MICROSTRUCTURE EVOLUTION DURING BONDING OF Al-Fe-Ce ALLOY WITH ZIRCALOY-4 USING Cu AND Zn INTERLAYERS

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Abstract. Bonding of Al-Fe-Ce alloy with Zircaloy-4 was carried out using Cu and Zn interlayers. Microstructural characterization was done using scanning electron microscopy and chemical composition of the phases was determined by energy dispersive X-ray spectroscopy. Formation of layered structure was observed in the diffusion zone of bonded structure of Al-Fe-Ce with Zircaloy-4 using Cu as an interlayer. Microhardness of different layers was determined. The results indicate that hardness of the AlZr phase is higher than the Al_2Zr_3. Microhardness of the bonded alloys also increased due to the small addition of Cu in the lattice. Microhardness of the bonded alloy did not change when the bonding of Al-Fe-Ce with Zircaloy-4 was carried out using Zn coating as an interlayer and a few microcracks were observed in the diffusion zone.

1. INTRODUCTION

The fabrication of components consisting of dissimilar materials by conventional welding is always difficult because their thermal properties like expansion coefficient, melting temperature, *etc.* differ significantly. Due to this difference, it is very difficult to have a controlled melting on both sides of fusion weld. The main problem in the conventional welding is the formation of intermetallics at the interface as well as in the heat affected zones (HAZ). It will not only change the microstructure but also the chemistry of the parent materials, which results in the degradation of the joints. In conventional welding size of the fusion zones (FZ) and HAZ is large. Due to the large difference in their thermal properties, joining materials suffer large thermal stresses in the FZ and HAZ.

Electron beam welding (EBW) and laser beam welding (LBW) are being used to join dissimilar materials because of the small FZ and HAZ as well

as small distortion, compared to the conventional welding techniques. Ahmad *et al*. [1,2] applied the EBW to weld the dissimilar materials such as stainless steel and Al-Fe-Ce alloy with Zircaloy-4, and characterized the formation of different phases in the FZ. They also observed cracks in the FZ of welded specimens of Al-Fe-Ce with Zircaloy-4. Cracks were attributed to the formation of brittle intermetallic layer ($AlZr_2$) at the joint. These cracks are deleterious and can be the cause of failure of materials under stress during service.

Diffusion bonding (DB) is being used to join materials which are very difficult to join using conventional welding techniques [3-12]. In DB a strong bond can be achieved at the interface without melting the parent materials. DB has been found to result in good metallic couples. For example, good interconnections in Ni/Al/Ni have been formed through diffusion soldering [13] and a low temperature fabrication bond has been developed in diffusion soldering of Cu/In/Cu interconnections [14].

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Table 1. Composition of Zircaloy-4 and Al-Fe-Ce alloy (wt.%).

Alloys	Elements							
	Al	C	Ce	Cr	Fe	Ni	Sn	Zr
Zircaloy-4	-	0.002	-	0.10	0.15	<0.005	1.52	Balance
Al-Fe-Ce	92.71	-	2.53	-	4.76	-	-	-

Transition liquid phase (TLP) bonding is a process through which strong bonds can be made without melting of the base materials. In TLP bonding, temperature gradient is very low as compared to fusion welding and hence microstructural changes within the joint can be prevented [3]. Akhter *et al.* [15] and Ahmad *et al.* [16] succeeded in bonding dissimilar materials like stainless steel and Zircaloy-4 using Ti and Ta as interlayers. Aluminum forms a eutectic with Cu and Zn at 548 °C and 382 °C respectively [17], and TLP bonding above this temperature can be helpful in bonding the Al-Fe-Ce to Zircaloy-4. In this study, DB has been used to join Al-Fe-Ce alloy with Zircaloy-4 using Cu and Zn interlayers.

2. EXPERIMENTAL PROCEDURE

The composition of the Zircaloy-4 and Al-Fe-Ce alloy is given in Table 1. For diffusion bonding trial samples were cut to the dimensions of 10 x 10 x 4 mm^3. The joining surfaces were prepared to a 1000 grit finish using SiC abrasive papers and then ultrasonically cleaned and stored in ethanol until used for bonding.

