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Tungsten/steel diffusion bonding using Cu/W−Ni/Ni multi-interlayer

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Abstract: Diffusion bonding between tungsten and 0Cr13Al stainless steel using a Cu/90W−10Ni powder mixtures/Ni multi-interlayer was carried out in vacuum at 1150 °C with a pressure of 5 MPa for 60 min. The microstructures, composition distribution and fracture characteristics of the joint were studied by SEM and EDS. Joint properties were evaluated by shear experiments and thermal shock tests. The results showed that the joints comprised tungsten/Cu−Ni sub-layer/W−Ni composites sub-layer/Ni sub-layer/0Cr13Al stainless steel. The W−Ni composites sub-layer with a homogeneous and dense microstructure was formed by solid phase sintering of 90W−10Ni powder mixtures. Sound bonding between tungsten base material and W−Ni composites sub-layer was realized based on transient liquid phase (TLP) diffusion bonding mechanism. Joints fractured at bonding zone of W−Ni composites sub-layer and Ni sub-layer during shear testing, and the average strength was 256 MPa. Thermal shock tests showed that joints could withstood 60 thermal cycles quenching from 700 °C to room temperature.

Key words: tungsten; diffusion bonding; sintering; interlayer

1 Introduction

Tungsten is well suited for most kinds of high-temperature/high-vacuum applications, such as nuclear components [1−3], because of its superior high melting point, high density, high atomic number and low vapor pressure. However, the fabrication of tungsten components with complex shape and large size is difficult and cost. Developing the tungsten/steel composite structure to substitute full tungsten structure is of great significance. Several techniques including diffusion bonding have been investigated to join tungsten with steel [4,5]. The key issue of tungsten/steel diffusion bonding is the large mismatch of their coefficients of thermal expansion (4.4×10^{-6} °C^{-1} for tungsten and ~12×10^{-6} °C^{-1} for steel), which could result in a large residual stress at the bonding interface [6].

In the case of diffusion bonding of dissimilar materials, minimization of thermal residual stresses of joints can be achieved by the use of some foils acting as stress relieving elements. These foils, commonly called stress relief interlayers, are introduced between the parts to be joined. Recently, most efforts of the stress relief interlayer researches for tungsten/steel diffusion bonding were focused on single-interlayer including metallic single-interlayer (Nb, Ni, Ti, etc) [4,7,8] and multi-component single-interlayer (Fe−Ti, 316L−W, etc) [9,10]. Similar researches on other dissimilar materials diffusion bonding indicated that the residual stress status and bonding strength of the joints could be further optimized using multi-interlayer with a proper combination of different interlayer materials compared with most single-interlayer [11−13]. Ni with low yield strength is favorable for the residual stress relaxation [14], and W−Ni composites which could be designed to have a close CTE to tungsten are helpful for limiting the development of the thermal stress. In this work, a novel multi-interlayer of Cu/90W−10Ni powder mixtures/Ni for tungsten/steel diffusion bonding has been designed and investigated.

2 Experimental

The base materials used for the dissimilar joining were pure tungsten (99.8% purity) and 0Cr13Al ferritic

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stainless steel. The specimens for a joining were cylinder with dimensions of *d*18 mm×8 mm. The W (99.9% purity, 13 μm) and Ni (99.9% purity, 38 μm) powders were selected to prepare powder mixtures. The powder mixture with a chemical composition of 90W−10Ni (mass fraction, %) was milled in a high energy ball mill for 10 h, and then was pressed into interlayer with dimensions of *d*18 mm×0.25 mm by the cold isotropic pressing method under a pressure of 300 MPa. A 0.3 mm-thick Ni foil (99.5% purity) and a 0.008 mm-thick Cu foil (99.98% purity) were used as the other two interlayers. The surfaces of the materials were polished and ultrasonically cleaned in an acetone solution for 15 min. They were then assembled with a top-to-bottom stacking sequence of tungsten, Cu, W−Ni, Ni and 0Cr13Al in a graphite mould (Fig. 1). The diffusion bonding was carried out at 1150 °C for 60 min with a constant pressure of 5 MPa in a hot-pressing furnace under vacuum (<6×10^{-3} Pa).

