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Diffusion brazing of tungsten and steel using Ti–Ni liquid phase forming interlayer

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A diffusion brazing process, for joining of tungsten (W) to steel using liquid phase forming interlayer, was developed for divertor application. Ti–Ni liquid forming interlayer was designed to overcome the large differences in physical properties between W and steel, and to eliminate the need for a high bonding pressure to achieve intimate contact between the bonded surfaces. The diffusion brazing experiment was conducted at 1050 °C for 1 h under 0.2 MPa uniaxial pressure of contact. Metallographic analysis revealed that a good bonding was obtained at both the W/Ni and Ni/steel interfaces, and the reaction products were identified in the diffusion zone. A hard reaction layer containing TiC with a high hardness value of about 20.3 GPa was found at the Ni/steel interface. Joint tensile strength at different testing temperatures was evaluated and all the fracture occurred in the brittle carbide layer.

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1. Introduction

The targeted development of helium cooled high performance divertors for fusion DEMO reactors requires the selection and joining of tungsten (W) or W alloys and ferritic–martensitic high chromium steels in present design concepts [1]. The method for realizing such kind of connection is restricted by a lot of problems related to the large differences in the physical properties of both materials. For instance, fusion welding technique is inapplicable for joining of them, due to the large differences of their melting points. Furthermore, W and steel have significant differences in their coefficients of thermal expansion (CTE), which causes high thermally induced residual stress in W–steel joints after welding and/or subsequent exposing to mechanical and thermal load.

Until now several joining techniques including brazing [2–4] and solid state diffusion bonding [5–9] have been developed for the joining of W and steel. Brazing has been successfully used for joining W to steel [10]. Although brazing alloys are metallurgically compatible with parent materials, the brazing temperature is usually much higher than the recrystallisation temperature of the materials (e.g., EUROFER97 [11]) to be joined. Thereafter, the upper working temperature of the assembly is also compromised by the presence of the lower-melting-point filler metal [12]. Diffusion bonding seems to be a suitable way to join W with steel due to its tolerable bonding temperature and the joint could be used at high temperatures. However, diffusion bonding tends to be limited as a production process because it is not tolerant of joints of variable width due to high loads have to be applied, moreover, its reliability is highly sensitive to surface cleanliness. To solve this problem, a liquid phase forming interlayer inserted between substrates is necessary to required to overcome the differences in their properties, and to eliminate the need for a high bonding pressure to achieve intimate contact between the bonded surfaces.

Previous studies have shown that nickel (Ni) can be used as the bonding interlayer for diffusion bonding of W to steel because its relative ductility can reduce the level of thermal stresses caused by the mismatch in CTE [6,7]. According to the Ti–Ni phase diagram (Fig. 1) [13], a low melting point liquid-phase can be formed above 942 °C between Ti and Ni. The aim of the present work is to propose a new way to join W to steel using Ti/Ni/Ti combination as interlayer through diffusion brazing. A liquid is formed at the bonding temperature resulting from the eutectic reaction between the core (Ni) and the outer (Ti) layer. When the Ti layer is very thin compared with the Ni layer in the joint, the liquid phase would occur

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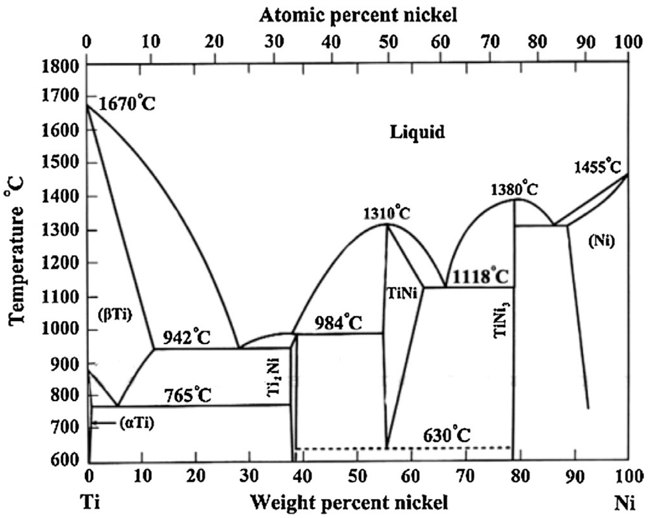

Fig. 1. Ti–Ni binary phase diagram.

at the interface at first, would decrease with time and finally disappear, since the liquid wets the surfaces of the base materials (W and steel) and interdiffuses with the core material (Ni) to transform into a more refractory solid material through re-solidification process.

