase diagram [14], both B-Ni and Ni-Si binary phase system could occur eutectic reaction which formed solid solution of γ-Ni, Ni_3Si phase

T a b l e 2. Elements content of different regions in Fig. 3

Region	W	Cu	Ni	Cr	Fe	Si
1	–	–	6.06	27.26	65.15	–
2	–	–	38.40	16.45	43.85	1.30
3	–	8.39	70.81	11.09	5.07	4.64
4	31.72	9.59	29.22	8.93	20.54	–
5	80.12	8.49	9.86	1.52	–	–
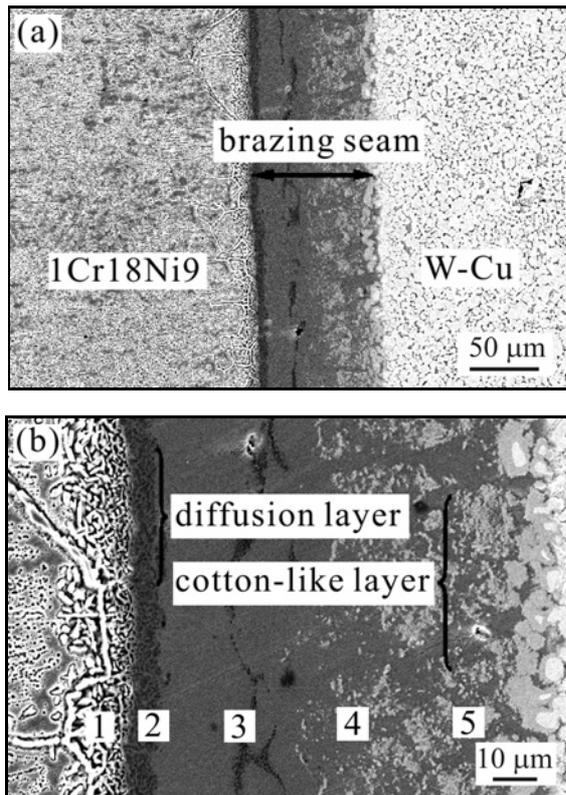
Fig. 4. The microstructure of W-Cu/1Cr18Ni9 steel brazed joint with clearance 50 μm: (a) microstructure of the brazed joint; (b) micromorphology and feature regions.

Fig. 5. The microstructure of W-Cu/1Cr18Ni9 steel brazed joint with clearance 20 μm: (a) microstructure of the brazed joint; (b) micromorphology and feature regions.

and Ni_3B phase, respectively shown as region-2 and region-3 in Fig. 3b. The size of brazing clearance was too big, so Cr element and Si element in the brazing seam could not completely diffuse across the brazing interface. Hence, some brittle and hard phases of boride and silicide still kept in the brazing seam region, which made the joint become fragile.

In the brazing seam region near the W-Cu composite side, cotton-like transition layer formed, which mainly contained W, Ni element and a small amount of Cu and Cr element. The solid solution of W(Ni) and Ni_2W phase was possible to be formed. Due to the abundant of Ni element diffused into W-Cu composite, Ni element of 5-region attained 80.12 % as shown in Fig. 3b. After the solid solution had reached supersaturation in the interface of brazed joint, redundant
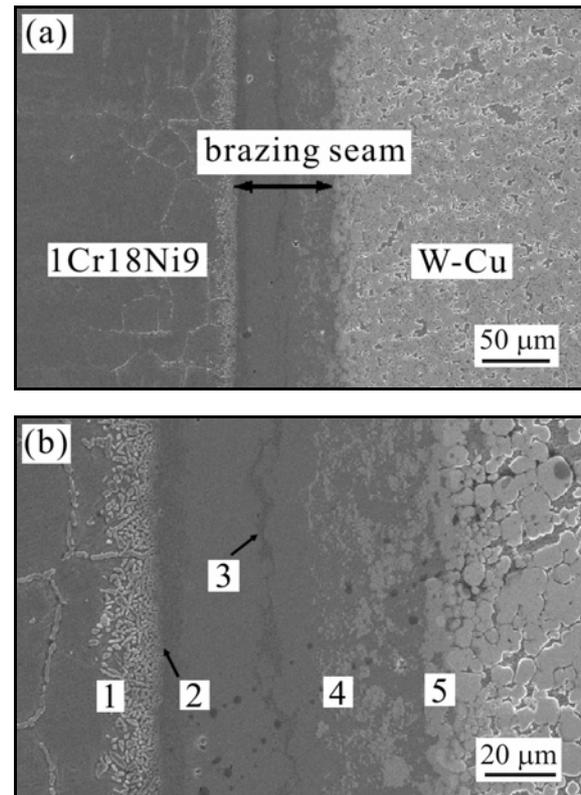
Ni elements and W would produce compounds that increased the tendency of brittleness in the brazing seam region.

