



Diffusion bonding of stainless steel to Zircaloy-4 in the presence of a Ta intermediate layer

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Abstract

Diffusion bonding of stainless steel to Zircaloy-4 in the presence of Ta as interlayer has been investigated using diffusion couples heat treated at 1150 °C under vacuum for 3 h. Different types of regions have been observed in stainless steel, Zircaloy-4 and Ta foil. A Cr rich layer formed in stainless steel is found to act as a diffusion barrier. A region observed with the Ta foil in Zircaloy-4 is rich in Cr, Ta, Fe and Zr while two zones are formed in Zircaloy-4 containing Zr, Ta, Cr, Fe and Ni. The zone rich in Zr gives black contrast and the other zone gives grey contrast and these are identified as eutectic phases of the type $\text{TaCr}_2\text{-Liq.-}\beta\text{-Zr}$ and $\text{Ta}(\text{Cr,Fe})_2\text{-Liq.-}\beta\text{-Zr}$.

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1. Introduction

Bonding of Zircaloy to stainless steel has been studied in the past by different methods by many authors [1–4]. They observed that the diffusion zone formed between the two alloys contains intermetallic compounds due to the reaction of diffused Fe, Cr and Ni from stainless steel with Zr. These intermetallic compounds are brittle in nature and reduce the strength of the bond. It was found that Ni diffuses to longer distance in the Zircaloy [3]. The diffused Ni in Zircaloy even in small quantity results in increased hydrogen absorption during oxidation at high-temperature in water and steam, which would be harmful.

The idea of using an intermediate layer was presented long ago. Tylecote [5] used an Al intermediate layer to facilitate welding of Cu/Cu. Kilbride and Adams [6] used a titanium interlayer in the welding of Zircaloy and

stainless steel to avoid the formation of an intermetallic compound. Shaaban and Hammad [7] used a thin Fe foil as interlayer in bonding of Zircaloy and stainless steel at 950 and 1000 °C for 5 min. Their results show that Zr_2Fe is formed in the diffusion zone. They also observed a $\text{Zr}_{18}\text{Sn}_{1.4}$ Fe phase in the diffusion zone. Sweeney and Batt [8] detected FeZr_3 , FeZr_2 and FeZr phases in the diffusion zone of the Fe/Zr couple heat treated at 875 °C for 113 h. There is no report available on the diffusion bonding of Zircaloy-4 to stainless steel using Ta as interlayer to the knowledge of the authors. The aim of the present paper is to study diffusion bonding of Zircaloy-4 and stainless steel 304L by introducing a Ta foil as interlayer and to characterize the phases that are formed in the different regions of the diffusion zone.

2. Experimental

The nominal composition of the two alloys is given in Table 1. Samples of the size $1 \times 1 \times 0.1 \text{ cm}^3$ were cut by spark erosion and then the two joining surfaces were polished on a lapping machine using diamond paste up to 0.25 μm . A thin Ta foil was placed between the

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Table 1
Nominal composition of the alloys

Alloys	Elements in wt%							
	Sn	Cr	Ni	Si	C	Mn	Fe	Zr
Zircaloy-4	1.52	0.10	<0.005	–	0.002	–	0.20	Bal.
Stainless steel 304L	–	19.0	10.0	1.0	0.03	2.0	Bal.	–

stainless steel SS 304L and Zircaloy-4 in such a way that it remained in touch with large surface area of the two alloys. Pressure of 4.9×10^3 bar was applied on the samples. After applying the pressure the samples were wrapped by a platinum–rhodium wire around them. The samples were then put in the vice machine for 24 h to introduce plastic deformation to both surfaces and Ta foil to enhance the diffusion rate. Afterwards the samples were heat treated at (1150 ± 2) °C for 3 h in vacuum maintaining a pressure of 7×10^{-8} bar and cooled down to room temperature under vacuum to avoid any oxidation. Samples were again polished on the lapping machine after removing about 2 mm of the sample surface to get the real view of the diffusion zone. The samples were etched in a solution as given previously [9] to reveal the microstructure of the diffusion zone. The investigation of the diffusion zone was done using a scanning electron microscope attached with the energy dispersive system (EDS). Quantitative analysis was done using the ZAF correction.

