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2019

Wang, M., Chen, Y., Li, X., Hua, P., Gao, L., Zhou, W., & Wu, Y. (2019). Effect of Cu interlayer on the microstructure and strength for brazing of Tungsten/316L steel. *Journal of Materials Engineering and Performance*, 28(3), 1745-1752. doi:10.1007/s11665-019-3871-7

<https://hdl.handle.net/10356/144448>

<https://doi.org/10.1007/s11665-019-3871-7>

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Effect of Cu interlayer on the microstructure and strength for brazing of tungsten/316L steel
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Abstract: Brazing is known as an effective joining technique to bond tungsten and steel. However, the high residual stresses are produced for the different coefficients of thermal expansions (CTE) between W and steel. Compared with the direct brazing with BNi-2 foil filler, a Cu interlayer was selected as an interlayer to reduce the residual stresses between W and 316L steel. The brazing experiments were conducted at 1050°C for 25 min with different thickness of Cu foils. The investigation results show that successful brazing joints were achieved under different experimental conditions. The interface microstructure shows that intermetallic compound NiW formed at the W/BNi-2 interface, which has detrimental influence on the joint strength. The microhardness of different diffusion zones is higher than that of the substrates due to the formation of intermetallic compound and solid solution. All shear test specimens fractured at the W/BNi-2 interface close to W substrate, and the average strength of joints is 197 MPa, 275 MPa and 268 MPa with BNi-2 foil plus copper interlayer of 0.2 mm, 0.1 mm and 0.05 mm thickness, respectively; while the average strength of joints is 143 MPa with BNi-2 foil filler. The joint shear strength increases evidently with a Cu interlayer due to the excellent plasticity and toughness of copper.

Keywords: tungsten; steel; brazing; welding; shear strength

1.Introduction:

Divertor works in a complicated environment with high heat flux, high particle flux and heavy

neutron irradiation (Ref 1) , it is important to select appropriate plasma facing material (PFM) to enhance the lifetime of the component itself (Ref 2) . Due to its remarkable properties of low activation, high Z material, high thermal conductivity, high sputtering resistance and low deuterium/tritium retention (Ref 3,4) , tungsten is considered as one of the most promising divertor materials. However, tungsten is difficult to machine due to its brittle nature at room temperature, and usually tungsten (as first wall material) and steel (as structural material) need to be bonded for a divertor (Ref 5) . The bonding of dissimilar materials is a difficult issue due to their large differences in physical properties, such as different melting points (T_m : 3407°C for tungsten and 1300°C-1500°C for steels) and large mismatch of coefficients of thermal expansion (CTE: $4.5 \times 10^{-6} \text{ K}^{-1}$ for tungsten and $10-18 \times 10^{-6} \text{ K}^{-1}$ for steels), which will result in large residual stresses along the bonding interfaces. Based on the above reasons, traditional fusion welding is inapplicable to join tungsten and steels, brazing and solid-state diffusion bonding have been applied and reported in some research papers.

In solid-state diffusion bonding process (Ref 6-8) , the interlayer is widely used to reduce the residual stresses. Compared with the direct diffusion bonding (Ref 9) , the property of the joints is significantly improved by inserting interlayer, e.g. Zhong used Ti and Ni as the interlayer to bond W and steels (Ref 6,10) . For brazing (Ref 11-13) , although the joint was successfully obtained with the high temperature nickel-based filler for its excellent wettability and mobility, interlayer was not widely used and studied. In this work, a copper interlayer with low yield strength and low elastic modulus (Ref 14) was inserted between nickel based filler to reduce the residual stress in the joints. The interface microstructure, microhardness and mechanical property of brazing joints with Cu interlayer were analyzed.

2. Experimental procedures

The pure tungsten (99.95wt.%) and 316L steel (Fe-17Cr-13Ni-3Mo-2Mn, wt.%) used in this work were cut by wire-electrode cutting to a dimension of 10 mm^L×10 mm^W×5 mm^T respectively. Meanwhile, commercial copper interlayer (99.9 wt.% purity, 0.05 mm, 0.1 mm, 0.2 mm thickness) and a 0.03 mm thick BNi-2 filler (Ni-6Cr-5Si-3.5Fe-3.5B, wt.%, solidus temperature 971 °C, liquidus temperature 999 °C) were cut to a size of 10mm×10mm. Before the brazing process, the bonding surfaces of these specimens were ground with SiC emery paper and polished with diamond suspension. To remove the oil contamination, these samples were cleaned in an ultrasonic acetone bath for 15 min. The assembly of samples (shown in Fig. 1) was brazed in a molybdenum wire vacuum furnace. Since brazing temperature is generally 50 °C higher than the filler's liquidus temperature, the brazing was carried out at 1050 °C. The temperature was firstly raised to 800 °C at a rate of 10 °C/min and then heated up to the assigned brazing temperature at a rate of 10 °C/1.5min, and held it on the brazing temperature for 25 min under vacuum(< 10⁻³Pa), finally cooled with furnace to room temperature.