The microstructure of the brazed joint and composition analysis of typical regions with brazing clearance 50 μm thick are shown in Fig. 4 and Table 3. As it can be seen from Fig. 4a, when the brazing clearance decreased from 100 to 50 μm, the microstructure of the brazing seam changed, and all of the holes near the region of stainless steel side completely disappeared. The white and needle-like phases of boride are reduced. The dark-gray and striped phases begun to be isolated in the brazing seam, which appeared interrupted distributions. The cotton-like layer in W-Cu composite side became thinner than that in the brazed joint with a clearance of 100 μm thick. So tissues of

T a b l e 3. Elements content of different regions in Fig. 4

Region	W	Cu	Ni	Cr	Fe	Si	Mn
1	–	–	7.92	22.35	67.46	0.70	1.56
2	–	4.82	65.30	6.63	19.35	3.90	–
3	–	–	48.14	43.33	5.51	3.03	–
4	30.43	9.51	51.04	4.52	2.31	2.19	–
5	77.85	3.61	15.75	2.79	–	–	–

T a b l e 4. Elements content of different regions in Fig. 5

Region	W	Cu	Ni	Cr	Fe	Si	Mn
1	–	–	7.92	22.35	67.46	0.70	1.56
2	–	4.82	65.30	6.63	19.35	3.90	–
3	–	–	48.14	43.33	5.51	3.03	–
4	52.26	9.51	19.21	24.51	2.31	–	–

the brazing seam were more stable and homogeneous, and microstructure morphology had been obviously improved.

While the brazing clearance controlled to be 50 μm thick, microstructure of brazing seam was composed of the Ni-based solid solution, a limited solid solution of W-Ni, a certain amount of eutectic phase of Ni_3Si, Ni_3B and CrB phase and a small amount of W-Ni compounds. However, diffusion distance of elements became shorter due to a thickness of the brazing seam decreased. While being brazed, B, Si, and other elements diffused into base metals more sufficiently. Therefore, an amount of brittle phases constantly reduced in the region of the brazing seam that decreased production and propagation of a crack in the four-point bending test. The four-point bending strength of the brazed joint increased nearly five times compared with brazed joint of 100 μm brazing clearance. The strength of brazed interface was obviously enhanced.

The microstructure of the brazed joint and composition analysis of typical regions with brazing clearance 20 μm thick are shown in Fig. 5 and Table 4. As it can be seen from Fig. 5a, in addition to three reaction zones (BCr, Ni_3Si, and Ni_3B, Ni-W), the dark-gray brittle layer almost completely disappeared in the brazing seam, which only had intermittent dark-gray and striped phases. There were abundant elements of Ni in the region-3, most of Ni participated in reacting and producing a solid solution of γ-Ni. According to binary phase diagram, Ni and Si elements would react and produce eutectic phase of Ni-Si. A small amount of Cr element could still exist in the form of the B-Cr compound at brazed layer. However, the content of Cr was limited which almost has not affected the joint strength.

