Effects of processing parameters on the corrosion properties of dissimilar Friction Stir Welds of aluminium and copper

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Abstract

This paper reports the influence of friction stir welding processing parameters on dissimilar joints conducted between AA5754 aluminium alloy and C11000 copper. The welds were produced by varying the rotational speed from 600 to 1200 rpm and the feed rate from 50 to 300 mm/min. The resulting microstructure and the corrosion properties of the welds produced were studied. It was found that the joint interfacial regions of the welds were characterised by interlayers of aluminium and copper. The corrosion tests conducted revealed that the corrosion resistance of the welds improved as the rotational speed increased. The corrosion rates of the base metals compared to the welds improved for Cu and decreased slightly for aluminium. The lowest corrosion rates were obtained at welds produced at rotational speed of 950 rpm and feed rate of 300 mm/min.

Keywords: Aluminium alloy, Copper, Corrosion, Friction stir weld, Processing parameters.

1. Introduction

It is estimated that corrosion destroys one quarter of the world's annual steel production, which corresponds to about 150 million tons per year, or 5 tons per second **[1]**. Corrosion is not limited to steel but affects other materials used in various applications especially in welded joints. Corrosion is known to destroy a material or degrades its functional properties, rendering it unsuitable for the intended use **[1]**. Generally, the durability and the life time of welds, installations, machines and devices are critically dependent on their corrosion rate and wear resistance. Welded joints are specifically susceptible to corrosion when exposed to the environment and most especially dissimilar welds.

Friction Stir Welding (FSW) process employed in this study to join aluminium to copper is a solid state welding technique invented by Dr W. M. Thomas of The Welding Institute (TWI), United Kingdom in 1991 **[2]**. FSW is a continuous process that involves plunging a portion of a specially shaped rotating tool between the butting faces of the joint. A schematic of the process is presented in **Fig. 1**. The relative motion between the tool and the substrate generates frictional heat that creates a plasticized region around the immersed portion of the tool. The tool is moved relatively along the joint line, forcing the plasticized material to coalesce behind the tool to form a solid–phase joint **[3]**.

The resulting microstructures of friction stir welds are as described by Threadgill **[4]**. He identified and described the different zones as follows: The base metal (BM); which is the material remote from the weld that has not been deformed. It is not affected by the heat in terms of microstructure or the mechanical properties. The Heat Affected Zone (HAZ) which is a region, which lies closer to the weld centre, the material has experienced a thermal cycle that has modified the microstructure and/or the mechanical properties. However, no plastic deformation has occurred in this area. The Thermo Mechanically Affected Zone (TMAZ) which is a region in which the FSW tool has plastically deformed the material at the weld interface and lastly, the weld nugget which is the fully recrystallized area, sometimes called the Stir Zone (SZ) or Stir Nugget (SN), it refers to the zone previously occupied by the tool pin during FSW **[4]**.

