Fe-Al Diffusion Bonding: Effect of Reaction Time on The Interlayer Thickness

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ABSTRACT

A direct diffusion bonding of marine grade aluminium alloy AA6061 and steel A36 was carried out in an argon environment at 640°C. Specimens were heated in a furnace for a holding time of 90 to 120 min under a pre-pressed condition. An interface microstructural observation revealed an increase in the thickness of this reaction layer as bonding time increased. A 33.4 μ m thickness was recorded at the interfacial zone of the joint after a 120-min holding time condition. Hardness along the bonding line was measured using a micro-Vickers hardness tester. Diffusion of aluminium and iron was observed on both parent metals at the interfacial zone using scanning electron microscope (SEM) with energy dispersive spectroscopy (EDS). This experiment had proven that different content and thickness of alloy compounds were formed at various reaction time frames on the interfacial zone.

Keywords: diffusion bonding; interlayer thickness; dissimilar metals; solid state; aluminium;

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Introduction

Improved technology is seen as an increasing trend in industries to manufacture products consisting of a combination of different metallic components. An example is a car body that is sometimes made of aluminium and welded to a steel mainframe so as to reduce weight without compromising safety and quality. The same idea is applied in the shipbuilding industry, where an aluminium superstructure would be welded to the steel hull of a ship.

Conservatively, adhesive bonding, mechanical fastening and welding are typical modes of joining dissimilar metals. In welding, the joining of steel to steel would involve the fusion process, while diffusion process would be applicable in the joining of ferrous to non-ferrous metals.

Diffusion welding or diffusion bonding is a solid-state welding process, which also includes forge welding, friction stir welding (FSW), and explosive welding. The principal joining mechanism is the application of a pressure to bring the mating surfaces to within an inter atomic distance [1]. The joining process is accomplished through the application of pressure at an elevated temperature, about 50% - 90% of the melting temperature of parent metals; mostly conducted in a vacuum to reduce the effects of surface oxidation.

Diffusion bonding of materials in the solid state is a monolithic joint- making the process by forming bonds at the atomic level through the closure of the mating surfaces brought about by local plastic deformation at an elevated temperature to enable inter-diffusion to take place at the surface layers of the joining materials [2].

The application of the diffusion bonding process is normally in the fabrication of parts, when high-quality and high-strength welds are required, especially where part shapes are intricate and costly or impossible to manufacture by conventional means or when the materials used possess certain properties that may interfere with or are difficult to maintain during the conventional fabrication process.

Ferrous and non-ferrous are poorly weldable by fusion welding because they tend to crack in the near-weld zone both during and after welding. In fusion welding, the melted aluminium absorbs hydrogen released in the fusion process that promotes hydrogen embrittlement and makes the joint brittle. Failure of the reaction zone is attributed to this intermetallic embrittlement of compounds at the joint. The presence of aluminium induces its plastic fluidity that hasten the formation of Fe-Al intermetallic compound (IMC) at the interface [3].

Welding of aluminium and steel is possible through the diffusion bonding process since issues faced during fusion welding of these dissimilar metal joints can be eliminated; any alloying process between the metals is insignificant, and the potential structural failures, especially at the joint interface, can be reduced.

One method of diffusion bonding is FSW. A challenging issue in the joining of dissimilar metals using FSW is the ability and reliability of the handling tool to remain intact after reaching a certain temperature [4]. Consequently, the mechanical property of carbon steel is altered by the heating process [5].

Another method of diffusion bonding is the explosion welding process. The explosion welding approach is used in the mass production of 'triclad', i.e. of aluminium and steel joining, which is used as transition joints for ships made of the steel hull and aluminium superstructure. Some drawbacks of this process include the requirement for high energy explosive materials and the generation of incredible noise as it has to be conducted remotely.

The main factor for diffusion to occur is temperature; however, other important factors that affect the quality of a joint are holding time, holding pressure, heating rate, surface roughness and atmospheric condition [6, 7].

With regard to this hot press bonding approach, further investigation into the effects of reaction (holding) time towards the interlayer thickness is hereby undertaken. The hardness, interlayer thickness, microstructural development and properties (EDS analysis) of the joints are presented in this research. Priority is focused on the quality of joints as reflected in having a quality finish that is free from internal and surface defects [8].

Methodology

Materials used were bars of marine grade aluminium (A6061) and steel (A36). The 6061 alloy and A36 contain impurities of 0.70 wt % iron, and 0.015 wt % aluminium, respectively.

The specimens were machined to produce rods of $\emptyset 10 \text{ mm x } 20 \text{ mm}$; with faying surfaces set perpendicular to the extrusion direction, then polished to 1µm finish. No interlayer was used as this is a direct diffusion bonding process. The polished surfaces were rinsed in acetone and dried by hot air before joining. The schematic representation of fixture and furnace (Lyn furnace) used during diffusion is shown in Figure 1.