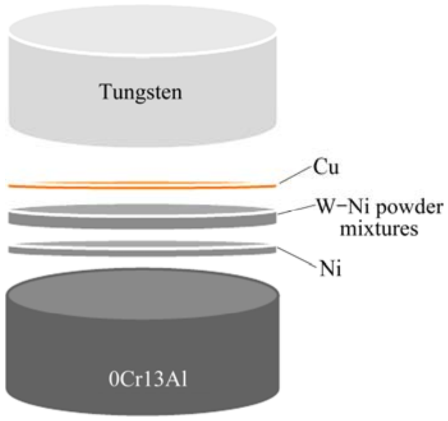

Fig. 1 Schematic diagram of specimen assembly

The microstructure and chemical composition of the joints were analyzed by a scanning electron microscope (SEM) equipped with an energy dispersive spectroscope (EDS). The mechanical property of the joints was tested at room temperature using samples with dimensions of *d*8 mm×16 mm using a shear test process at a shear speed of 1 mm/min. Fracture surfaces were examined using SEM. Thermal shock tests were also performed, during which the samples with dimensions of *d*8 mm×16 mm were heated in Ar atmosphere up to 700 °C and then quenched to room temperature in water. For comparison, three tungsten/steel TLP joints with Cu−5Ni interlayer, of which average shear strength was 213 MPa [5] and dimensions were *d*7 mm×~10 mm, also were performed the thermal shocking tests in the present work.

3 Results and discussion

3.1 Microstructure

From the micrographs in Fig. 2(a), it is seen that the tungsten/steel joint consists of 5 distinct regions, namely, the 0Cr13Al stainless steel base material, Ni sub-layer, W−Ni composites sub-layer, Cu−Ni sub-layer and the tungsten base material. W−Ni composites sub-layer was formed by solid state sintering of the initial W−Ni powder mixtures, and the content of W element reaches 14.57% in Ni phase (position F in Fig. 2(b)). The metallographic observation (Fig. 2(b)) indicates that W-particle is uniformly distributed in the Ni phase, forming a very dense microstructure. It is believed that W−Ni powder mixtures prepared by the high-energy balling milling played a key role in obtaining the W−Ni composites sub-layer with a homogeneous and dense microstructure (schematically shown in Fig. 2(d)). The bonding interface of W−Ni composites /Ni was irregular, most of which is Ni/W-particles bonding while few is Ni/Ni bonding. Figure 2(c) shows the bonding interface between the W−Ni composites and tungsten base material. The good integrity of the interface implied that the thin liquid Cu film formed during thermal bonding could adequately wet the surrounding W−Ni composites and tungsten.

Figure 3 shows the EDS composition analysis position of ~0.5 μm away from the tungsten base material (position A in Fig. 2(c)), the Ni element reaches a maximum level of 62% and the content of W element is 9%. This may be results from the strong affinity of the W atoms that were dissolved in this region to the Ni atoms, leading to the accumulation of Ni atoms in this area. At the middle position of the Cu−Ni sub-interlayer (position C in Fig. 2(c)), a minimum content of Ni element and a maximum level of Cu element were found, 30.27% and 69.73%, respectively, while no W element was detected. According to Cu−Ni binary alloy phase diagram [15], the melting point of Cu−Ni alloy with a composition of 30.27% Ni corresponding to the lowest level in the present Cu−Ni sub-layer is ~1200 °C. As the melting point of the Cu−Ni alloy is increased with the Ni content, the melting point of the whole Cu−Ni sub-layer should be higher than 1200 °C. This means that the Cu−Ni sub-layer was isothermal solidified at bonding temperature of 1150 °C. The bonding between tungsten and W−Ni composites sub-layer is achieved through a typical TLP diffusion bonding process, which could be described in detail as follows: 1) When Cu interlayer was melted during bonding, Ni atoms from W−Ni composites to diffuse into the thin liquid Cu film quickly and Cu−Ni liquid phase formed, and then the tungsten contacting with the Cu−Ni alloy liquid film gradually initiated the dissolution and diffusion; 2) With the continuous diffusion of Ni atoms, the melting point of the Cu−Ni liquid film progressively increased, resulting in an isothermal solidification of this alloy; 3) After solidification, Ni atoms continued to diffuse towards the