The benefits of this technology include: low distortion, greater weld strength compared to the fusion welding process, little or no porosity, no filler metals, little or no post-weld repair, no solidification cracking, no welding fumes or gases, improved corrosion resistance, and lower cost in production applications **[5-9]**. Because of the many demonstrated advantages of FSW over the fusion welding techniques, the commercialization of FSW is proceeding at a rapid pace. The FSW of aluminium and its alloys has been commercialized **[10]**; and recent research interest is focused on joining dissimilar materials such as aluminium and copper. Components consisting of aluminium and copper possess the beneficial properties of both. Aluminium is mainly required for its low cost, high corrosion resistance and high strength to weight ratio while copper is mainly used for its superior electrical conductivity and its high thermal expansion. Such applications include bus-bars, switchgears and heat sinks, and many other applications are being developed. By successfully joining these metals with superior corrosion resistance, the superior properties of both materials can be utilized in many applications requiring a combination of these properties. Friction stir welds of aluminium and copper being a dissimilar joint is susceptible to galvanic or bimetallic corrosion in which corrosion can result from the formation of an electrochemical cell between the two metals joined and the corrosion of the less noble metal is thus accelerated. Published literature in this regard include research study by Surekha *et al.*, **[11]** on the effect of processing parameters on the corrosion behavior of friction stir processed AA 2219 aluminium alloy. It was found that the resistance to corrosion increases as the rotational speed increases in the processed aluminium samples. This is due to the dissolution of the $CuAl_2$ particles during the friction stir processing which reduces the number of sites available for galvanic coupling and hence increases the corrosion resistance. Further study was conducted by Prasad Rao *et al.*, **[12]** on the effect of friction stir processing on the corrosion resistance of aluminium–copper alloy gas tungsten arc welds. It was found that the friction stir processing improved the corrosion resistance of the welds. Fusion welds of this grade of aluminium alloy are known to suffer from poor corrosion resistance due to the uneven distribution of copper in the welds which produces large differences in the electrochemical potentials **[13]**. $AlCu_2$ is the major intermetallic compound found, which imparts greater strength in this alloy but decreases the corrosion resistance. This is due to the formation of galvanic cells between the noble $AlCu_2$ and the aluminium matrix **[13]**. To improve the corrosion resistance, it is necessary to create a uniform level of copper in the weld. Other studies on corrosion properties of friction stir welds include a report by Paglia and Buchheit **[14]** on the corrosion properties of friction stir welds of 7075-O aluminium alloy and they found that the welds are susceptible to intergranular corrosion. They however suggested that short-term post-weld heat treatments with temperatures similar to the welding temperatures can be used to modify the microstructure and improve the corrosion resistance of the welds. The effect of welding parameters on the corrosion behavior of friction stir welded AA2024–T351 was also conducted by Jariyaboon *et al.*, **[10]**. They found that the rotational speed has the greatest influence on the corrosion sensitivity on the weld cross sections. It was concluded that for low rotational speeds, the corrosion attack is in the nugget region due to the significant increase in the anodic reactivity in this region. For higher rotational speeds, the corrosion attack is in the HAZ region owing to the presence of sensitized grain boundaries in this region. Bousquet *et al.*, **[15]** conducted a research study on the relationship between the microstructure, microhardness and corrosion sensitivity of friction stir welded joints of AA 2024-T3 and found that the HAZ close to the TMAZ is the region most sensitive to intergranular corrosion because of the presence of the continuous lines of intergranular precipitates at the grain boundaries and the pitting corrosion observed was due to the presence of intermetallic particles at such regions. However, the majority of these studies are limited to joining similar materials especially aluminium and its alloys.

In view of the foregoing, concerted efforts are geared towards optimizing the processing parameters to produce metallurgically sound joints of aluminium and copper using FSW **[16-19]** which will ultimately lead to its commercialization. It is very important to have an insight into the corrosion properties of such joints in order to produce joints that will meet the service requirements and be guided accordingly. To the authors' knowledge, there is no published literature on the corrosion properties of friction stir welds of aluminium and copper. The main objective of this paper therefore is to report on the corrosion properties of dissimilar friction stir welds of aluminium and copper produced at different process parameter combinations.

2. Materials and methods

2.1 Materials preparation

Friction stir welds between aluminium alloy AA5754 and copper C11000 having 3.175 mm thicknesses were produced using an Intelligent Stir Welding for Industry and Research Process Development System (I-STIR PDS) platform. The chemical compositions of the alloys used are provided in **Table 1**. The welds were produced using an 18 mm shoulder diameter tool with a tool pin diameter of 5 mm. The copper sheet was placed at the advancing side while the tool pin was plunged in the aluminium alloy and made to touch copper during the welding process. This is an optimized tool displacement setting as reported by Akinlabi *et al.*, **[20]**. Rotational speeds of 600, 950 and 1200 rpm were employed which represent a low, medium and high speed settings, respectively, while 50, 150 and 300 mm/min were the transverse feed rates considered, which also represents a low, medium and high feed rate settings, respectively. The weld matrix is presented in **Table 2**.