2. Experimental procedure

The base materials used in this study are a high-Cr ferritic steel (Fe–17Cr–0.1 C in wt%, China TISCO) produced by mechanical alloying methods and commercially available W (99.95 wt% purity, Xiamen Tungsten) obtained by powder metallurgy and forging along the radial direction. The samples were cut into size of Ø25 mm × 13 mm. Diffusion brazing bonds were made between W and steel using Ti–Ni liquid forming interlayer. The commercial Ti foil (99.9 wt% purity, 20 μm thick) and Ni sheet (99.95 wt% purity, 400 μm thick) were chosen as eutectic formers. The surfaces of the materials to be joined were polished to a 1000 grit finish and ultrasonically cleaned in acetone. The bonding process was performed in a chamber under a vacuum pressure of 10^{-3} Pa. A sandwich-like bonding couple of W/Ti/Ni/Ti/steel (Fig. 2a)was mounted in the bonding chamber and a uniaxial pressure of about 0.2 MPa was applied to maintain the alignment and initial contact of sample during the thermal cycle. The bonding temperature of 1050 °C was selected based on the lowest eutectic reaction in order to achieve a bond with minimum microstructural changes in the base metals. A bonding time of 60 min was investigated with a fast heating rate of 20 °C/min. After the bonding process, the joint was cooled to 650 °C at a rate of 5 °C/min and held for 120 min and followed by furnace cooling to room temperature (RT) in vacuum.

After diffusion brazing, the bonded sample was sectioned into small specimens consisting of tensile test specimens and also specimens for microstructure and hardness investigations. The sectioning outline is presented in Fig. 2b, the microstructure and the chemical composition along the bond seams were analyzed using field-emission scanning electron microscope (SEM) and electron probe microanalysis (EPMA). The local hardness distribution across the bonding interface was examined by a nanohardness tester (VNHT) with a load of 5 mN. To determine the tensile properties of bonded specimens, transverse tensile test specimens were manufactured with a gauge length of 13 mm shown in Fig. 2c. The coupons were machined in such a way that the weld was positioned at the center of gauge length. Tensile tests were conducted by a tensile testing machine (Instron-3369) at a temperature range from RT to 650 °C with a strain rate of 0.001 s^{-1}. Fracture surfaces were observed in secondary electron mode of SEM combined with energy dispersive spectroscopy (EDS). The different phases on the fracture surface created during the bonding process were identified by means of X-ray diffraction (XRD) with Cu Kalpha radiation.

3. Results and discussion

The diffusion brazing of W and steel was successfully done using Ti/Ni/Ti liquid forming interlayer. The representative interfacial microstructure of the W/steel joint is given in Fig. 3. Both W/Ni and Ni/steel interfaces are free from discontinuities or pores. However, few transverse micro-cracks can be observed at the W/Ni

