

Transient liquid phase bonding of Al-6063 to steel alloy UNS S32304

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Abstract

Transient liquid phase (TLP) bonding of Al-6063 and UNS S32304 was performed using copper foil as an interlayer between the base metals. A compression load was applied normal to the specimens. Metallurgical examination of the produced joints showed three distinct regions including a reaction zone, diffusion affected zone and the base metals. The diffusion of copper into aluminum resulted in an Al-Cu eutectic structure. However, the oxide layer on the aluminum surface controlled the dissolution behavior of copper and the extent of its wettability with the base metals. Although voids and intermetallic compounds were detected at the interfaces of the processed joints, a defect free joint was produced at 570°C. In addition, the results from corrosion tests showed that the use of copper as an interlayer decreased the corrosion resistance of the joints. However, increase in thickness of the joining reaction zone with increasing bonding temperature was observed to increase corrosion resistance.

Keywords: TLP bonding; Eutectic; Aluminum alloys; Duplex stainless steel.

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1. Introduction

There is a growing demand for cost effective materials with enhanced engineered properties. This involves the use of dissimilar metals. Although dissimilar metal-couples are being used in the aerospace and oil and gas sectors, the bonding of dissimilar metals is still facing numerous challenges and difficulties. The difference in thermal expansion coefficients of dissimilar metals can result in residual stresses within the joint region which can eventually cause failure of the joint when conventional fusion welding is used. Furthermore, the large difference in melting point between different metals (e.g. aluminum and steel) makes the use of fusion welding processes difficult to apply to these metals.

Stub ends, flanges and pipe fittings are examples of parts and structures, where dissimilar metal joints are used. Furthermore, many engineering components where corrosion susceptibility is high, the potential difference between the dissimilar materials is of great concern. The formation of intermetallic compounds (IMCs) during welding is a challenge for scientists and researchers, such compounds can easily result in brittle fracture of the joints. Therefore, all these factors must be considered in order to produce good dissimilar metal-couples [1-3].

Several methods have been used to bond dissimilar metals [4]. Each one has its own advantages and limitations. Selecting the most appropriate method is vital for accomplishing successful joints. Fusion welding processes, solid-state bonding, adhesive bonding, brazing and soldering are the most well-known methods of bonding dissimilar metals. Unlike fusion welding, solid-state diffusion bonding does not require melting of the base metals. Many researchers have investigated different methods of bonding metals and their behaviors [5-7]. However, the number of published articles on transient liquid phase (TLP) bonding and hybrid bonding processes, is

still quite few compared to arc welding process such as gas tungsten arc welding (TIG), shielded metal arc welding (SMAW) and gas metal arc welding (MIG).

In this paper a feasibility study is undertaken to fabricate joints between an Al-6063 aluminum alloy and a duplex stainless steel (UNS S32304), using a pure Cu foil as an interlayer between the base metals and at a temperature lower than conventional welding processes. The advantage of using Cu between Al and steel, is that Cu can make an eutectic reaction with Al, and at the same time one avoids the formation of IMCs with Fe [8].

2. Materials and Methods

UNS S32304 and Al-6063 were supplied by (ThyssenKrupp Nirosta, Germany) and (Qalex, Qatar), respectively. Samples were machined to rectangular dimensions of 30 mm X 15 mm. Aluminum samples were fabricated from an extruded Al-6063 sheet of 2 mm thickness, while duplex stainless steel samples were in the form of coupons with a thickness of 1 mm. A copper foil with 99.9% purity and 10 μ m thickness was obtained from Goodfellow, UK, was used to form an interlayer between the aluminum and stainless steel sheets. The base metal surfaces to be bonded, were ground to 1000 grit surface finish using SiC paper, cleaned with ethanol and dried using hot air. Each joint consisted of two overlapping samples with a piece of copper foil sandwiched in between.

The TLP bonding process was executed using a thermo-mechanical simulator of type Gleeble 3800, in an inert atmosphere (argon) and a heating rate of $100^{\circ}\text{C min}^{-1}$. A thermocouple wire was welded to the edge of the aluminum sample; approximately in the middle of the overlap region in order to ensure an accurate monitoring of the specimen temperature. A compression load of

0.2 KN was applied to the specimens. The processing temperature to form the joints was varied between 550°C, 555°C, 560°C and 570°C. Fig. 1 shows a sample setup in the Gleeble system.