Figure 1: Diagram of Specimens Assembly (not to scale)

The basic fundamental to joining components via diffusion bonding process is through the application of a low pressure at a high temperature. Faying surfaces of aluminium and steel were assembled using a G-clamp fixture. The clamp was pre-pressed by hand prior to positioning inside a programmable electric furnace. The temperature during the bonding process was monitored. The inert atmosphere of argon gas was maintained at 15 litre/min, and the heating rate of the furnace was set at 10°C/min. The specimens were heated at 640°C, just above the solidus line but below the solvus line of the Al-rich solidus and solvus of 6061 alloy [9] for 90, 100, 110 and 120 min. The specimens were cooled down inside the furnace under a protective atmosphere at 5°C/min. A schematic diagram of specimens that underwent an experiment at 90 min. holding time is shown in Figure 2 below.



Figure 2: Diagram of joining cycle at 90 min holding time; T=temperature, t=time.

Diffusion of aluminium and iron was analysed to observe the microstructure and micro-constituent on both parent metals at the interfacial zone using scanning electron microscope (SEM) with energy dispersive spectroscopy (EDS).

Results and Discussion

For the purpose of determining the effects of time to the thickness of diffusion layer, observations, findings, non-destructive test results and characterizations are expressed as follows.

1. Micro hardness Vickers (HV)

After the joining process, specimens were tested for hardness measurements. Micro-hardness measurement was carried out using Vickers Hardness Scale under 200g load.

Hardness values at the joint of specimens are presented in Figure 3 below. Both sides of the interface were observed to decrease in micro hardness due to the formation of IMC at the mating surface, caused by the migration of aluminium and iron near the interface.

As the materials were exposed to heat for a longer period of time, the grain size became bigger. Comparing the four graphs, the hardness value of the holding time 120 min is the lowest due to the bigger grain size it has. The average grain size increases with time. According to Hall-Petch equation, the grain size is determined by plastic deformation in the existence of heat.

The shape and grain size reduction improves toughness and strength of materials. According to the hardness versus tensile strength conversion for steel, the lower the Vickers hardness (HV), the lower the tensile strength at the steel part. The similar pattern can be observed at the aluminium part, the holding time of 120 min has the lowest hardness. A short range diffusion of atoms from one metal to the other is due to the movement of the boundary and/or atom in an opposite direction.



Figure 3: Micro hardness (HV) distribution of interlayer after 90, 100, 110 and 120 min.

2. SEM and EDS analysis on diffusion layer

Microstructures at the section of the joint specimens were obtained using SEM. A protruded uniform flat interface was found between the intermetallic layer and the weld on the steel side as shown in Figure 4.

Diffusion thickness of joint increases at an elevated holding time. The average thickness of the interface width at central zone as a function of time are 16, 30 and 33 μ m, respectively, as shown in Table 1. It was found that when joining was carried out at a holding time of 120 min, the average thickness of the diffusion layer is 33 μ m. These thickness measurements indicate that the effect of holding time is significant of the joint integrity.

Figure 4 below shows SEM images of the interfacial region of the joints. In Figure 4(a), at t=90, the reactant was not observed at the joint interface at the holding time of 90 min. Occasionally, the reactant started to appear at the interface at the holding time of 100 min. At the holding time of 110 min, a reaction layer was in a continuous shape. In this study, the average thickness of 33 μ m was observed at the reaction layer when the holding time reached 120 min as summarised in Figure 5, the graph of diffusion layers thickness as a function of holding time.



Figure 4 (a), (b), (c), (d): Average thickness of joins after 90, 100, 110 and 120 min holding time, respectively.

Holding Time (min)	Diffusion Layer Thickness (μm)	Boundary Layer Hardness (Hv)
90	0	39.2
100	16.3	49.9
110	30	51.4
120	33.4	32.4

Table 1: Diffusion Layer of the Direct Diffusion Bonding

The effect of holding time on the diffusion layer thickness of the joint at a bonding temperature of 640°C is indicated in Figure 5 below. Figure 5 represents an average thickness of interfacial layers at joints after 90, 100, 110 and 120 min holding time, respectively.