An optical microscope (Olympus BX51M) and scanning electron microscope (VEGA 3) were used for the microstructural evaluation of the joint interfaces. A weld length of 160 mm was produced for each setting and the samples for corrosion testing were cut at 50 mm length from the weld start in a transverse direction. The aluminium alloy samples were etched with Flicks reagent and the copper etched with a solution of 25 ml distilled water, 25 ml ammonia water and 15 ml hydrogen peroxide. Vickers microhardness profiles were measured along the cross sections of the welds with a load of 200 g and a dwell time of 10 seconds, using an MH3 microhardness indenter.

2.2 Electrochemical corrosion testing

Potentiodynamic polarization techniques were used to study the corrosion behavior of the welded joints. The corrosion experiments were carried out using AUTOLAB PGSTAT30 with GPES electrochemical software. All the experiments were carried out using a three-electrode corrosion cell set-up with saturated Ag/AgCl as reference and platinum rod as counter electrode. The corrosion tests were conducted on both the top surface and the cross-sectional areas of the welds. The samples were cold mounted in polyester resin. The areas exposed to the electrolyte were 2.25 cm^2 for the weld surface and 0.45 cm^2 for the cross-sections of the welded zone. All the tests were conducted at room temperature (25 ± 2 ^oC). The electrolyte used was 3.5% NaCl. Potentiodynamic polarization measurements were carried out using a scan rate of 0.167 mV/s at a potential initiated at -150 mV to +1500 mV versus corrosion potential. Before starting the polarization scan, the specimens were cathodically polarized at -1000 mV for 5 minutes followed by stabilization for about 1 hr. In all cases, triplicate experiments were carried out to ensure reproducibility. Corroded surfaces were observed using ultra high resolution scanning electron microscope (FE-SEM JSM 7600F).

3. Results and discussion

3.1 Structure of welded samples

Fig. 2 shows the optical micrographs of the parent materials – aluminium (AA5754) and copper (C11000). The microstructure of the aluminium alloy consisted of fairly elongated grains while the grains of the copper were equiaxed. **Fig. 3** shows the surface appearances of the friction stir welded samples as taken from 75 mm of the welded length. The weld appearances were typical of friction stir welds and without visual defects. The micrograph of an interfacial region of a typical weld produced at 950 rpm and 150 mm/min is presented in **Fig. 4**. Under these welding conditions, optimum mechanical properties could be obtained **[15]**. It was observed that the joint interface of the weld was characterized with an onion ring structure indicating good material flow and good mixing of both materials joined [**21**]. It can be inferred that this region has undergone dynamic recrystallization during the FSW process.

The microhardness profile of the weld produced at 950 rpm and 150 mm/min super imposed on the micrograph is presented in **Fig. 5**. The average microhardness of the aluminium and copper parent materials used in this study were 70 and 85 HV, respectively. A constant hardness of approximately 70 HV was observed in the aluminium side until 1 mm to the centre where the hardness increased sharply to a peak of about 180 HV. This is due to the transition from aluminium material to an intermetallic (Al_2Cu) present in the copper material. The high hardness value of 271 HV measured at 3 mm into copper corresponds to the presence of an intermetallic compound (Al_4Cu_9) in the weld as was confirmed by the XRD results which has been reported elsewhere **[16]**.