3. Results and discussion

Diffusion bonds between Ta and stainless steel and between Ta and Zircaloy-4 are shown in the scanning electron micrograph as given in Fig. 1. It shows that the bond on the stainless steel side is very smooth while slightly rough on Zircaloy-4. The diffusion zone shows

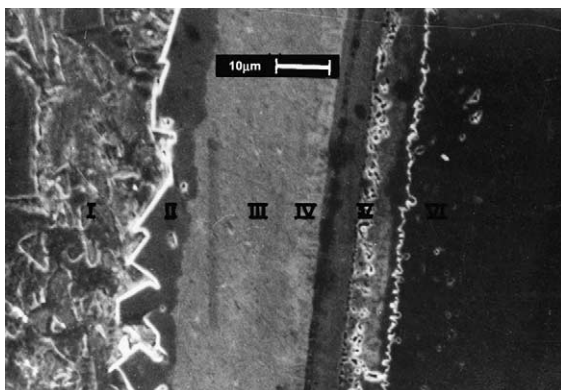


Fig. 1. SEM micrograph shows the different regions in the diffusion zone.

six different regions. These regions are named as I, II, III, IV, V and VI. Regions named I and II are on the Zircaloy-4 side and regions V and VI are on the stainless steel side. Region IV is formed in Ta on the side of stainless steel while region III consists of the remaining area of the Ta foil.

Fig. 2(a) gives a view of the diffusion zone at high magnification. In order to observe the concentration variation of various elements in different regions, line scans have been taken across the line shown in Fig. 2(a). These line scans are given in Fig. 2(b). The presence of different regions in the diffusion zone is also evident from the X-ray mapping of different elements as shown

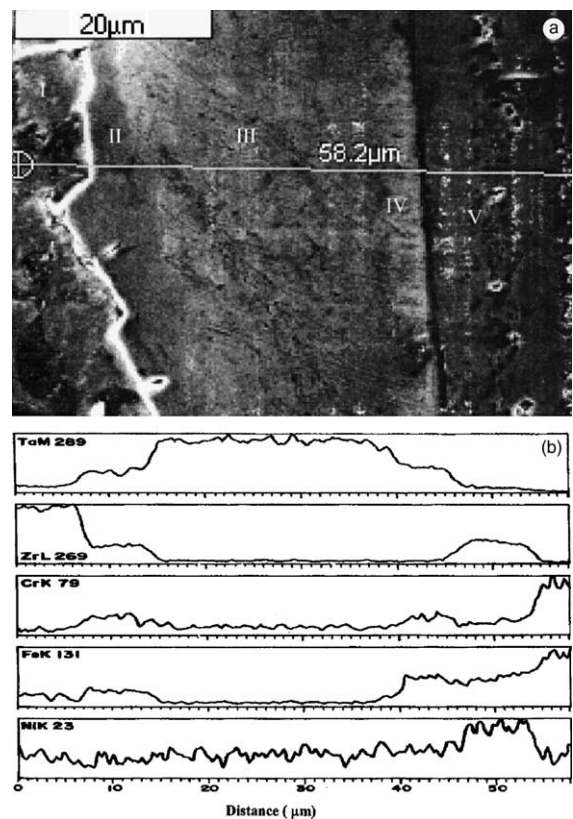


Fig. 2. (a) SEM micrograph of the diffusion zone at high magnification. (b) Line scan across the line shown in Fig. 3(a) showing the X-ray intensity variation of the elements Ta(M_{α}), Zr(L_{α}), Cr(K_{α}), Fe(K_{α}) and Ni(K_{α}).