The cross-sections of brazing bonded joints were cut and prepared for metallographic examination. The microstructure observation and the fractured surfaces analysis was carried out under field-emission scanning electron microscope (SEM). The chemical composition and phase analysis of the joints was analyzed with energy dispersive X-ray spectrometry (EDS) and XRD. The mechanical property of bonded joints was evaluated by shear test (specimens size: 4 mm^L×3 mm^W×3 mm^T, presented in Fig. 2) on CMT5150 Universal tester with a constant displacement rate of 0.5 mm/min, and the average value of three specimens test results was taken. Microhardness was tested across the bonding interface by VTD401 microhardness tester with a holding time of

15 seconds and a load of 100 g.

3. Results and discussion

3.1 Microstructural investigation of joints

The brazing between W and 316L is successfully carried out. No cracks and unbounded regions are observed along the joint bonding interfaces. The micrographs and elemental intensity profiles of the different joints are presented in Fig. 3 and Fig. 4 respectively, which reveals that each joint is composed of several layers investigated by EDS line scanning.

3.1.1 The analysis of the 316L/W joint with BNi-2 plus Cu interlayer

Fig. 3(a) shows back-scattered electron image of 316L/Cu/W with 0.2 mm Cu interlayer. The high magnification detail of 316L/BNi-2/Cu interface and Cu/BNi-2/W interface are shown in Fig. 3(b) and Fig. 3(c) respectively.

As for the 316L/BNi-2/Cu interface, 316L/filler diffusion layer (D1), filler (D2), filler/Cu diffusion layers (D3) are produced. As for the Cu/BNi-2/W interface, it includes Cu/filler diffusion layer (D4), filler (D5), filler/W diffusion layer (D6). Apparently, these diffusion layers are produced during the brazing process due to the interdiffusion effect. The intergranular infiltration occurs in the 316L near the D1 region, which is the result of the elements diffusion from the filler to the 316L steel during brazing.

The point A is consisted of 62.17 at.% Ni, 5.68 at.% Cr, 3.56 at.% Fe, 1.54 at.% W, 11.07 at.% Si, 15.98 at.% Cu, and the element content of point C is similar to point A, which is confirmed to be Nickel based solid solution. The point B is consisted of 31.18 at.% Cr, 66.17 at.% B, 0.26 at.% Si, 0.53 at.% Cu, 0.38 at.% Fe, 1.48 at.% Ni. However, the boron element (B) may be not exactly detected by EDS due to boron is light element. It is also observed that the Cr profile shows steep fluctuation respectively in D2 zones (as shown in Fig. 4(a)), which may result from Cr-B intermetallic compounds (CrB, Cr₅B₃, Cr₂B) (Ref 15) . The point D is detected to contain approximately 11.4 at.% Cr, 33.9 at.% Ni and 54.7 at.% W, which is between NiW and NiW₂ intermetallic compound according to the W-Ni binary diagram, as shown in Fig. 5. The presence

of NiW and Ni(W) phase is confirmed by XRD, as shown in Fig. 6. Also, a small amount of Cr elements has been dissolved in NiW intermetallic compound.

Although the same fillers are used in this work, it is interesting to find that the thickness of the filler (D2) becomes thicker than that of filler (D5). It is suggested that the different thickness of fillers is attributed to the following reasons: 1) copper can improve the wettability of BNi-2 filler (Ref 16) . And there is a good compatibility between Fe and Ni, Cu and Ni, while Ni and W are not compatible well. 2) The melting point of tungsten (3040 °C) is much higher than the brazing temperature (1050 °C), so the tungsten atom is relatively difficult to be activated.

3.1.2 The analysis of the 316L/W joint with BNi-2

Fig. 7(a) shows back-scattered electron image of 316L/BNi-2/W. The high magnification detail of 316L/BNi-2/W interface and the elemental intensity profile are shown in Fig. 7(b) and Fig. 7(c) respectively.

As for the 316L/W bonded joint, there are three diffusion layers formed during brazing process: 316L/filler diffusion layer (A1), filler (A2), filler/W diffusion layer (A3). The point E is consisted of 70.22 at.% Ni, 5.93 at.% Cr, 4.94 at.% Fe, 8.47 at.% W, 10.43 at.% Si, which is confirmed to be Nickel based solid solution. The point F with 10.2 at.% Cr, 36.6 at.% Ni and 53.2 at.% W is also between NiW and NiW₂ intermetallic compound which is confirmed by XRD analysis, as shown in Fig. 8. The result is similar to the analysis of point A.