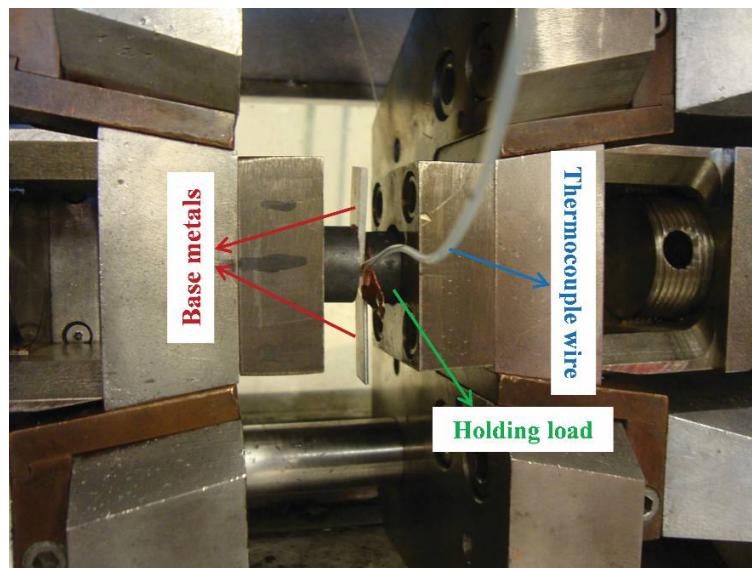


Fig. 1 Sample set up for bonding in the Gleeble system.

The microstructural development at the joint interface of the copper interlayer was characterized in a scanning electron microscope (SEM, Nova NanoSEM 450) equipped with energy dispersive X-ray spectroscopy (EDX) and a X-ray diffraction analysis unit (XRD, PANalytical Empyrean). Al-6063 and UNS S32304 were grinded and polished to a 1 μm finish using a diamond suspension. After polishing, Al-6063 was etched with Weck's reagent, while UNS S32304 was etched with Kalling's reagent. An optical microscope (Leica DM IRM) equipped with Clemex image analysis software, was used to measure the size of aluminum grains. Micro-hardness measurements across the interface were conducted in accordance to ASTM E384 using a load of 50 grams in a Vickers micro-hardness tester (Future-Tech ARS900). The hardness profile included 11 points measured from the center of the joint to 500 μm on both sides of the interface.

The indentation spacing was 0.1 mm. Further, corrosion rates of the joints were evaluated using potentiodynamic polarization tests in a Gamry 600 potentiostat, and a 3.5% NaCl solution.

3. Results and Discussion

3.1 Microstructural development in joints

The resultant area of bonding in various samples can be categorized into three distinct zones (Fig.2), i.e. base metal (Z1), reaction zone (Z2) and diffusion affected zone (Z3); The latter is the area adjacent to Z2 at both sides extending into the Al-6063 and UNS S32304, respectively.

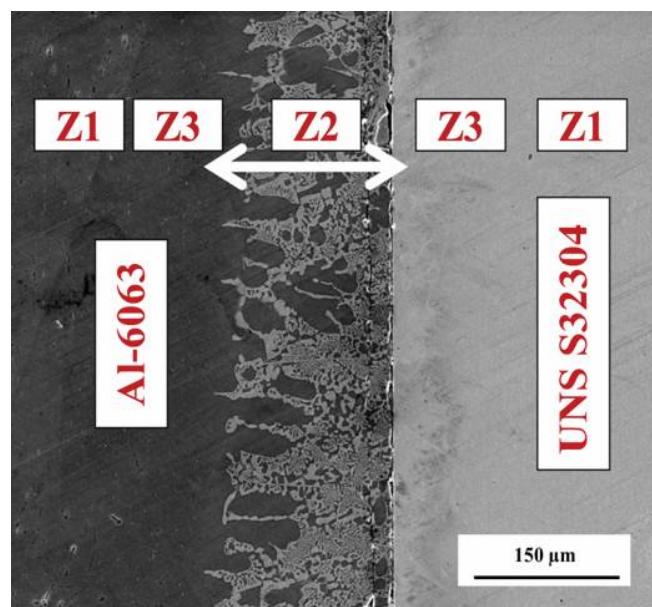


Fig. 2 SEM micrograph showing resultant reaction zones after the bonding process. (TLP-560°C-5min).

In general, the wettability of liquid Cu on the Al-6063 substrate is relatively high. Hence, liquid copper spread easily along the whole joint interface. On the contrary, the wettability of Cu on UNS S32304 is low and no intermetallic zones formed at the Cu/Fe interface. The formation of Al-Cu eutectic phase occurred at the interface. As soon as the eutectic liquid forms, Cu will tend

to diffuse into Al rather than the steel. This occurs as a consequence of the much higher value of the diffusion coefficient of Cu in Al than for Cu in Fe. An undissolved portion of Cu was observed at the joint interface of some samples, while the samples processed at 570°C showed a defect-free joint with a thick reaction zone having a dense sub-zone closed to the interface (See Fig. 3-c).