Two types of interlayer were used as filler metals in order to compare the bonding behavior. A pure Cu metal foil of thickness 22 μm was placed between the surfaces being joined and a bonding temperature of 550 °C was used (above the Al/Cu eutectic temperature of 548 °C). The samples were bonded in a vacuum of 5.33 x 10^{-7} bar using a slight pressure to hold the bonding surfaces together. The bonding time was varied from 1 to 15 minutes. The other interlayer was a Zn coating of 1 μm thickness sputter deposited onto the Zircaloy-4 and Al-Fe-Ce alloy surfaces. The diffusion bonding procedure was the same as that used for the Cu interlayer except the bonding temperature. A temperature of 385 °C was used which was above the eutectic temperature of the Al-Zn system (380 °C). Furthermore, solid-state diffusion bonding was also made using the Zn coating and a bonding temperature of 370 °C was used with an applied load of 0.7 MPa and bonding time varied from two seconds to five minutes.

In order to assess the quality of the diffusion bonds, metallurgical samples were prepared by cutting transverse sections through the joint region and the surfaces were polished to a 0.25 μm finish for metallographic examination. The microstructure of the alloys was revealed by etching with 50 ml H$_2$O$_2$, 47 ml HNO$_3$, and 3 ml HF. The bonded interface was examined under the scanning electron microscope (SEM), LEO 440i. Changes in the composition of the bonded interface were analyzed using energy dispersive X-ray spectroscopy (EDX). A Leitz mini-load micro-hardness tester with a load of 100 g was used to obtain hardness profiles and homogeneity across the joint region in case of Cu interface. A lower load of 25 g was used at the bonded interface in case of Zn coated specimens. Microhardness measurements were done in the center of the each layer along the length of specimens and average of 5-6 measurements were taken for interpretation.

3. RESULTS AND DISCUSSION

3.1. Microstructure

SEM secondary electron image (SEI) at low magnification showing the microstructure features of the diffusion bonded sample using Cu as an interlayer is shown in Fig. 1a. It shows four different layers. The layers are designated as A, B, C, and D. Layer A is actually Zircaloy-4 containing small amount of interdiffusing elements like Al, Ce, and Cu. The other layers are formed in Al-Fe-Ce alloy due to the diffusion of Zr from the Zircaloy-4 towards the Al-Fe-Ce alloy. Backscattered electron image (BSI) at higher magnification also confirm the formation of different layers with smooth interfaces clearly shown in Fig. 1b. The width of the layer B, C, and D are 100, 430, and 175 μm respectively. It indicates that a higher gradient exists for the formation of layer C as compared to the layers B and D. The interfaces among the layers and between the Zircaloy-4 and Al-Fe-Ce alloy are smooth and almost defects free. Few voids can be seen in Zircaloy-4. It shows the transfer of Zr- atoms towards Al-Fe-Ce alloy. EDX analysis re-

Table 2. Quantitative analysis of layers A, B, C, D in the diffusion bonded sample of Al-Fe-Ce and Zircaloy-4 using Cu interlayers.

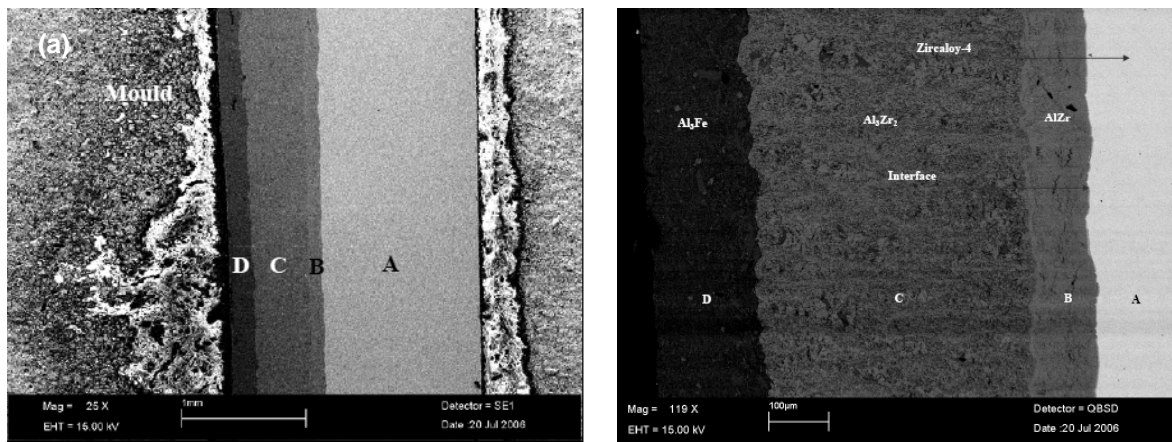| Sample layers designation	Elements (at. %)							Phases
	Al	Fe	Cr	Zr	Sn	Cu	Ce	
A	0.03	0.16	0.06	97.40	1.64	0.02	0.04	Zircaloy-4
B	54.64	0.17	0.02	44.74	0.31	0.30	0.25	ZrAl
C	61.17	0.37	0.01	38.32	0.15	0.38	0.60	Zr_2Al_3
D	76.65	21.10	0.02	0.17	0.16	1.82	1.56	Al_3Fe