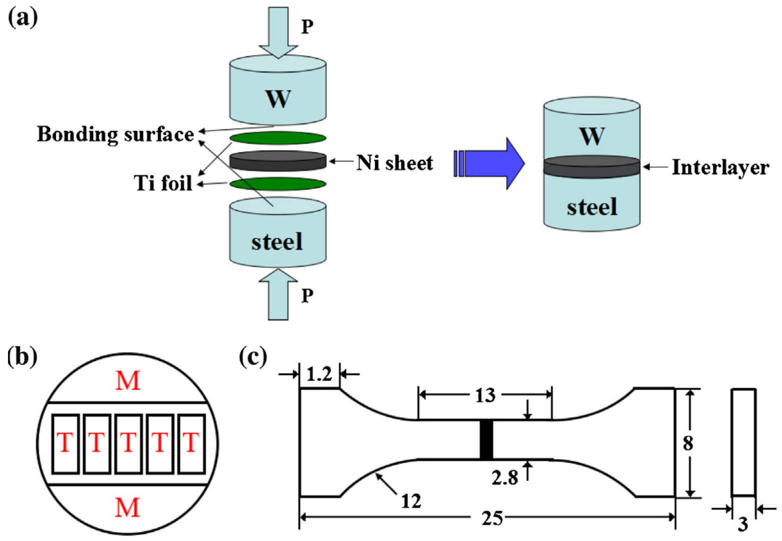

Fig. 2. (a) Schematic description of the sample preparation. (b) Sectioning outline on the top view of diffusion brazed sample, M: specimens for microstructure and hardness investigations, T: tensile test specimens, (c) schematic drawing for tensile testing (all dimensions are in mm).

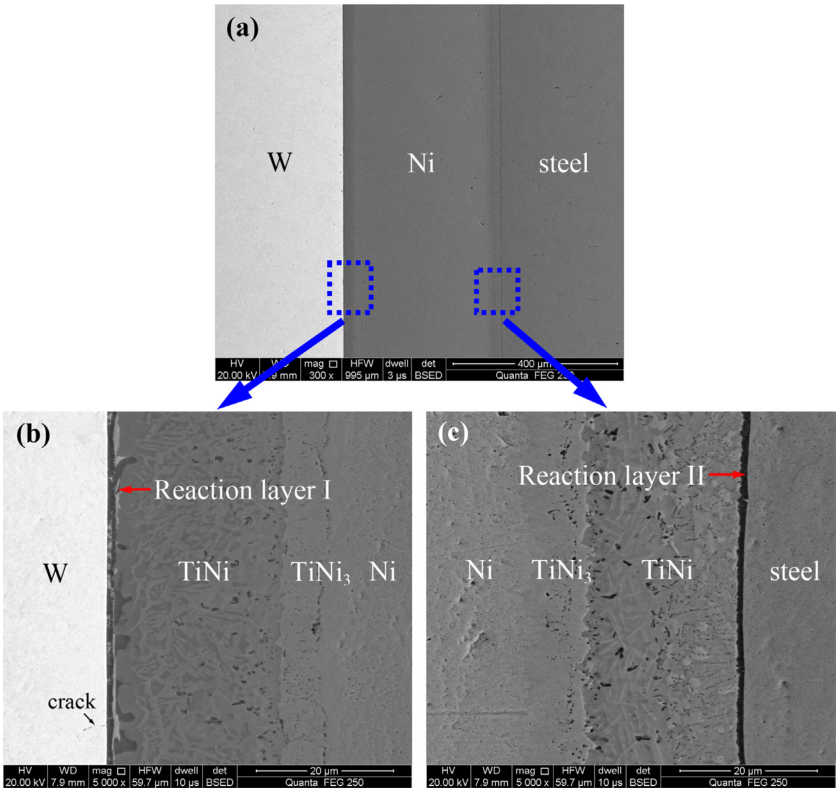

Fig. 3. (a) Representative interfacial microstructure of W/steel bonded joint and high magnification details of (b) W/Ni and (c) Ni/steel interfaces.

interface. This is believed to be due to the residual stress induced by the CTE mismatch and the different atomic sizes of W and Ti [8,14]. As shown in Fig. 3b and c, it can be seen that several reaction layers were observed at both interfaces. To investigate the elemental composition and its distance in the layers, elemental concentration profiles were measured across the bonded interfaces by means of EPMA. Fig. 4a shows such kind of mutual diffusion area at the interface of W/Ni with a thickness of ~40 μm. A similar area with a thickness of ~36 μm can be seen at the interface of Ni/steel in Fig. 4b.