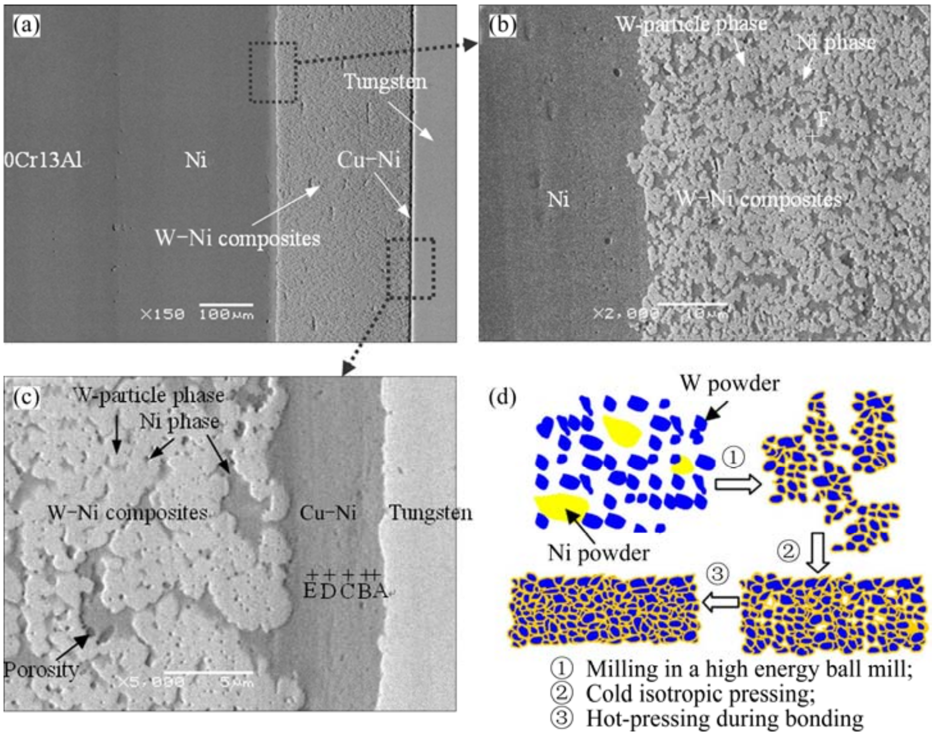

Fig. 2 SEM images of joint: (a) Joint structure (0Cr13Al/Ni/W−Ni composites/Cu−Ni/tungsten); (b) Interface of Ni/W−Ni composites; (c) Interfaces of W−Ni composites/Cu−Ni and Cu−Ni/tungsten; (d) Schematic drawing showing formation of W−Ni composites

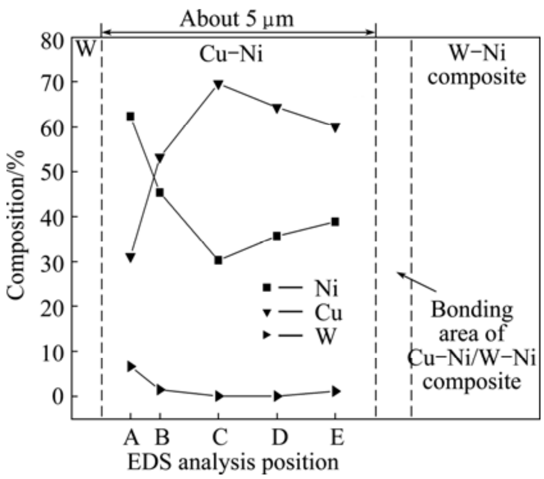

Fig. 3 EDS analysis results of Cu−Ni sub-layer

Cu−Ni interlayer through the solid-state diffusion at the high bonding temperature, eventually forming a TLP joint with high Ni contents and high temperature resistances.