Even thickness of brazing seam reduced to 20 μm, main transformation of elements diffusion was inter-crystalline diffusion, and elements of Si and B diffused into base metals, but there were still a few elements of Si and B existing at brazed layer. Moreover, CrB phase was difficult to be eliminated, due to its chemical stability. A few eutectic phases of Ni-B and Ni-Si were formed, existing in the form of discontinuous and striped phases of multiple compounds at brazed layer. The solid solution phase produced in connection region and near two sides of base metals formed a special pattern of "soft clip hard". This pattern could buffer stress and deformation, and improve the overall performance of the brazed joint. The bending strength of Ni-based brazed joint with a brazing clearance of 20 μm thick was 333 MPa; obviously higher than that of the brazed joint with brazing clearance 50 and 100 μm.

3.3. Influence of brazing clearance on elements distribution

Elements distribution of the brazed joint with brazing clearance 100 μm is shown in Fig. 6. Cr element from the filler metal was obviously gathered near the interface of stainless steel side. The peak intensity existed at the brazed seam, which located at the dark-gray and striped phase. Cr and Ni elements appeared obvious concentration gradient at the brazed interface. B element could be combined with Cr and Ni, and produced a series of compounds of B-Cr and eutectic phases of B-Ni. Ni element was mainly concentrated in brazed layer that produced the uniform solid solution of γ-Ni. When the elements of filler metal uniformly diffused into the base metal, Fe, W, and Cu from both sides of base metals appeared diffusion phenomenon at the brazed interface. Especially the diffusion phenomenon of granulated W crystal was obvious. Solid solution and compound phase could be formed between W and Ni elements, reducing the interfacial free energy of W-Cu composite side. However,
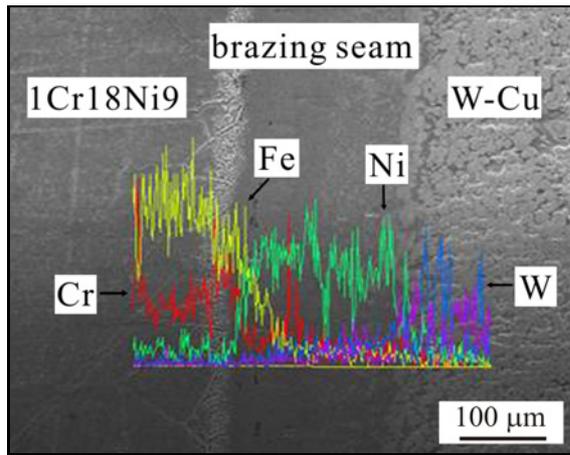
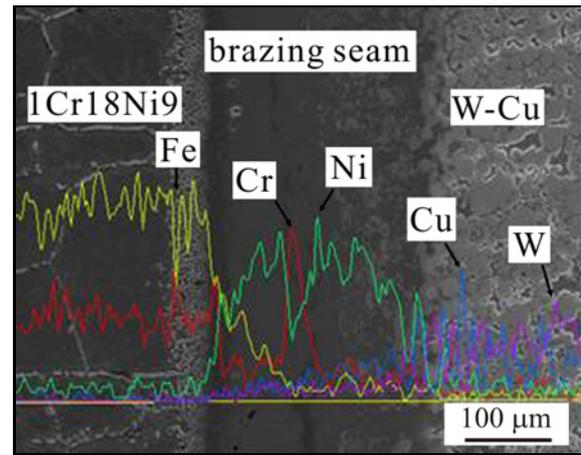Fig. 6. Elements diffusion of the brazed joint with brazing
clearance 100 μm.