At the W/Ni interface, a thin diffusion zone (Reaction layer I) has been found close to the W base metal and the composition changes gradually for W, Ti and Ni. Besides the formation of the reaction layer I, intermetallic TiNi and $TiNi_3$ layers were also formed during the diffusion brazing process. The formation of Ti–Ni intermetallic compounds is consistent with the Ti–Ni phase diagram, as shown in Fig. 1. These results demonstrated that the inserted active Ti foil had been molten and consumed during the formation of the reaction layer I and the intermetallic compound layers, leading to the presence of the interfacial structure of W/reaction layer I/TiNi/$TiNi_3$/Ni. As for the Ni–steel interface, similar to that observed for W/Ni interface, intermetallic TiNi and $TiNi_3$ layers were also noted. However, a significant change has been observed at the Ni–steel interface. A dark thin reaction layer (Reaction layer II) containing large amounts of carbon (C) atoms was appeared between TiNi and steel. According to the XRD investigation on the fractured surfaces of a tensile specimen both at the W side and steel side, the reaction layer II as shown in Fig. 5 consist of TiC, TiFe, $TiFe_2$, $TiCr_2$ and TiNi. The existence of the reaction products TiC, TiFe, $TiFe_2$ and $TiCr_2$ has been observed by the present authors for diffusion bonding of Ti or Ti alloys to steel [15–17].

The alteration of microstructures and the change of the element composition at the bond interfaces usually can influence the mechanical properties of materials around those regions significantly. Based on the microstructure investigations, the hardness of the substrates (W and steel), interlayer (Ni) and reaction zones at W/Ni and Ni/steel interfaces were measured by means of nanoindentation. The results are presented in Fig. 6. In the figure, the hard region was found in the reaction layer II, which was a mixture of TiC, TiFe, $TiFe_2$, $TiCr_2$ and TiNi. The maximum hardness value was about 20.3 GPa, which was much higher than the hardness of Ti–Ni compounds layers. This can be justified by the formation of the hard carbide in the reaction layer II. Likewise, the reaction layer I shows a hardness value (~7.4 GPa) higher than W (~6.3 GPa), which can be explained by the migration of Ti and Ni into W. This observed region-dependent hardness is ascribed to the intermetallic compound formation and solid solution strengthening effect which is related to reaction or interdiffusion process. Considering the whole bonded specimen, the reaction layer II that contains titanium carbide could be considered as the hardest region and therefore should be the weakest region against the tensile loading.

Tensile tests with miniaturized specimen of the bonded sample were carried out at room temperature and elevated temperatures. The experimental result is shown in Fig. 7. After testing, it was evident that all tensile test specimens were broken between Ni interlayer and steel. The fracture surfaces of the specimen tested at RT are shown in Fig. 8. The surface morphologies on the W side (Fig. 8a) and steel side (Fig. 8b) did not differ from each other. The fracture surfaces of the specimens tested at 500 °C and 650 °C were similar to that of the specimen tested at RT. The fracture surface is characterized by the appearance of the faceted grains, suggesting