For any Particle Diffusion to a Wall, by dint of the Brownian motion with Fick's Law; the diffusion boundary layer thickness, δ_c is given as

Asmawi Ismail et al.

$$\delta_c = \sqrt{\pi D t} \tag{1}$$

The total number of particles deposited in an interval dt is given as

$$dN = Jdt = C_o \sqrt{\frac{D}{\pi t} dt}$$
(2)

By integrating dN, the total number of particles deposited per unit area in the time interval 0 to t can be calculated. Thus,

$$N = C_o \sqrt{\frac{4Dt}{\pi}} \tag{3}$$

Bonding temperature, pressure and time are the main inter-related bonding parameters. Aside from temperature and pressure, this dissimilar metal joining of aluminium and steel was still successful when there was enough diffusion in a reasonable time. Time adequacy is to stimulate an interdiffusion reaction in promoting contact of faying surfaces (i) by way of plastic deformation through eliminating the residual voids between surfaces, and/or (ii) via plastic flow that exists within the parent metals during the bonding process.

There is a relative study reported by José Lemus-Ruiz, on the definite effect of the metal layer thickness towards the mechanical properties of a joint, at which the reaction layer thickness has got to be controlled as to ensure good joint strength [10]. The formation of bonds via chemical reaction, as a result of which, the growth of new phases at the interface seems to be the main concern to overcome. As summarised in Figure 4 and 5 above, with respect to time, it is strongly believed that at a certain diffusion layer thickness, the optimised value of strength at the joint is possible to be determined. Bonding time is partially involved to influence the strength quality at the join because as time prolongs, the chemical reactions form bridges that enhance the joining quality. Inasmuch as the inter-diffusion reaction of iron into the aluminium, this thin intermetallic diffusion layer dominated at the aluminium side of the joint.



Figure 5: Diffusion Layer Thickness as a function of holding time

3. EDS Analysis

Reaction time setting affects the morphology of intermetallic layer, which finally has an effect on the strength of the joint. Characterizations of the intermetallic layer are as shown in Figure 6 below that represent the EDS analysis near to the joint interface corresponding to the interlayer morphology at various holding reaction time frames.

The identification of phases, the weld zone and base metal were carried out by EDS spot analysis; as important parts of the compositional transition analysis. The development of diffusion by line scan near the interface of the joint was from the condition of 90, 100, 110 and 120 min reaction time, respectively.

A concentration of aluminium was scanned along a line across the bond centreline. In Figure 6 (a), at the 90 min of holding time, diffusion layer was about to form, nevertheless, neither aluminium nor iron occurred at the interlayer.

Asmawi Ismail et al.



Figure 6: Line scan result representing concentration variation of aluminium and iron around the bonded interface after 90 (a), 100 (b), 110 (c) and 120 (d) min of holding time. (brown: represents Al; yellow: represents Fe).

The elements of aluminium and iron were detected in the reaction layer and their content was lower as compared to the as-received original parent metal samples. It was relatively a small amount of parent metals that were detected inside the intermetallic layer.

In Figure 6 (b), at the 100 min, diffusion started to grow. Initially, the concentration of aluminium at AA6061 side was more than 90 wt%. The aluminium concentration of more than 60 wt% was observed in the interdiffusion layer, which was about 75% of the initial aluminium concentration. At the steel A36 side, the initial concentration of iron was more than 80 wt%. The iron concentration of more than 20 wt% was observed in the diffusion layer, which was about 25 wt% of the initial iron concentration.

In Figures 6 (c) and (d), the thickness of the diffusion layer increased as the holding time increased. At certain parts of the reaction layer, the (map) specific chemical composition analysis showed that the average

mapping or atomic concentration of aluminium and iron was 69.9 wt% and 27.8 wt%, respectively. It was detected that there was a possible IMC of AlFe [11].

The content of the main element such as aluminium and iron was stable at the diffusion layer, indicating the possible existence of IMC. At a particular reaction time, the migration of aluminium and iron occurs at the elevated joint temperature, about 75 wt% aluminium and 20 wt% iron were diffused into the parent metals during the bonding process, especially on the 120-min sample.

As the holding time increased, the amount of iron content was generally decreased at the interface layer. On the other hand, the aluminium distribution increased with respect to the reaction time.

Conclusion

As a conclusion, there are few important points that can be drawn as follows:

- (1) A direct joining of aluminium A6061 to steel A36 without application of a strip layer or filler metal in an Argon-controlled environment or protective atmosphere is possible.
- (2) The reaction layer thickness is related to the bonding time of the joint, as the thickness increases with time. The content of aluminium and iron were comparatively balanced inside the intermetallic layer at a higher reaction time.
- (3) The longest holding time demonstrates the lowest hardness values on both parent metals' intermetallic layer due to the heating process during bonding.
- (4) A movement of iron atoms occur at the correct combination of temperature and holding time, as the grain size is a time and temperature dependent variable. The EDS results indicated that the content of iron in the intermetallic layer varies with different reaction time at the interface of aluminium and low carbon steel.

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