3.2 Shear testing

The shear tests on four joint samples showed that the average shear strength of the joint reaches 256 MPa. All of the samples fracture at the bonding regions of Ni /W−Ni composites in tungsten/steel joints (Fig. 4(a)). Figure 4(b) clearly shows that the fracture surface exhibits many big dimples, indicating that the good bonding of Ni/W−Ni composites, which contributes to a higher strength of tungsten/steel joints. The fractographic observation on W−Ni composites (Fig. 4(c)) also shows a ductile fracture of Ni phase in W−Ni composites. The small Ni phase dimples were formed during the debonding between Ni phase and little W-particles phase.

Our previous work reported in Ref. [5] indicated that the maximum shear strength for the tungsten/steel joints bonded using the Cu−5Ni single sub-layer via TLP is 213 MPa, and the corresponding fracture of the joints mainly occurred in the tungsten matrix. These results implied that a large residual stress may exist in the region of the tungsten matrix close to the joint interface, leading to the premature fracture of the tungsten matrix at a shear stress level much lower than its intrinsic strength. By contrast, the shear strength of the tungsten/steel joints obtained in the current work improves greatly, and moreover, the fracture after the shear tests was not observed in the tungsten matrix. This comparison suggests that the Cu/W−Ni/Ni multi-interlayer used in the bonding readily decreases the residual stress level at the tungsten interface and improves the bonding strength of the joint.

3.3 Thermal shock tests

Four tungsten/steel joint samples with Ni/W−Ni

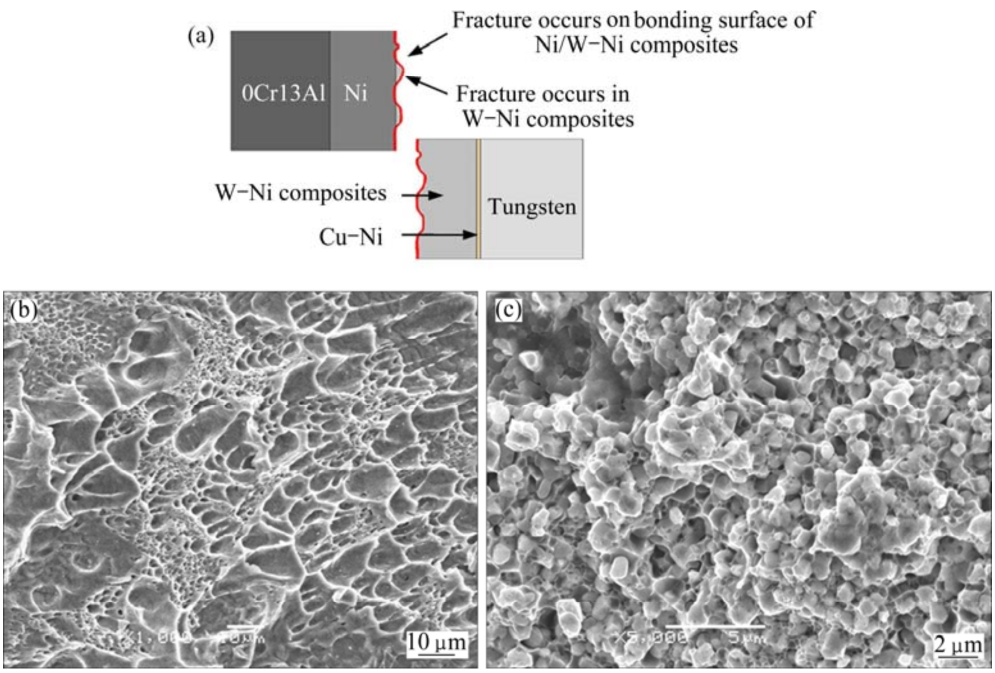

Fig. 4 Fractographs of joint (steel side): (a) Schematic diagram of fracture path; (b) Fracture surface of Ni/W−Ni composites joint surface; (c) Fracture surface of W−Ni composites