3.2 Effects of friction stir welding parameters on electrochemical corrosion behavior

Typical electrochemical corrosion behavior of FSW of aluminium alloy and copper in 3.5% NaCl is shown in **Fig.6**. **Fig.6a** shows the corrosion behavior on the sample surface and **Fig.6b** the cross section. Both the surface and the cross section samples indicated similar polarization curves with no stable passivity features. At a potential of about 0.9 V versus Ag/AgCl, the current density of the surface samples first decreased as the applied potential was increased, indicating the formation of surface film on the sample surface, and then increased sharply with a slight increase in the applied potential (**Fig.6a**). The decrease in the current density was absent in the cross section samples (**Fig.6b**). The reason could be due to the presence of high concentration of aluminium in the surface samples than the cross section samples resulting from the fact that the tool pin was plunged in the aluminium alloy and made to touch the copper during the welding process. Aluminium with a lower melting point in this regard became plasticized and then got mixed with the copper due to the stirring action of the tool during the FSW. The corrosion potentials of the surface samples were about the same (-1.017 $V_{Ag/AgCl}$) whereas the corrosion potentials of the cross section samples varied slightly from -1.031 to -0.704 $V_{Ag/AgCl}$. The current densities of both surface and cross section samples after corrosion potential increased sharply which indicates the possibility of pitting corrosion occurring. The relatively higher corrosion rate of surface sample produced at 950 rpm and 50 mm/min could be as a result of the high concentration of Cu present at the surface of this sample which increased the galvanic interaction. The high concentration of Cu at the welded joint was confirmed by XRD results indicating the presence of both Al_2Cu and Al_4Cu_9. At higher feed rate (300 mm/min) only Al_2Cu was present **[24]**.

The corrosion rates of both the surface and the cross section samples were calculated and the results are presented in **Fig.7**. In the calculation of the corrosion rate, the equivalent weights of the specimen were used by determining the chemical compositions of the welded zones. It is observed that as the rotational speed increased, the corrosion rate decreased for both the surface and the cross section samples. Thus, the rotational speed has a direct relationship with the corrosion rate. In all instances, there is a significant reduction in the corrosion rate when the rotational speed was increased from 600 rpm to 950 rpm. From 950 rpm to 1200 rpm, the reduction in the corrosion rate was minimal. There was no strong correlation between the feed rate and the corrosion rate. It is interesting to note that for a specified rotational speed, the lowest corrosion rate was observed at the maximum feed rate employed (i.e. 300 mm/min) for most of the samples. This can be attributed to the fact that the welds produced at the highest feed rate of 300 mm/min were conducted with less heat input which has resulted in the less mixing of both materials compared to the welds produced at the 50 and 150 mm/min. Hence, in this regard, less heat input into the welds resulting in excellent characteristics of the joint interface is desired. Additionally, at high rotational speed, Al_2Cu phase was mainly identified in the welded zones whereas at low rotational speed Al_4Cu_9 was identified. It has been reported by Chen and Hwang **[22]** that the activation energy of Al_4Cu_9 is higher than that of Al_2Cu. This implies that the galvanic interaction between these intermetallics and the base metals would be higher for Al_4Cu_9 than Al_2Cu; hence the observed corrosion behavior.

The surface corrosion rates of the aluminium alloy and pure copper were calculated to be 0.00112 mm/yr and 0.0367 mm/yr, respectively; the corresponding corrosion rates of the cross section samples were 0.00129 mm/yr and 0.048 mm/yr, respectively. It could be observed that the corrosion resistance of copper in 3.5% NaCl was enhanced. This behavior has been reported by Wharton and Stokes **[23]** where Al forms a film of hydrated oxide/hydroxide.

Figs. 8 and **9** show the microstructures of the surface and cross section of the FSW samples after corrosion, respectively. Localized corrosion could be observed on the samples after corrosion testing in 3.5% NaCl. It is reported **[14]** that the chemistry of Al alloy has significant effect on the corrosion behavior of the welded piece. Stress corrosion cracking was intense at the surfaces compared to the cross section samples. This is because of the presence of high concentration of Al on the surface samples. Aluminium alloy is susceptible to stress corrosion cracking; pitting and intergranular corrosion after the alloy microstructure has been sensitized. It could be said that the attack on the friction stir welded samples initiates as pits and propagate as stress corrosion cracking. This agrees with the polarization results presented in **Fig. 6**. At the cross sections (**Fig.9**), intergranular attack was observed at the nuggets.