Fig. 8. Elements diffusion of the brazed joint with brazing
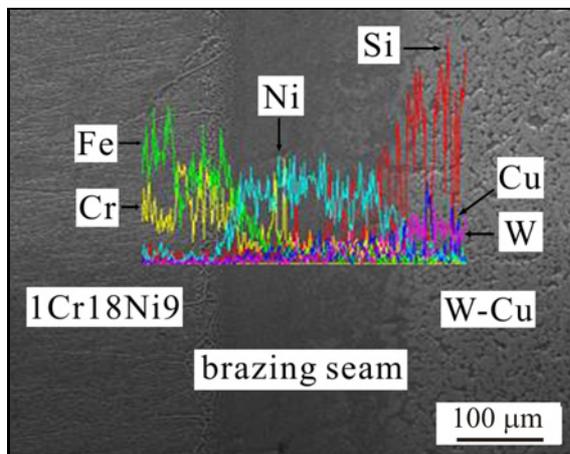clearance 20 μm.

Fig. 7. Elements diffusion of the brazed joint with brazing
clearance 50 μm.

the thickness of brazed layer was too wide to appear phenomenon of micro holes and lack of penetration in the brazing seam.

While the brazing clearance being controlled to be 50 μm, as it can be seen from Fig. 7, diffusion curve of Ni and Fe elements changed to be smooth and micro holes near the interface of stainless steel disappeared. This phenomenon stated that decreasing the brazing clearance could improve homogeneity and metallurgical stability of various elements at the brazed layer. The possibility of the brazing flaw was reduced, such as micro holes and crack, assured the stability of the brazed joint. Also, Si element from the filler metal diffused into the W-Cu composite, which improved the wettability and metallurgy during the brazing process.

While the brazing clearance being controlled to be 20 μm, as it can be seen from Fig. 8, the elements from the filler metal were easier to diffuse into the base metal, due to the shorter diffusion distance. From

the interface near stainless steel side to the center of brazed seam, the content of Cr element was decreased firstly and then increased, appeared a crest value in the brazing seam. Elements of W and Cu were not obvious solubility at brazed layer, which mainly concentrated on the brazed layer of W-Cu composite side. It provided a favorable condition for the solid solutions of W-Ni to form. Near the interface of stainless steel side, a transition layer mainly containing Fe and Cr elements was formed, ensured joining of brazed layer and stainless steel. Hence, the brazed joint with a clearance of 20 μm thick had higher strength and excellent mechanical properties.

4. Conclusions

1. As the brazing clearance was decreased from 100 to 20 μm while W-Cu composite and 1Cr18Ni9 steel were brazed with Ni-based filler metal, the bending strength changed from low to high. From the perspective of mechanical strength, the brazing clearance 20 μm was an optimal choice for brazing W-Cu and 1Cr18Ni9 steel. Meanwhile, the bending fracture was a typical characteristic of ductile fracture.

2. Microstructure of the brazed joint with brazing clearance 100 μm was mainly composed of γ-Ni solid solution, W-Ni solid solution phase, CrB, and a part of eutectic phases of Ni_3Si and Ni_3B. Moreover, the phenomenon of partial melting appeared. From the perspective of microstructure and element diffusion, the microstructure of brazed joint with brazing clearance 20 μm was more uniform and more stable. Microstructure and element of brazed joint almost transformed into single-phase solid solution. The quantity of borides, silicides, and nickel-tungsten obviously reduced, which provided well plastic reserves for the brazed joint. Microstructure and mechanical proper-

ties of brazed joint with brazing clearance 50 μm performed between that of the former two.

3. Diffusion of alloying element during the brazing process was changed by the brazing clearance. When the thickness of the brazing seam region was too wide, elements of B, Si from the Ni-based filler metal could not completely diffuse into base metals. In the brazing seam, the content of Ni solid solution exceeded its saturability, inducing brittle layer and brazing defect to be formed at the brazing interface. When the thickness of brazed joint was reduced, elements of filler metals were completely diffused in the brazing process. Meanwhile, the micro holes disappeared, and a number of brittle phases was reduced to assure the performance of the brazed joint of W-Cu and 1Cr18Ni9 steel.

Acknowledgements

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