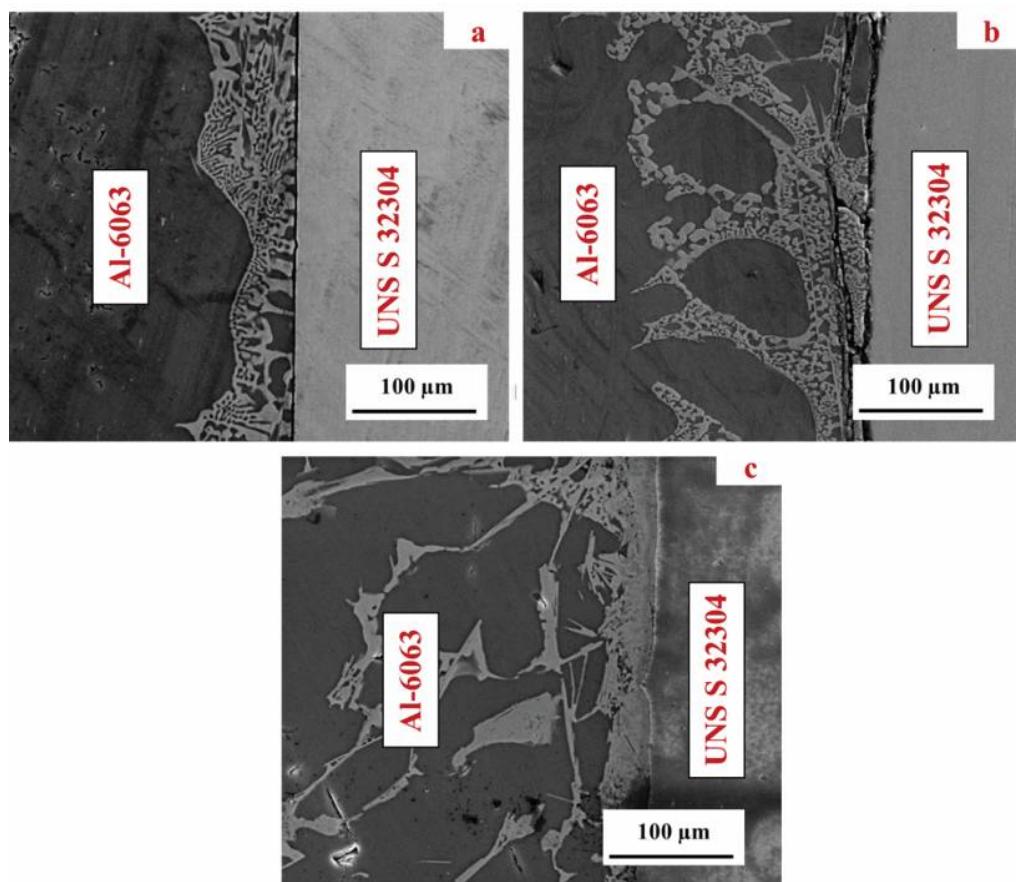


Fig. 3SEM micrographs of Al-6063-UNS S32304 joint interface produced by TLP bonding at (a) 550°C; (b) 555°C; (c) 570°C.

The thickness (Z) of the reaction zone $Z2$ is a function of temperature (T) and time (t) and can be expressed by the following parabolic equation :

$$Z = (2Kt)^{0.5}$$

and

$$K = K_0 \exp(-Q/RT) \quad (1)$$

Hence, K is the growth velocity (m^2s^{-1}); K_0 a growth constant (m^2s^{-1}); R is the gas constant (8.314 KJmol^{-1}) and Q is the activation energy (KJmol^{-1}) [9, 10].

Further, a direct correlation between the bonding temperature and reaction zone thickness could be found, i.e. see Fig. 4. The average thickness of Z2 increased from 96 μm at 550°C to 217 μm at 570°C.

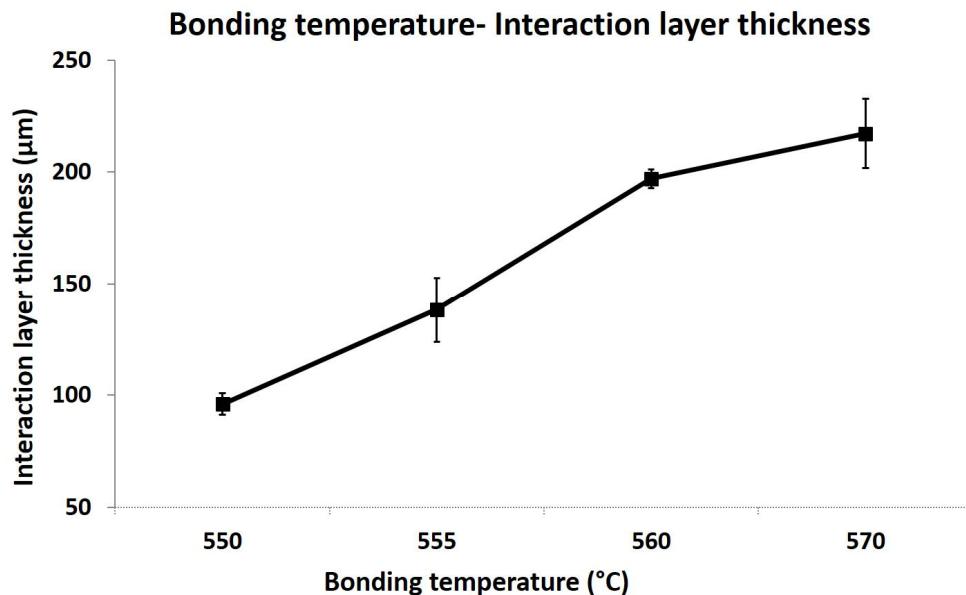


Fig. 4 The relationship between bonding temperature and thickness of the reaction zone.