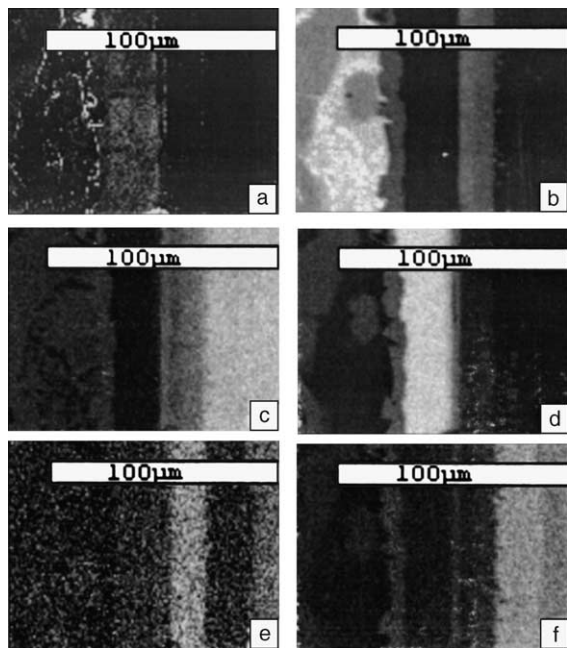


Fig. 3. Secondary electron image (a) and corresponding X-ray maps showing the concentration variation of the diffusing species like Zr L_{α} (b), Fe K_{α} (c), Ta M_{α} (d), Ni K_{α} (e) and Cr K_{α} (f) in the diffusion zone area.

in Fig. 3. The results of line scans and X-ray mapping clearly indicate the diffusion of the elements Fe, Ni and Cr from stainless steel towards the Ta foil and Zircaloy-4, and diffusion of Zr and Ta from Zircaloy-4 and the Ta foil towards stainless steel. The diffusion of Ta takes place towards both the alloys. Region I on the side of Zircaloy-4 is rich in Zr and Ta and region II contains Zr, Cr, Ta, and Fe. Region III shows maximum concentration of Ta. Region IV has higher contents of Fe, Cr and Ta and region V contains Zr, Ni, and Fe in higher quantities. In this region the concentration of Zr decreases while the concentration of Fe increases with the increase in width towards region VI. In region VI the concentration of Zr is almost zero and the concentration of Cr is about 25% higher while Ni is 50% lower compared to the matrix of SS 304L. The X-ray images further show the presence of two zones in region I with a variation in Zr and Ta concentration.

In order to obtain quantitative data, point analyses were carried out at several points in each region and the average composition is given in Table 2. The results show that there are two zones within region I. The concentration of Ta varies in both the zones with an increase in distance from the Ta foil towards Zircaloy-4. The zone with higher content of Ta is also rich in Cr and Fe, and the zone containing the higher Zr content has a less concentration of Fe and Cr. The diffusion of the elements Fe, Cr, Ni and Ta towards Zircaloy-4 lowers

the temperature of Zircaloy-4 and as a result eutectic reactions of Zr occur with these elements. The zones formed in Zircaloy-4 due to the above eutectic reactions give grey and black contrast in the secondary electron image as shown in Fig. 4. These eutectic phases are identified as $Ta(Fe,Cr)_2-Liq.-\beta-Zr$ and $TaCr_2-Liq.-\beta-Zr$. The region II containing Ta, Cr, Zr and Fe forms a layer of eutectic phases of the type $Ta(Fe,Cr)_2-Zr(Fe,Cr)_2-solid\ sol.(Zr,Ta)$.

Two regions have been observed on the Ta foil. The major portion of the foil, marked as region III, does not contain an appreciable amount of any other element. This may be due to the fact that the solid solubility of Fe, Cr, Zr and Ni is negligible in Ta at room temperature. These elements may form different phases with Ta like $TaFe_2$, $TaCr_2$, $ZrCr_2$, Zr_2Ni , etc. However, these phases may be too small and thin to determine by EDS. Region IV which is rich in Fe, Cr and Ta is formed in Ta foil near stainless steel and is identified as a phase of the type $Ta(Fe,Cr)_2-Ta_3Ni_2$. This may be due to the fact that the series of solid solution of Fe, Cr and Ni is available as this region is near the stainless steel.

