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Diffusion bonding of low carbon steel and pure zirconium with Cu-base amorphous interlayer

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ABSTRACT

A composite plate of low carbon steel and pure zirconium was fabricated by diffusion bonding with Cu-base amorphous interlayer at a temperature of 700 °C and pressure of 3 MPa. The effect of interlayer thickness on the microstructure and properties of the composite plate were investigated. The results show that all the samples produced with various interlayer thicknesses are composed of the reaction layer (RL) with a multilayer structure. Two dendritic structures are observed in the RL with 90 μm thick interlayer. The maximum shear strength across the composite plate and the displacement is 88 MPa and 0.33 mm, respectively. The maximum bending strength and strain of the composite plate reaches 1079 MPa and 47.6%, respectively.

1. Introduction

Diffusion bonding is a simple technique which is widely used to bond dissimilar materials. Bhanumurthy et al. (2001) claimed that this technique can fabricate composite plates with a minimum macroscopic deformation and metallurgical bonding through interdiffusion of chemical elements on both sides of the interface at proper temperature and pressure. However, Taouinet et al. (2013) found that thin and brittle intermetallic compounds formed in the reaction layer (RL) owing to the significant differences in physical and chemical properties of the two dissimilar materials and deteriorated mechanical properties of the composite plate.

Terai et al. (1997) revealed that zirconium base alloys, which possess high-temperature corrosion resistance, creep resistance and low neutron absorption cross-section, are extensively used in the fields of chemical reaction vessels and nuclear reactors. Steels are traditional structural materials with reasonable mechanical properties and acceptable cost than zirconium base alloys. Diffusion bonding of composite laminates between zirconium alloys and steels have been investigated by many researchers. Ahmad et al. (2008) discovered that primary elements (Fe, Cr and Ni) in steels would react with zirconium to form brittle and hard intermetallic compounds, and cracking occurred during bonding Inconel 625 in zircaloy-4 at 1050 °C and 1100 °C under 4.9 MPa. Lee et al. (2009) carried out the diffusion bonding of zircaloy-4 and stainless steel (304 L) without interlayer at 850 °C–1020 °C under 11 MPa, and massive brittle intermetallic phases $Zr(Fe, Cr)_2$, and $Zr_2(Fe_{1-x}Ni_x)$ with $0.15 \leq X \leq 0.25$ were observed in the diffusion zone. Furthermore, high residual stress develops at bonding face due to the large difference in coefficients of thermal expansion between dissimilar materials, and the result leads to the formation of microcracks in the RL.

Hence, utilizing interlayer to bond steel and zirconium is a simple and feasible alternative. Early works (Shaaban et al., 1978; Wayman et al., 1986) used pure Fe and Pt as an interlayer to prevent the reaction of Cr, Ni with Zr, respectively. However, a few intermetallics including FeZr and Pt_3Zr were also still detected in the RL. Ahmad et al. (2003) reported that pure Ta foil as interlayer bonded steel and zirconium to study microstructure evolution of the RL. There were various intermetallic phases with different microhardness in the RL and voids or cracks could not always be eliminated at high temperature. Atabaki (2010) and Atabaki et al. (2012) investigated partial transient liquid phase diffusion bonding of Zircaloy-4 (or Zr-2.5Nb) and SS321 with Ti based interlayer to control the formation of intermetallics in the RL. The results showed that the solid state growth rate of the intermetallic compounds was linear with the square root of time at a given temperature and exponential with temperature. According to Chen et al. (2014), the RL of the composite plate of Zr-Sn-Nb alloy and 304SS increased from 55 μm to 135 μm by using pure Ni interlayer, and the wide RL was beneficial to the relaxation of residual stresses, but the bond strength was not greatly improved owing to the formation of amounts of brittle $Zr(Cr, Fe)_2$ and $Zr_2(Ni, Fe)$. Nicholas (1998) concluded that there is no constant interlayer thickness which can be defined as

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necessary for effective diffusion bonding and strong composite plates, however, the thickness of the RL can directly determine the bonding strength. Most researchers prefer to obtain a suitable thickness of the RL by control temperature and time. Such as Aboudi et al. (2017) attempted to join Zircaloy-4 and SS304 L using 50 μm thick copper interlayer at a holding time of 45 min under 29 MPa, but huge cracks were formed in the RL with more than 200 μm thickness at 1050 °C. Elrefaey and Tillmann (2009) suggested that 100 μm thick Cu-based alloy (Cu-Mn-Ni) was a good interlayer material to bond titanium and steel at an optimal temperature of 850 °C for 90 min. The maximum shear strength of 105.2 MPa was obtained. Dong et al. (2014) also studied the microstructure evolution and the bonding strength of TC4 titanium alloy/304 stainless steel composite plates with Cu-base amorphous foil (Cu-Ti-Ni-Zr-V) by diffusion bonding. The results showed that Fe-Ti, Cu-Fe-Ti intermetallic compounds formed near 304 stainless steel in the RL, and the maximum shear strength of the composite plate was 116 MPa. Even though the various alloys as the interlayer for bonding steels and zirconium have been investigated, different thicknesses Cu-base amorphous interlayer (Cu-Ni-P-Sn) were never reported to join steels and zirconium.