Fig. 5 and Table 1 show the location of the EDX analysis and the corresponding, chemical compositions, across the interface, respectively. The diffusion coefficients of Cu in Al ($D_{\text{Cu} \rightarrow \text{Al}}$) and Cu in Fe ($D_{\text{Cu} \rightarrow \text{Fe}}$), at same temperature, are about six order of magnitude different.

Therefore one observed that Cu diffused more easily into the Al-alloy than the steel [8, 11]. In comparison insignificant amounts of Cu (≈ 0.8 at. %) were detected in points D and E, e.g. along the joint interface at the UNS S32304 side. The stainless steel bonded at 570°C, absorbed Cu to a content close to 0.8% (See Table 1). In fact, in the literature it was reported that the solubility of Cu in Fe can be enhanced by raising the bonding temperature [8].

The highest Cu concentration between 28-33 at.% was observed at Point C, located in the Al-alloy. Furthermore, diffusion of Cu extended into the Al-alloy away from the joint. The decrease in Cu content at the joint interface resulted in isothermal solidification at the eutectic temperature [12].

Moreover, diffusion of Fe into Al was also observed. An approximate amount of 0.3 at.% of Fe was detected at point B. The lowest percentage of Al detected at points D and E were 0.32 and 0.31 at.%, respectively which is due to the large difference in the diffusion coefficients between Fe in Al ($D_{Fe \rightarrow Al} = 1.00 \times 10^{-13} \text{ m}^2 \text{s}^{-1}$) and Al in Fe ($D_{Al \rightarrow Fe} = 9.94 \times 10^{-19} \text{ m}^2 \text{s}^{-1}$) [13].

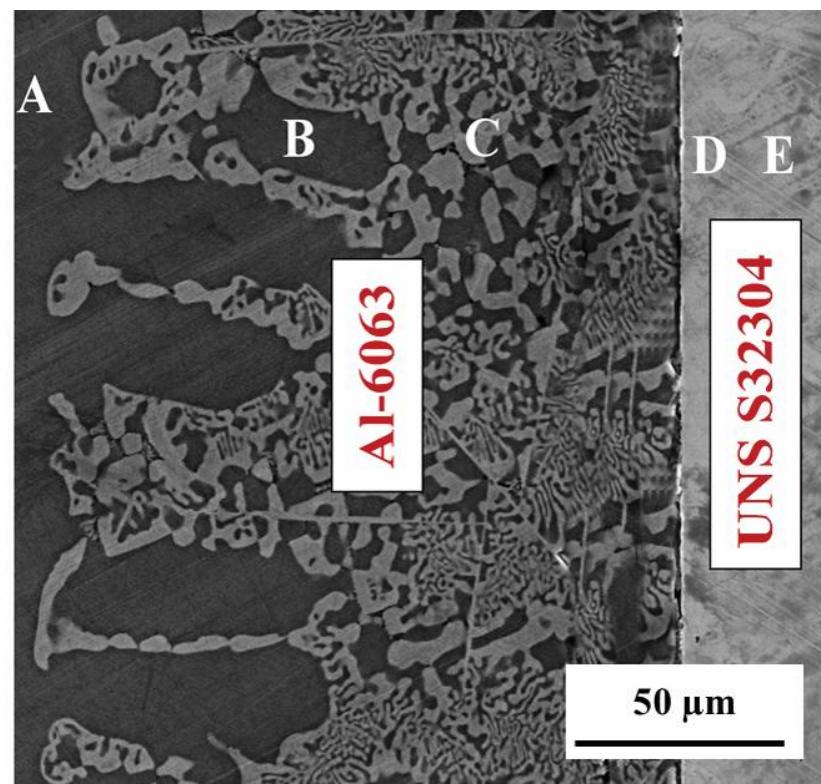


Fig. 5 SEM micrograph showing the selected five points (A to E) for EDX analysis (TLP-560°C-5min).

Table 1 Shows the composition for points selected for EDX analysis in Fig. 5.

Point	Region	Composition (at%)
A	At Al-6063 side adjacent to the joint interface (zone Z3)	Al 94.29%, Cu 2.46%, Fe 0%.
B	Area surrounded by Al-Cu eutectic structure (zone Z2)	Al 95.48%, Cu 3.15%, Fe 0.32%.
C	Al-Cu eutectic structure (zone Z2)	Al 67.04%, Cu 32.96%.
D	At the duplex stainless steel side	Al 0.32%, Cu 0.79%, Fe 51.61%.
E	adjacent to the joint interface (zone Z3)	Al 0.31%, Cu 0%, Fe 71.81%.

The shown XRD pattern revealed the existence of Al_2Cu at the interface near the Al-6063 side (See Fig. 6). This is in good agreement with the EDX data (Table 1). The presence of FeAl_3 as indicated by Sun et al. (2015) has not been observed in the present case, which is presumably because Cu acted as a transition material between Al and Fe, preventing direct contact and hence suppressing the formation of Fe-Al intermetallic compounds [14].