composites/Cu−Ni multi-interlayer and three tungsten/steel joint samples with Cu−5Ni single-interlayer were performed for thermal shock tests, and up to 60 test cycles were carried out for every sample. After each cycle, the samples were examined for possible cracks. Four tungsten/steel joints with Ni/W−Ni composites/Cu−Ni multi-interlayer passed the thermal tests without apparent cracks appearing during the test, which showed good thermal resistance. In contrast, the tungsten/steel joint with Cu−5Ni single-interlayer exhibited worse thermal shocking resistance, with the cracking emerged after 29, 22 and 35 cycles of thermal shock tests for 3 separate samples, respectively. The thermal tests result indicated that the use of Ni/W−Ni composites/Cu−Ni multi-interlayer could effectively relax the thermal stress of tungsten/steel joints.

According to Turner's model [16], the applied 90W:10Ni composites material possessed a linear CTE value of 5.3×10^{-6} °C^{-1}, which is close to that of the W base material (4.4×10^{-6} °C^{-1}). Besides, through the local plastic deformation of the relatively soft Cu−Ni layer between the W−Ni composites and the tungsten matrix, the thermal stress generated in the bonding and thermal shocking test processes could be effectively released. Additionally, as the Cu−Ni layer was very thin (~5 μm), the thermal stress arising from the mismatch of the thermal expansion coefficients between the Cu−Ni layer and the adjacent materials is limited. As a result, the thermal stress in the bonding region between the W−Ni composites sub-layer and the tungsten matrix could be minimized, resulting in the improvement of the bonding strength and the resistance to the thermal shocking. For the W−Ni composites/Ni interface, although there is large difference in the linear expansion coefficients of the two constitutions, the irregular bonding interface between them was favorable for the joint strength and residual stress relaxation of interface. Furthermore, due to the relatively low yield strength of Ni, the Ni layer in the interface area could be deformed slightly, and therefore releases the thermal stress at the interface of the W−Ni composites and Ni layers.

In summary, the thermal stress at the interface of the tungsten/steel joint maintained at a low level during the bonding and the thermal shocking processes, contributing to the improved bonding strength and the thermal shocking resistance.

4 Conclusions

1) A Cu/90W−10Ni powder mixtures/Ni multi-interlayer was successfully used as an interlayer in diffusion bonding tungsten to 0Cr13Al stainless steel.

2) The W−Ni composites sub-layer of joints were formed by solid phase sintering of 90W−10Ni powder mixtures. Bonding between tungsten base material and W−Ni composites sub-layer was realized based on TLP diffusion bonding mechanism.

3) In the shear tests, all the joint samples fractured in the bonding region of Ni sub-layer/W−Ni composites sub-layer in tungsten/steel joints under shear testing, and ductile fracture characteristics were observed obviously, and the average joint shear strength was 256 MPa.

4) Thermal shock tests of tungsten/steel joints indicated that the Cu−Ni/W−Ni composites/Ni multi-interlayer could effectively relieve the thermal stress of joints.

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Cu/W−Ni/Ni 多中间层的钨/钢扩散连接

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摘 要：采用铜箔/90W−10Ni(质量分数)混合粉末/镍箔多中间层，在加压 5 MPa、连接温度 1150 °C、保温 60 min 的工艺条件下，对纯钨(W)和 0Cr13Al 铁素体不锈钢进行真空扩散连接。利用 SEM、EDS、电子万能试验机及水淬热震实验等手段研究接头的微观组织、成分分布、断口特征、力学性能及抗热震性能。结果表明，连接接头由钨母材/Cu−Ni 合金层/W−Ni 复合材料层/镍层/钢母材五部分组成。接头中的 W−Ni 复合材料层由 90W−10Ni 混合粉末固相烧结而生成，其组织均匀、致密。W−Ni 复合材料层与钨母材以瞬间液相扩散连接机制来实现良好结合。接头剪切强度达到 256 MPa，断裂均发生在 W−Ni 复合材料层与镍层的结合区域，断口形貌呈现为韧性断裂。经过 60 次 700 °C 至室温的水淬热震测试，接头无裂纹出现。

关键词：钨；扩散连接；烧结；中间层

(Edited by Hua YANG)