Two different forms of white compound are observed: a continuous layer close to W substrate and a number of discontinuous areas distributed on the filler (A2), which could be related to the process of intermetallic compound's growth. Because there are a large number of vacancies and dislocations on the W substrate surface, the nucleation of the intermetallic compound grow along the W surface firstly, when the intermetallic compound grows into a whole, the direction of

growth becomes horizontal.

According to the Ni-W phase diagram, three kinds of intermetallic compound (Ni_4W , NiW , NiW_2) are stable. However, NiW_2 and Ni_4W are not found in the present work. For the growth of intermetallic compounds, many investigations are carried out (Ref 18-21). The accepted view by some authors (Ref 1,18-21) is that the growth rate of compound is related to the interdiffusion coefficient in accord with kinetic analysis, which means that, if the interdiffusion coefficient for one intermetallic compound is much smaller than for other compounds, the intermetallic compound cannot grow up. The NiW is also observed at 1273 K for 100 h and at 1311 K for 112-600 h by Walsh and Donachie (Ref 22). These observations imply that the interdiffusion coefficient is much smaller for Ni_4W and NiW_2 than for NiW . According to reference (Ref 20), however, Ni_4W , NiW and NiW_2 are formed by annealing at $T=1073\text{-}1273\text{ K}$, $T=1073\text{-}1363\text{ K}$ and $T=1073\text{-}1313\text{ K}$, respectively. In summary, different results are reported by different researchers with different experimental parameters. Meanwhile, the mentioned-above experiments are all finished by solid diffusion bonding. For brazing, the growth of intermetallic compound is not investigated in depth (Ref 11-13). In the present work, the NiW may be formed due to the change of the interdiffusion coefficient. Compared with pure Ni foil used as an interlayer in solid diffusion bonding, BNi-2 filler in our work contains nickel element, boron, silicon, these elements may influence the interdiffusion coefficient. In order to understand the growth of intermetallic compounds, more work about interdiffusion coefficient should be done in the future.

3.2 Microhardness across the interfaces

To evaluate the microhardness of the bonded joints, microhardness is tested on the polished cross-section of the brazed joint for the 316L/Cu/W and 316L/W respectively. The substantial

variation indicates the changes in microhardness across profiles of the 316L/Cu/W and 316L/W, as displayed in Fig. 9 and Fig. 10 respectively.

At the D1 and A1 regions, the microhardness is improved due to formation of Ni-Fe solid solution. Despite the formation of Cu-Ni solid solution at D3 and D4 regions, the hardness of Cu-Ni solid solution is lower than of the Fe-Ni solid solution. The highest microhardness of the joint is 600 HV next to W for 316L/Cu/W joint, which clearly indicates the formation of intermetallic compound analyzed by EDS and XRD results. The hardness value of the 316L near the joint interface is higher than that of the initial 316L (< 200 HV) due to the penetration of BNi-2 filler into the grain boundary.

3.3 Shear strength and fracture surface analysis

The shear test specimens cut from the bonded joints are tested in CMT5150 Universal tester and five test results are obtained for each specimen. The results shown in Fig. 11 indicate that the strengths of joint with copper interlayer are higher than that of the joint without a copper interlayer, which is analyzed that soft copper interlayer can absorb some residual stresses along the joints during brazing(Ref 22). Zhang et al. (Ref 23) simulated the effect of intermediate layer with different thickness on the residual stress between Al_2O_3 and SS304 stainless steel. The results show that the residual stress of the joint decreases rapidly with the increase of the thickness of the intermediate layer at first and then the residual stress decreases slowly when the thickness of the intermediate layer reaches a certain extent, as shown in Fig. 12. Additionally the diffusion of Cu into Ni-based filler can improve the wettability and mobility of BNi-2 filler (Ref 16). Considering these changes of brazing filler, the amount of solid solution and the width of diffusion zone, the shear strength of joints with Cu interlayer are increased (Ref 13). From the point of welding

structure mechanics, when the copper interlayer thickness is appropriate, the amount of Cu-based solid solution and the rest of softer Cu are proper to improve the joint strength. When the copper interlayer thickness is too thick, the rest of softer Cu is left too much in the joint, which will decrease the joint strength; if the copper interlayer thickness is too thin, most of the area in the joint is fulfilled with Cu-based solid solution, which will limit the plastic deformation ability of joint. In our experiments, the shear strength of the joint with 0.2 mm copper interlayer is lower than that of 0.1 mm and 0.05 mm thickness, which show that the appropriate thickness of Cu interlayer should be around 0.1 mm. The study of Zhu Ying (Ref 24) and Kevin M (Ref 25) also indicate this rule that the shear strength increases first and then decreases with the increase of the thickness of the interlayer. As can be seen from Fig.13, the toughness of the joint is greatly improved by the addition of copper interlayer and 100 μ m thickness intermediate layer is the best.