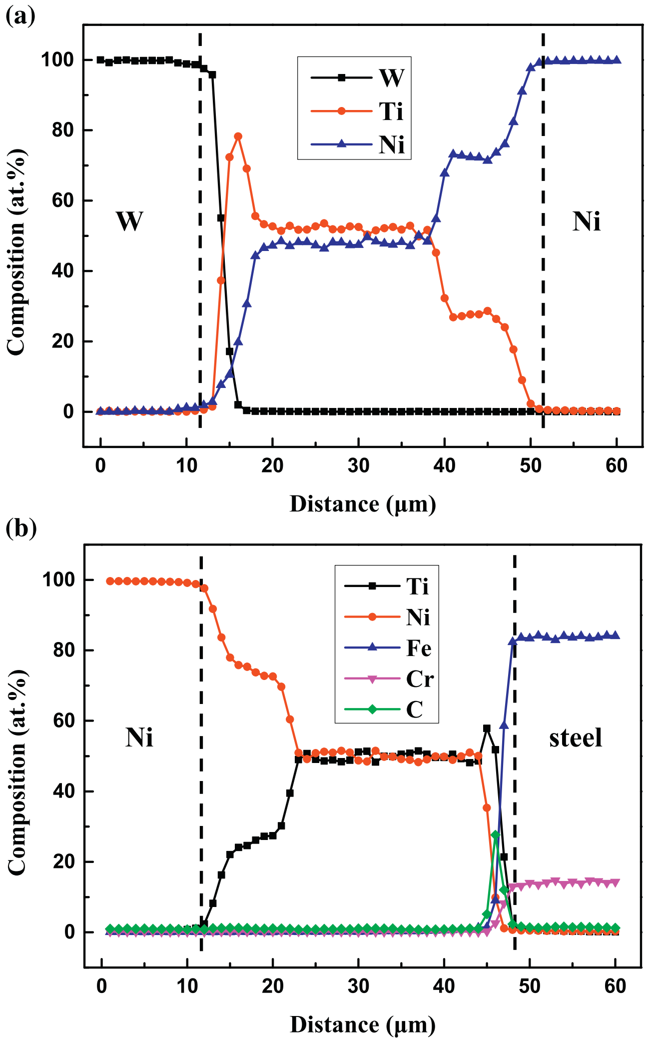

Fig. 4. EPMA analysis of element concentration across the: (a) W/Ni and (b) Ni/steel interfaces.

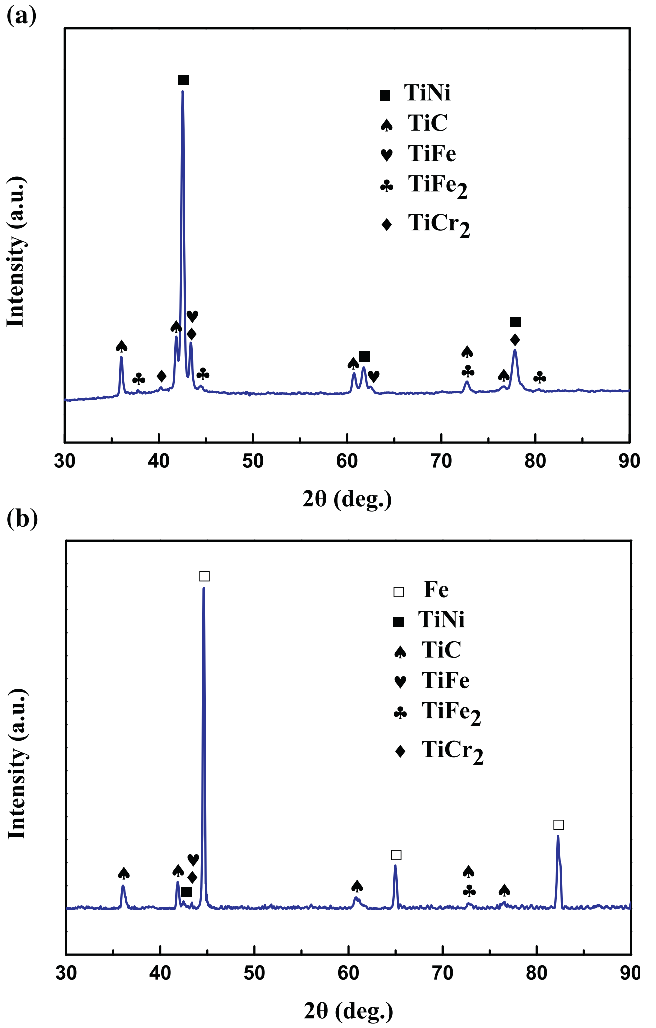

Fig. 5. XRD pattern of the fractured surfaces of (a) W side and (b) steel side for a tensile specimen tested at RT.