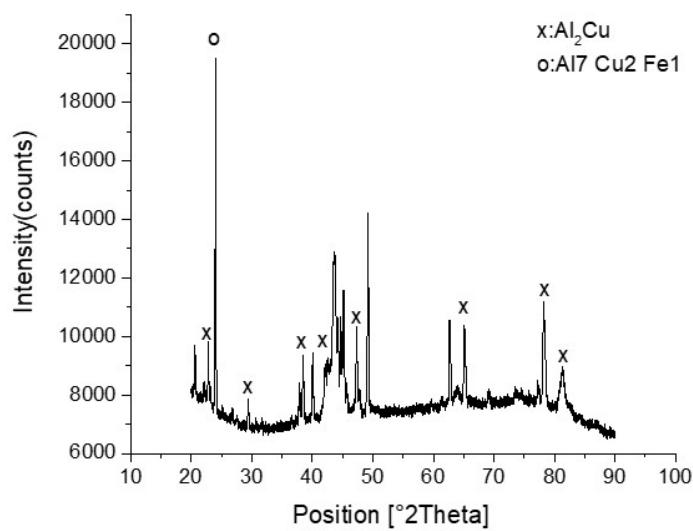


Fig. 6 XRD spectrum of the eutectic region of a TLP joint.

The highest magnification view of the microstructure shown in Fig. 7 indicates that Cu diffused from the joint interface into the Al-alloy along grain boundaries. Therefore, one can claim that grain boundary diffusion is dominant over lattice diffusion during the present bonding process.

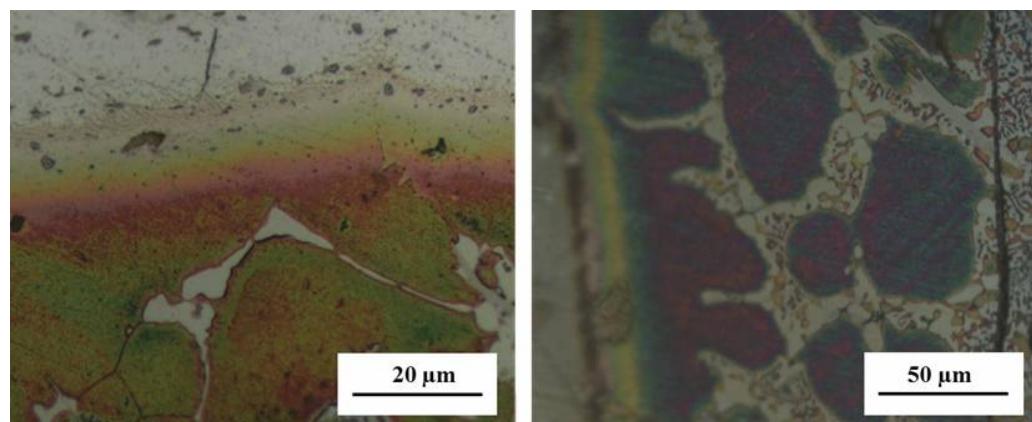


Fig. 7 Close-up of microstructure of Al-6063 in zone Z2 after the bonding process.

The diffusion path depends on several factors such as microstructure, temperature and the interface quality between the metal and adjacent layers [15]. The Arrhenius' law describes the effect of activation energy (E_a) on the diffusion coefficient (D) :

$$D = D_0 e^{-E_a/RT} \quad (2)$$

Grain boundary diffusion has a fast diffusion path due to the open structures of the boundaries. The latter formed a network along the whole area of the specimen. Also, voids and imperfections along Al grain boundaries render the diffusion of Cu easier than diffusion through the lattice [15-17].

Fig. 8 shows the microstructure of UNS S32304 steel before and after the bonding process. Unlike Al, because of the presence of Cu, the etching process was more aggressive on the post-welded samples than the as-received ones. It was also noticed that ferrite grains (α -Fe) were significantly more etched than the austenite grains (γ -Fe). This could be attributed to the significant difference between Cu diffusivities in α -Fe and γ -Fe

(4.4×10^{-9} and $9.4 \times 10^{-12} \text{ cm}^2 \text{s}^{-1}$, respectively). In addition, α -Fe (BCC) has a more open atomic structure than γ -Fe (FCC) [17, 18].