The microstructures of the typical fracture surface of the 316L/Cu/W joint and 316L/W joint are shown in Fig.14 and Fig. 15 respectively. The fracture surface of the 316L/W joint is characterized by the appearance of the faceted grains (region G) and river patterns (region H). The average composition of region G is 99.92 at.% W, 0.08 at.% Ni, which suggests the region G is located on the substrate W adjacent to the brazing seam. While the region H is composed of 59.56 at.% W, 40.44 at.% Ni, the NiW can be expected in this region by XRD. For the fracture surface of 316L/Cu/W joint, in addition to faceted grains (region I) and river patterns (region J), dimples (region K) are visible on the fracture surface. The average element content of region I and region J is similar to the element content of region G and region H, respectively, which suggested region I is in W near the brazing seam, and the region K is NiW. The region K is comprised of 80.22 at.% Ni, 5.93 at.% Cr, 13.85 at.% W, the presence of Nickel based solid solution can be expected in this

region. On the base of what have been discussed above, the fracture mode of 316L/W is brittle fracture, while the fracture mode of 316L/Cu/W is a mixed mode of brittle fracture and dimple fracture.

Besides the effect of brittle metallic compound, the induced residual stresses play an important role in the joining of dissimilar materials, which strongly affects the strength of joints. During the cooling process, the contraction of 316L steel is larger than that of the W owing to different CTE, which causes residual stresses located adjacent to W (the low CTE material). In addition, the brittle nature of tungsten also may result in fracture at the W substrate.

4. Conclusion

The effect of copper interlayer on the microstructure and mechanical property of 316L/W brazed joints are investigated and the following conclusions are summarized.

1) The bonding between tungsten and 316L steel can be successfully achieved by brazing without microcracks and unbounded regions at the interfaces. No intermetallic compound is formed at 316L/BNi-2 interface and BNi-2/Cu interface due to the good compatibility between Fe and Ni, Ni and Cu, NiW analyzed by XRD is observed at W/BNi-2 interface.

2) The substantial changes of microhardness are attributed to the formation of solid solution and intermetallic compound. The maximum microhardness is tested at the interface next to W for W/Cu/316L joint and W/316L joint respectively.

3) The average shear strength of the brazed joints increased with Cu foil interlayer addition, and the shear strength value is also affected by the thickness of Cu foil interlayer; the appropriate thickness of Cu foil interlayer should be around 0.1 mm to reach the maximum joint shear strength in our experiments. All shear test specimens fractured in W near to the W/BNi-2 interface. The

fracture mode of 316L/W is brittle fracture, while the fracture mode of 316L/Cu/W is a mixed mode of brittle fracture and dimple fracture.

Acknowledgement

This work is supported by National Magnetic Confinement Fusion Program (Grant No.2014GB121001 and 2014GB121001B) and The Foundation of Laboratory of Nonferrous Metal material and Processing Engineering of Anhui Province (15CZS08031).

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Fig.1 Assembly drawing of the brazing specimens:(a)316L/W,(b)316L/Cu/W

Fig.2 Schematic diagram of shear strength tests

Fig.3 Back-scattered electron micographs and elemental intensity profiles of the joint with copper interlayer. (a)General view; (b) High magnification detail of the 316/Cu interface; (c) High magnification detail of the Cu/W interface

Fig.4 (a) elemental intensity profiles of 316/BNi-2/Cu; (b) Cu/BNi-2/W interface

Fig.5 Ni-W state diagram

Fig.6 XRD pattern of the fracture surface on W side for 316L/Cu/W

Fig.7 Back-scatter electron micographs and elemental intensity profiles of the joint without copper interlayer (a) General view; (b) High magnification detail of the 316L/W interface; (c) elemental intensity profiles of 316L/W

Fig.8 XRD pattern of the fracture surface on W side for 316L/W

Fig.9 Microhardness distribution 316L/Cu/W joint

Fig.10 Microhardness distribution 316L/W joint

Fig.11 Shear strengths of joint with copper interlayer

Fig.12 The residual stress along the y-axis near to the free side surface for different interlayer thickness

Fig.13 Stress-strain plot of the 316L/Cu/W joint

Fig.14 Fracture surfaces of the 316L/Cu/W joint

Fig.15 Fracture surfaces of the 316L/W joint