Region V, rich in Zr and Ni, is formed because Ni has a strong interaction with Zr due to the large difference in their electronegativities. The concentration of Cr and Fe is lower in this region compared to stainless steel. This layer is characterized as a mixture of phases of the type $Zr(Fe,Ni)_2-TaCr_2$. The $Zr(Fe,Ni)_2$ phase has already been observed by [11–13] during the analysis of the interaction between Zircaloy-4 and Inconel-718. Region VI is found to have a lower Ni content and an increase in Cr as compared to the matrix stainless steel. The Ni depletion in the Cr rich region promotes the ferrite structure. This region, rich in Cr and depleted in Ni, has already been reported by Lucuta et al. [4], Blanc et al. [10], Uetsuka et al. [11] and Ahmad et al. [9] on the stainless steel side. This region is found to act as a diffusion barrier for Ta and Zr towards stainless steel. Some carbides and voids which give black contrast, as shown in Fig. 1, are also observed in regions V and VI. These carbides are formed in stainless steel near Ta due to the diffusion of Ta towards the stainless steel and because Ta is also known as strong carbide former.

4. Conclusions

In the diffusion bonding of stainless steel 304L and Zircaloy-4 in the presence of a Ta interlayer, eutectic phases like $Zr(Fe,Ni)_2-TaCr_2$ and $Ta(Cr,Fe)_2-Ta_2Ni_3$ are formed on the stainless steel side in the Ta foil and in stainless steel whereas $Ta(Cr,Fe)_2-Zr$ ($Fe,Cr)_2-Zr_3Ta_2$ and $Ta(Fe,Cr)_2-Liq.-\beta-Zr$ and $TaCr_2-Liq.-\beta-Zr$ are formed in Zircaloy-4. A Cr-rich layer observed on the stainless steel side acts as a diffusion barrier. Carbides and voids are also observed in stainless steel on the Ta

Table 2
Quantitative analysis of regions/layers formed in the diffusion zone

Regions/layer	Elements	Concentration (wt%)	Phases
I In Zircaloy-4 (i) Grey contrast	Cr	2.00	Ta(Fe,Cr) ₂ -Liq.-β-Zr
	Fe	9.85	
	Zr	66.45	
	Ta	20.00	
	Ni	1.15	
	Sn	0.55	
(ii) Black contrast	Cr	1.20	TaCr ₂ -Liq.-β-Zr
	Fe	7.00	
	Zr	77.00	
	Ta	11.50	
	Ni	1.30	
	Sn	2.00	
II Layer near Ta	Cr	5.60	Ta(Fe,Cr) ₂ -Zr(Fe,Cr) ₂ -solid sol.(Zr,- Ta)
	Fe	11.90	
	Zr	33.50	
	Ta	49.00	
	Ni	–	
	Sn	–	
IV Ta-Fe rich region	Cr	4.85	Ta(Fe,Cr) ₂ -Ta ₃ Ni ₂
	Fe	25.55	
	Ni	2.60	
	Sn	0.50	
	Ta	66.50	
	V Zr-Ni rich layer on the side of S.S.	Cr	
Fe		33.60	
Ni		10.50	
Zr		44.50	
Ta		6.82	
IV Cr-rich layer		Cr	25.50
	Fe	69.20	
	Ni	5.30	

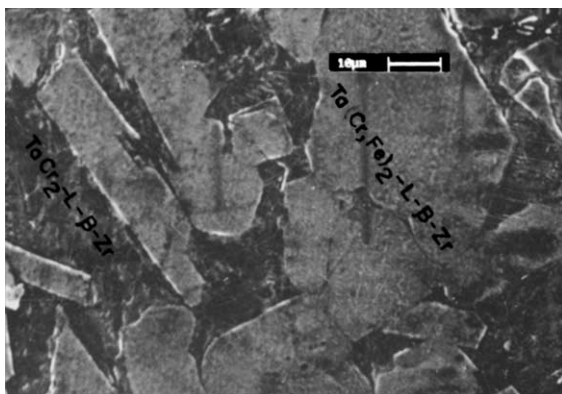


Fig. 4. The SEM micrograph shows the dark and grey contrast with different composition having eutectic like phases.

foil side. Different phases that are formed in the Ta foil are so small and thin, that they are not detected by EDS.

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