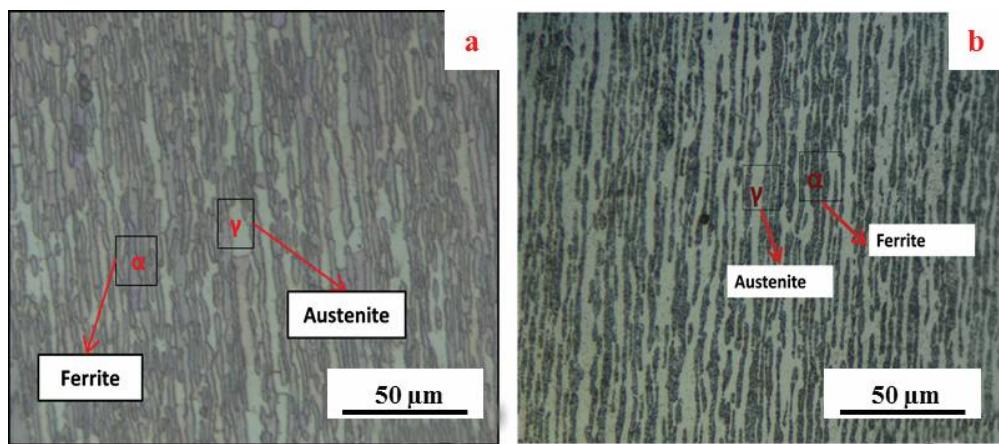


Fig. 8 Austenite and ferrite grains of UNS S32304 (a) before the bonding process;(b) after the bonding process at (550-570°C).

3.2 Micro-hardness evaluation

Fig. 9 shows the micro-hardness profile for samples made by TLP bonding at 550°C, 555°C, 560°C and 570°C. The average micro-hardness values at the joint interface were in the range of 125-175 HRB. A gradual decrease in the hardness occurred away from the interface, towards the Al-6063 side. The micro-hardness of Al-6063 decreased after a distance of 200 μm from the joint interface by 44-50% (from an average of 132 HRB to 58-67 HRB). The higher hardness of the post-welded Al-6063 was due to diffusion of Cu [19]. Furthermore, the joint formed at 570°C exhibited a significant increase in hardness within the Al-6063 alloy compared to the other joints made at lower temperatures. This is due to the maximum spread of the eutectic phase for the joint produced at 570°C. On the UNS S32304 side, the micro-hardness profile gave an average value of 266 HRB in a position 200 μm from the joint interface.

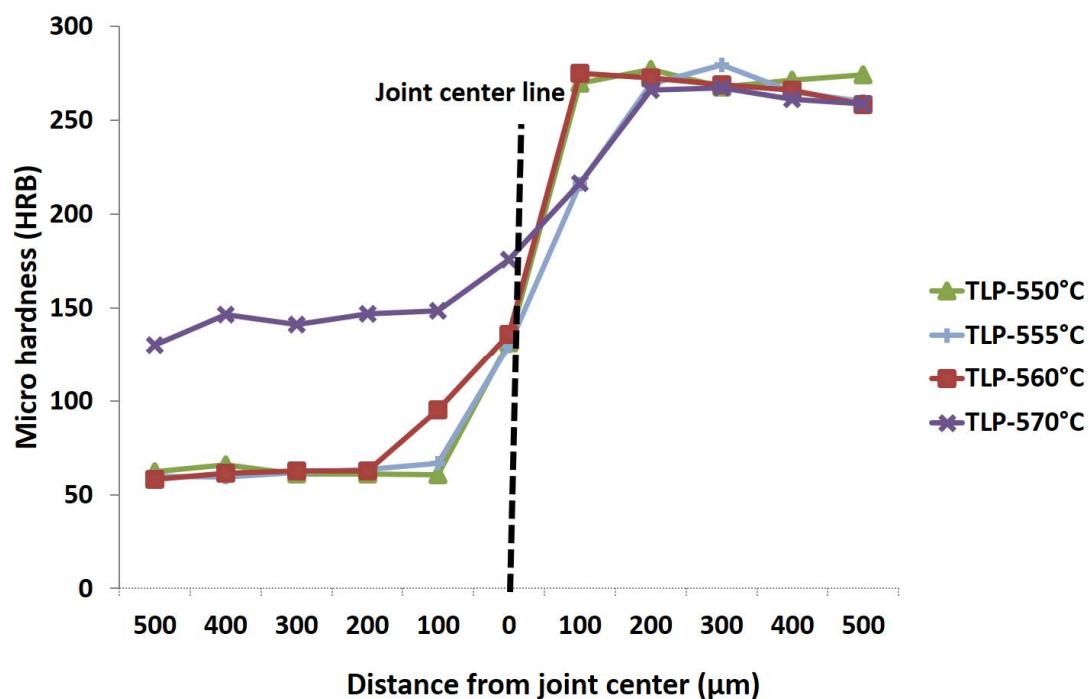


Fig. 9 Micro-hardness profile of joints made by TLP bonding at various temperatures. The Al-alloy and steel alloy to the left and right, respectively.

3.3 Corrosion resistance

The potentiodynamic polarization curves for TLP bonding joints as a function of bonding temperature are shown in Fig. 10. I_{corr} and E_{corr} were calculated by extrapolating the tafel plots of each test to the zero potential. Each test was repeated twice and found to be reproducible. As shown in Fig. 10, all the joints exposed to the same experimental condition, showed similar behavior in the polarization test. However, I_{corr} decreased at higher bonding temperature with the lowest I_{corr} value obtained for the joint produced at 570°C. It was found that the thickness of the reaction zone had a direct relation with the bonding temperature (See Fig. 4). Therefore, the joint produced at 570°C formed the thickest reaction zone compared to other joints formed at lower temperatures. According to the findings in [21], the denser the reaction zone, the higher the corrosion resistance of the joint. The present polarization curves in Fig. 10 clearly confirmed

these observations. The shift in curves to a more positive potential with increasing bonding temperature from 550°C to 570°C indicated a better corrosion resistance of the joint.