Fig. 1. (a): SEM micrograph, SEI at low magnification, showing diffusion layers formed during TLP bonding using Cu as an interlayer, (b): SEM micrograph, BSI, at high magnification showing diffusion layers formed during TLP bonding using Cu as an interlayer.
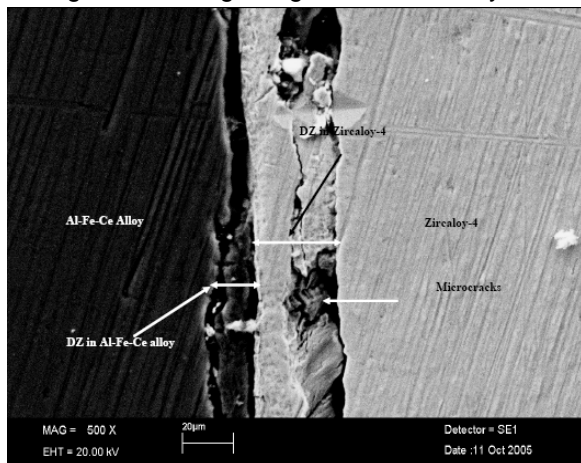

Fig. 2. SEM micrograph showing a solid-state bond made using a Zn coating.

sults of the layers, giving the concentration of the diffusing elements from the Al-Fe-Ce alloy towards Zircaloy-4 and vice versa, are summarized in Table 2. It clearly indicates that very small amount of Al diffuses towards the Zircaloy-4, i.e. layer A. Layer B has a very small width compared to layers C and D, which is rich in Al and Zr and their ratios corre-

spond to the AlZr phase. The composition of layers C and D correspond to Al_2Zr_3 and Al_3Fe phases respectively. The concentration of the Fe and Al varies significantly from the diffusion zone (DZ) to the end point of the Al-Fe-Ce alloy. The concentration of Fe is higher in layer D which indicates that diffusion of Al occurs very fast towards DZ. While diffusion of Fe does not take place towards DZ, rather it seems that it diffuses from DZ to layer D. It is important to note that Cu interlayer completely disappears and diffusion of Cu is higher towards the Al-Fe-Ce alloy. Interestingly the concentration of Cu increases from layer A to layer D having a maximum value of 1.82 at.%. The results suggest that Cu diffuses towards the Al-Fe-Ce alloy indicating its higher diffusion coefficient in Al- alloys as compared to Zircaloy-4.

TLP diffusion bonds made using Zn coating broke during cutting of the bonded samples for metallurgical analysis. This suggested that the joints were very weak. In contrast the solid-state diffusion bonded samples were cut successfully and prepared for
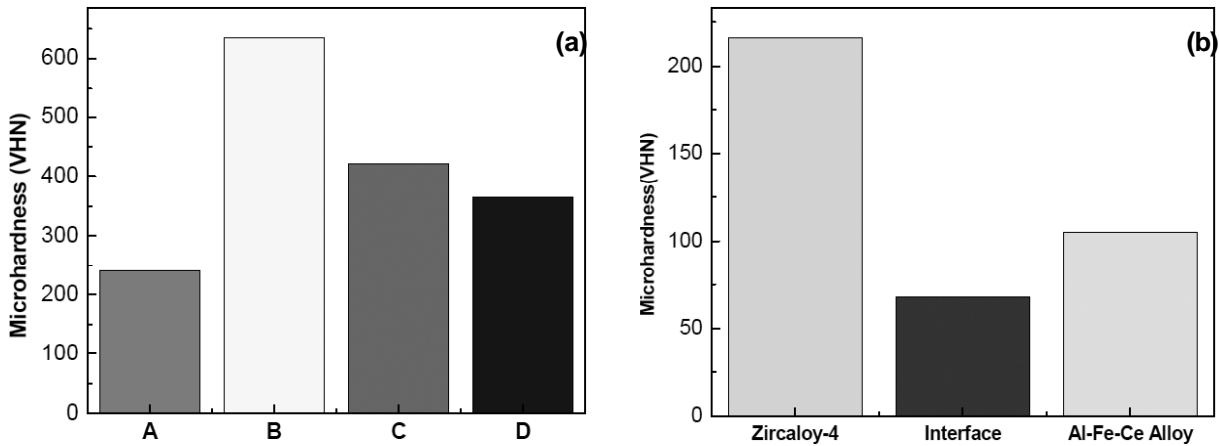
Fig. 3. (a): Changes in microhardness across the layers formed in DZ when bonding Al-Fe-Ce alloy to Zircaloy-4 using Cu as an interlayer, (b): Changes in microhardness across the solid-state bonded interface when Zn using as a coating interlayer.