that the bonded specimen failed in a brittle mode. It is interesting to note that the fracture surface is divided into two regions, A and B. Fig. 8c and d are the higher magnification view of region A and B, respectively. The average composition of the tiny grain phases (region A) identified by EDS on W side is Ti (~40.8 at%), C (~16.3 at%), Fe (~32.6 at%) and Cr (~7.6 at%) with a small amount of Ni (bal.), suggesting the existence of Ti–Fe–Cr base intermetallic phases and TiC. While the region B is comprised of Ti (~50.2 at%), Ni (~27.5 at%), C (~21.3 at%) with very low quantities of Fe and Cr, TiC + TiNi phase mixture can be expected in this region. Furthermore, it was found that the fracture surfaces (magnified image of region B in Fig. 8d) were covered with grooves that possessed river structures on their slopes. This made clear that the bonded specimens were not fractured exactly along the bonded interface and the material in this fracture region was brittle. The XRD investigations result in Fig. 5 revealed that the tensile specimens were broken in the brittle reaction layer II that contains TiC.

The quality of the joint will need to be improved by further design of the liquid forming interlayer due to the formation of brittle intermetallic phases with metal carbides at the bonded interfaces. However, it is expected that the diffusion brazing have a potential to be used to produce a W/steel joint for divertor application, which has the advantage of lower bonding temperature,

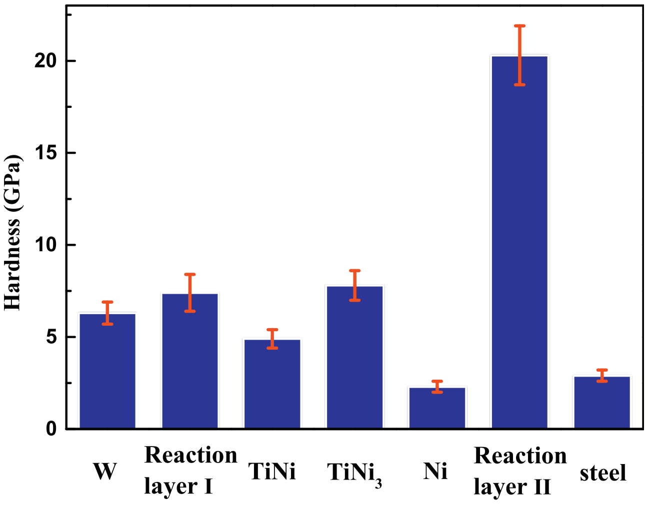

Fig. 6. Nanohardness of the substrates (W, steel), interlayer (Ni) and reaction zones at W/Ni and Ni/steel interfaces.

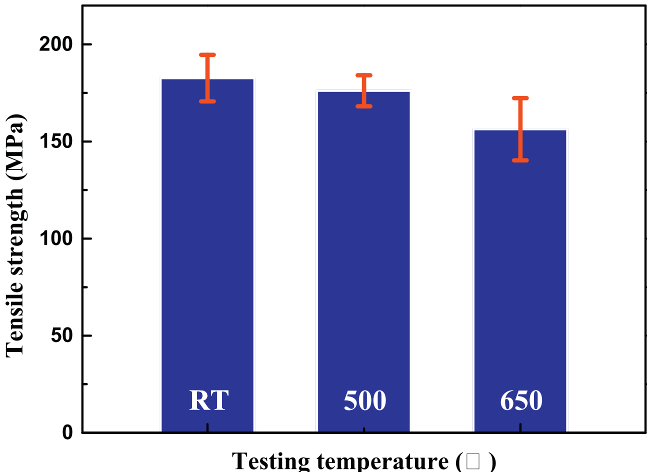

Fig. 7. Tensile strength of the W/steel joint as a function of testing temperature.

lower bonding pressure and less surface finish requirement than solid-state diffusion bonding.