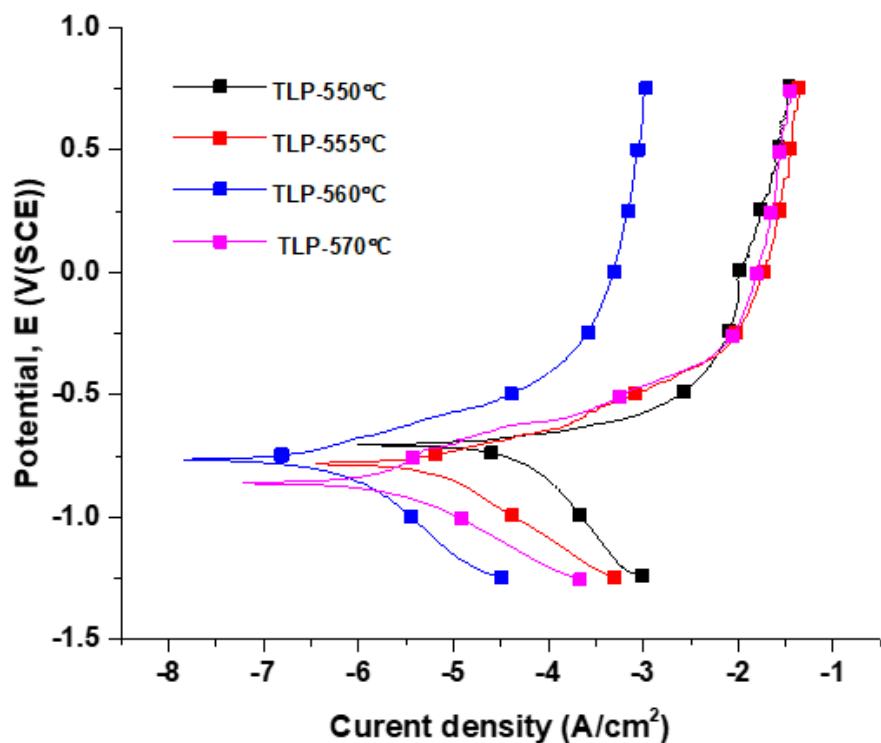


Fig. 10 Comparison of polarization curves for TLP bonding joints produced at various temperatures.

4. Conclusions

Transient liquid phase bonding of Al-6063 to UNS S32304 was studied using a thin Cu interlayer. The effects of the Cu interlayer and bonding temperature (550°C, 555°C, 560°C and 570°C) on the microstructure, corrosion resistance and micro-hardness of the resulting joints were investigated.

- The resultant area of TLP bonding consisted of three distinct zones including base metal, reaction zone and diffusion affected zone. Cu diffused into the Al alloy and formed an eutectic phase. However, no reaction was observed on the UNS S32304 side.

- As the bonding temperature increased from 550°C to 570°C, the thickness of the reaction zone increased by over 100%.
- Although voids and intermetallic compounds (Al_2Cu_1) were found at the interface, a TLP joint was produced successfully at 570°C.
- Employing Cu foil as an interlayer suppressed the formation of Fe-Al intermetallics.
- Hardness was increased on the Al-6063 side as a result of Cu diffusion. However, changes in hardness for the UNS S32304 steel was negligible.

Acknowledgements

This work was funded by Qatar Aluminium (Qatalum). The authors would also like to acknowledge the technical staff at Hydro Research and Technology Development Center in Norway for their support.