microstructural characterization. Fig. 2 shows the bond interface in case of solid-state bonding of Al-Fe-Ce to Zircaloy-4 using Zn coating interlayer. It is clear that the Al interface has moved towards Zircaloy-4 and has defects like voids and microcracks clearly marked in Fig. 2. The interface movement of Al in Zircaloy-4 is larger than Zr interface in Al-Fe-Ce alloy. This indicates that the diffusion coefficient of Zr is lower in Al-Fe-Ce as compared to Al in Zircaloy-4. The concentration of Al is very low in the DZ marked in Zircaloy-4 whereas 50 at.% Zr and 50 at.% Al is found in the start of diffusion region formed in Al-Fe Ce alloy marked by arrows in Fig. 2. The concentration of Zr is decreasing towards the Al-Fe-Ce alloy. Phases of Zr with Al were not observed in the DZ formed in Zircaloy-4. The reason for the absence of these phases may be the formation of AlZr layer which acts as barrier for further diffusion of Al towards the Zircaloy-4. The Al diffused in the Zircaloy-4 before the formation of barrier layer spreads in larger area in the atomic form.

3.2. Microhardness

Microhardness of the designated layers A, B, C, and D is shown in Fig. 3a in the form of histogram for samples bonded using Cu as interlayer. It clearly indicates that the hardness of the layer A nearly corresponds to the value of the Zircaloy-4. However hardness of the layer B is higher than layers C and D. This dictates that the layer B is harder than all the layers formed in the DZ. The phases given in Table 1 are responsible for hardness in different layers. The layer B is composed of AlZr phase whereas

layer C has the composition of Al_2Zr_3 and layer D has a composition different to that of the Al-Fe-Ce alloy and a phase composed of Al_3Fe observed instead of original alloy.

Microhardness of the Al- Fe-Ce alloy and Zircaloy-4 as well as the interface in case of Zn coating on both surfaces of Zircaloy-4 and Al-Fe-Ce alloy is shown in Fig. 3b. The results indicate that hardness of both the alloys did not change after solid-state bonding, whereas the microhardness value of the interface was even lower than that of either the Al-Fe-Ce alloy or Zircaloy-4. Furthermore, when a solid-state bonding is used with a Zn coating, hardness lower than that of the parent alloys is observed at the interface.

These results suggest that TLP bonding using a pure Cu interlayer produces a good quality bond with an interlayer free of defects and pores. However, a DZ is formed throughout the bonded region and a significant difference in hardness values across the region is observed.

4. CONCLUSIONS

Bonding of Al-Fe-Ce and Zircaloy-4 has been carried out successfully using Cu and Zn interlayers. The Cu interlayer formed a eutectic with aluminum and a metallurgical joint was achieved using TLP bonding. A layered structure occurred in the Al-Fe-Ce alloy and each layer corresponds to the formation of metallic compounds. The solid-state bonding of the Al-Fe-Ce alloy with Zircaloy-4 was produced through solid-state diffusion by Zn coating on both the interfaces of Zircaloy-4 and Al-Fe-Ce alloy. The DZ formed in Zircaloy-4 is larger com-

pared with the DZ occurred in Al-Fe-Ce alloy and microcracks were observed in the DZ formed in Zircaloy-4. Phases of Al with Zr were not observed in diffusion region of Zircaloy-4 in both the cases whereas the layer structures having different composition were observed in Al-Fe-Ce alloy bonded using Cu interlayer. Hardness of intermetallic layers was found higher as compared to the parent alloys in case of Cu interlayer.

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