This work focuses on an approach to join low carbon steel and pure zirconium by diffusion bonding at low temperature and low pressure with Cu-base amorphous alloys foil as interlayer. The effects of interlayer thickness, temperature and hot-pressure-stress on the microstructure evolution and mechanical properties of the composite plate of low carbon steel and pure zirconium are investigated.

2. Experimental procedures

Wrought low carbon steel was selected as base plate and pure zirconium was used as cladding plate. Cu-base amorphous alloy foil (30 um thick) was selected as interlayer. The chemical compositions of low carbon steel, pure zirconium and Cu-base amorphous alloy foil are presented in Table 1. The low carbon steel and pure zirconium plates were prepared with dimensions of $100 \times 100 \times 6\,\text{mm}^3$ and $100 \times 100 \times 1.5\,\text{mm}^3$, respectively. All plate surface was kept clean before the diffusion bonding. All the bonding surfaces were ground by SiC paper and then ultrasonically cleaned by acetone for 2 min. In this study, one layer (30 μm), two layers (60 μm) and three layers (90 μm) of Cu-base alloy foil were applied, separately, to control the thickness of interlayer. Hereafter, the corresponding composite plates with the three thicknesses of interlayer are labeled as SZ30, SZ60 and SZ90, respectively. Fig. 1 shows a schematic illustration of the diffusion bonding process. The assembly was set into a vertical cylindrical furnace. An axial pressure of 1 MPa was applied to the assembled sample to make the interlayer closely contact with the base metal. The diffusion bonding was carried out at 700 °C and uniaxial load of 3 MPa for 1 h. The samples were heated under a rate of 10 °C/min and cooled in the furnace. All the above procedures were performed under a vacuum less than 1×10^{-3} MPa.

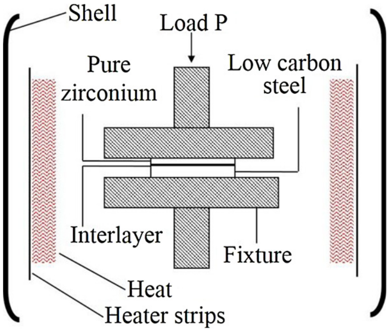

Fig. 1. A schematic illustration of diffusion bonding of low carbon steel and pure zirconium.

The diffusion bonded joints were sectioned along a plane normal to pure zirconium/low carbon steel interface and polished to a surface finish of 1 μm. The joint microstructures were investigated using a FIE-Quanta 200FEG scanning electron microscope (SEM) in back scattered mode (BSE) with an acceleration voltage of 20 kV. The phase compositions of the RL zones were determined by energy dispersive spectrometer (EDS, FIE, Quanta 200FEG) and X-ray diffraction (XRD, Panalytical, Empyrean). Line scanning of EDS analysis was performed from the steel side to the zirconium side to obtain the composition distribution crossing the RLs. The step of line scanning is 0.25 μm for SZ30, SZ60 and 0.45 μm for SZ90. The phase constitution was also identified using XRD on the shear-fractured surface (Zirconium side). The size of X-ray beam is 2.5 mm × 1 mm. A step of 0.05° was applied to scan from 20–90°.

The microhardness of the RL were measured using an HVS-1000Z tester with 10 g load with a testing of 25 s on various zones. Shear test and bend test were performed with a San-Si tester in the compression direction to investigate the properties of the samples. Fig. 2 shows the shear test specimen and a schematic illustration of the shear test apparatus. A compression speed of 1 MPa/s was used. According to Yan et al. (2010), the shear stress (τ) was calculated from the following relations:

$$\tau = \frac{P}{ae} \tag{1}$$

Where a and e are the width and height of test interface, respectively. In this study, a and e are 25 mm and 3 mm. After the shear tests, the fracture morphologies and cross-sectional microstructures of fractured samples were observed in order to analyze the failure mode.

In order to investigate the influence of various thickness interlayer on the mechanical properties of the diffusion bonded low carbon steel/pure zirconium composite plate were measured by three-point bend test. Specimens were cut into 90 mm × 20 mm × 7.5 mm (the thickness

Table 1
Nominal chemical composition of experimental materials (wt. %).

Alloy type	Elements						
Pure zirconium	Zr + Hf	Hf	Fe + Cr	H	N	O	C
	≥99.2	4.5	0.2	0.005	0.025	0.16	0.05

Alloy type	Elements										
Low-carbon steel	Fe	Nb	V	Ti	Cr	Ni	Mo	C	Mn	Si	P	S
	Bal.	≤0.07	≤0.15	≤0.2	≤0.30	≤0.012	≤0.1	≤0.2	≤1.7	≤0.5	≤0.04	≤0.035

Alloy type	Elements			
Cu-base amorphous alloy foil	Cu	Ni	P	Sn
			7	
	Bal.	5.4	7	10