References

1. Muralimohan, C.H; Haribabu, S; Reddy, Y.H; Muthupandi, V.; Sivaprasad. Evaluation of Microstructures and Mechanical Properties of Dissimilar Materials by Friction Welding. *Procedia Mater. Sci.* **2014**, *5*, 1107-1113. DOI: <http://dx.doi.org/10.1016/j.mspro.2014.07.404>.
2. Torbati, A.M; Miranda, R.M; Quintino, L; Williams, S; Yapp, D. Optimization procedures for GMAW of bimetal pipes. *J. Mater. Process. Technol.* **2011**, *211*, 1112-1116. DOI: <http://dx.doi.org/10.1016/j.jmatprotec.2011.01.013>
3. Kouadri-David, A. Study of metallurgic and mechanical properties of laser welded heterogeneous joints between DP600 galvanised steel and aluminium 6082. *Mater. Des.* **2014**, *54*, 184-195. DOI: <http://dx.doi.org/10.1016/j.matdes.2013.07.093>.
4. Martinsen, K; Hu, S.J; Carlson, B.E. Joining of dissimilar materials. *CIRP Annu. Manuf. Techn* **2015**, *64*, 679-699. DOI: <http://dx.doi.org/10.1016/j.cirp.2015.05.006>.
5. Jiangwei, R; Yajiang, L; Tao, F. Microstructure characteristics in the interface zone of Ti/Al diffusion bonding. *Mater. Lett* **2002**, *56*, 647-652. DOI: [http://dx.doi.org/10.1016/S0167-577X\(02\)00570-0](http://dx.doi.org/10.1016/S0167-577X(02)00570-0).
6. Sammaiah, P; Suresh, A; Tagore, G.R.N. Mechanical properties of friction welded 6063 aluminum alloy and austenitic stainless steel. *J. Mater. Sci.* **2010**, *45*, 5512-5521. DOI: [10.1007/s10853-010-4609-y](https://doi.org/10.1007/s10853-010-4609-y).
7. Jafarian, M; Khodabandeh, A; Manafi, S. Evaluation of diffusion welding of 6061 aluminum and AZ31 magnesium alloys without using an interlayer. *Mater. Des.* **2015**, *65*, 160-164. DOI: <http://dx.doi.org/10.1016/j.matdes.2014.09.020>
8. Atabaki, M.M; Wati, J.N; Idris, J. Transient liquid phase diffusion brazing of stainless steel 304. *Weld. J.* **2013**, *92*, 57-63
9. Qiu, R; Shi, H; Zhang, K; Tu, Y; Iwamoto, C; Satonaka, S. Interfacial characterization of joint between mild steel and aluminum alloy welded by resistance spot welding. *Mater. Charact.* **2010**, *61*, 684-688. DOI: <http://dx.doi.org/10.1016/j.matchar.2010.03.015>
10. Guo, W; Zhao, X; Song, M; Fing, J; Yang, B. Diffusion bonding of Ti-6 Al-4 V to QAl 10-3-1. 5 with Ni/Cu interlayers. *J. Mater. Sci. Technol.* **2006**, *22*, 817-820.
11. Na, H; Kim, J; Jeong, B; Kang, C. Effect of brazing conditions on the microstructure and mechanical properties of duplex stainless steels to Cr-Cu alloy with Cu-base insert metal. *Met. Mater. Int.* **2007**, *13*, 511-515. DOI: [10.1007/bf03027911](https://doi.org/10.1007/bf03027911).
12. Wu, M-F; Si, N-C; Chen J. Contact reactive brazing of Al alloy/Cu/stainless steel joints and dissolution behaviors of interlayer. *Trans. Nonferr. Met.* **2011**, *21*, 1035-1039. DOI: [http://dx.doi.org/10.1016/S1003-6326\(11\)60818-2](http://dx.doi.org/10.1016/S1003-6326(11)60818-2).
13. Watanabe, T; Doi, Y; Yanagisawa, A; Konuma, S. Resistance Spot Welding of Mild Steel to Al-Mg Alloy. *Q. J. Jpn. Weld. Soc.* **2005**, *23*, 491-495. DOI: [10.2207/qjjws.23.491](https://doi.org/10.2207/qjjws.23.491).
14. Sun, J; Yan, Q; Gao, W; Huang, J. Investigation of laser welding on butt joints of Al/steel dissimilar materials. *Mater. Des.* **2015**, *83*, 120-128. DOI: <http://dx.doi.org/10.1016/j.matdes.2015.05.069>.
15. Orio, R.L.D. Electromigration modeling and simulation Technical University of Vienna, Vienna, 2010.
16. Suzuki, A; Mishin, Y. Atomic mechanisms of grain boundary diffusion: Low versus high temperatures. *J. Mater. Sci.* **2005**, *40*, 3155-3161. DOI: [10.1007/s10853-005-2678-0](https://doi.org/10.1007/s10853-005-2678-0).

17. Padron, T; Khan, T.I; Kabir MJ. Modelling the transient liquid phase bonding behaviour of a duplex stainless steel using copper interlayers. *Mater. Sci Eng A.* **2004**, 385, 220-228. DOI: <http://dx.doi.org/10.1016/j.msea.2004.06.034>.
18. Roberts, G.A; Krauss, G; Kennedy, R.L; *Tool Steels*, 5th ed.; ASM International, 1998.
19. Porter, D.L; Totemeier, T.C, Mechanical properties of metals and alloys. In *Smithells Metals Reference Book*, 8th ed., Gale, W.F; Totemeier, T.C, Butterworth-Heinemann: Oxford, 2004, pp.1-162.
20. Jin, Y; Khan, T. Effect of bonding time on microstructure and mechanical properties of transient liquid phase bonded magnesium AZ31 alloy. *Mater. Des.* **2012**, 38, 32-37.
21. Khan, T.I; Kabir, M.J; Bulpett R. Effect of transient liquid-phase bonding variables on the properties of a micro-duplex stainless steel. *Mater. Sci Eng A.* **2004**, 372, 290-295. DOI: <http://dx.doi.org/10.1016/j.msea.2004.01.023>.