4. Conclusions

Due to the technological interest for fusion applications, using Ti–Ni liquid phase forming interlayer for diffusion brazing of W to steel has been developed. The interfacial microstructure and mechanical properties of the bonded specimen were examined. The obtained main results are as follows:Liquid phase bonding of W to steel was successfully achieved by using Ti/Ni/Ti combination as interlayer at a relatively low pressure of about 0.2 MPa.The formation of Ti–Ni intermetallics and Ti diffusion caused the Ti foil to molten and consume during joining process.A hard reaction layer consisting of TiC, TiFe, $TiFe_2$, $TiCr_2$ and TiNi was identified at the Ni/steel interface. With a hardness of about 20.3 GPa, this region was the hardest part in the whole bonded sample and therefore can be considered as the weakest part especially against the tensile loading.The joint strength of more than 150 MPa was retained up to 650 °C. The surface fracture analyses on the tensile specimens tested at RT revealed a brittle fracture in the reaction layer that contains TiC.

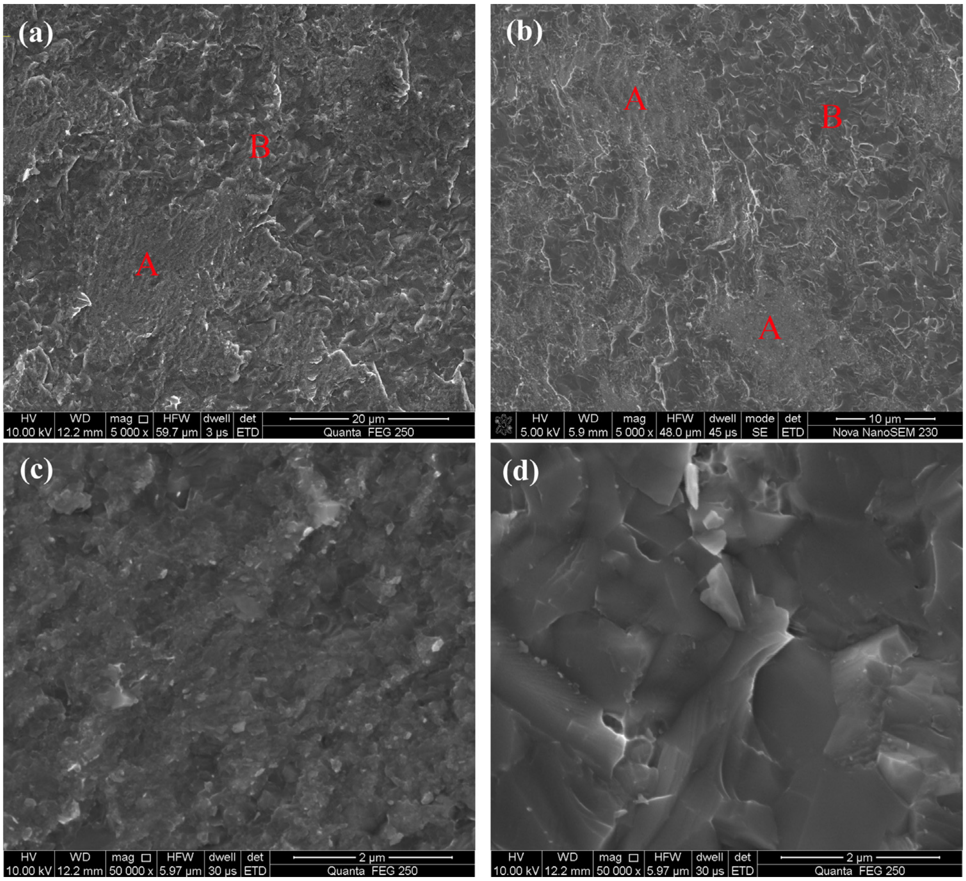

Fig. 8. SEM micrographs of the fracture surfaces on (a) W side and (b) steel side of diffusion brazed specimen, tensile tested at RT. The fracture surface on steel side is divided into two regions A and B, (c) and (d) are the higher magnification view of region A and B, respectively.

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