4. Conclusion

The corrosion behaviors of friction stir welds of Al-Cu joints have been characterized. Rotational speeds and feed rates were varied and the corrosion behavior studied. Microstructural evaluation of the interface revealed the formation of onion rings which are an excellent characteristic of friction stir welds. The corrosion results indicated that the transverse feed rate has little or no effect on the rate of corrosion. However, the electrochemical corrosion resistance of the welded Al-Cu alloys is enhanced as the rotational speed is increased due to the presence of Al_2Cu intermetallic phase at higher rotational speed as compared to Al_4Cu_9 and lower rotational speed. Optimum corrosion resistance was obtained for welds produced at 950 rpm at 300 mm/min which was correlated to the low heat input in these welds.

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List of Figures

Table 1: Chemical composition of Al and Cu materials

	Si	Pb	Mg	Cr	Ti	Zn	Al	Cu
AA5754	0.40	0.80	3.50	0.30	0.15	0.50	96.10	0.03
C11000	0.0005	0.0005	0.0001	0.0003	0.0002	0.009	0.001	99.859

Table 2: Weld matrix of investigated materials

Sample ID	Rotational speed (rpm)	Feed rate (mm/min)
A1	600	50
A2	600	150
A4	600	300
C1	950	50
C2	950	150
C4	950	300
L1	1200	50
L2	1200	150
L4	1200	300

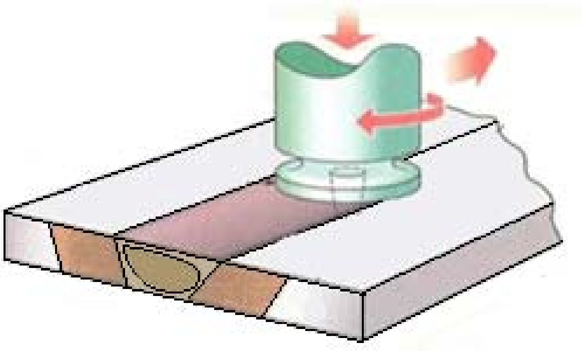


Figure 1

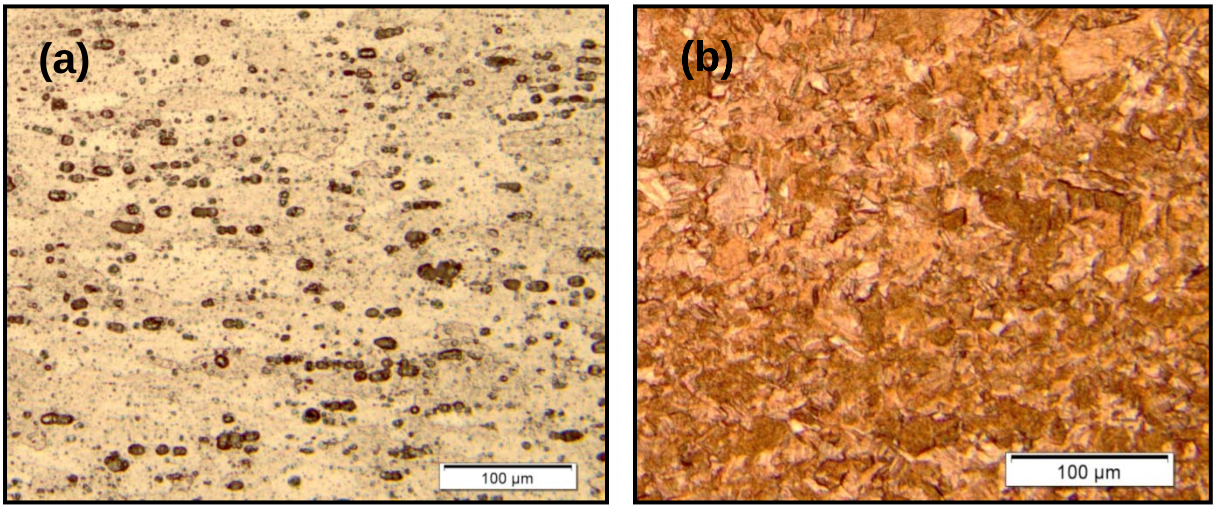


Figure 2

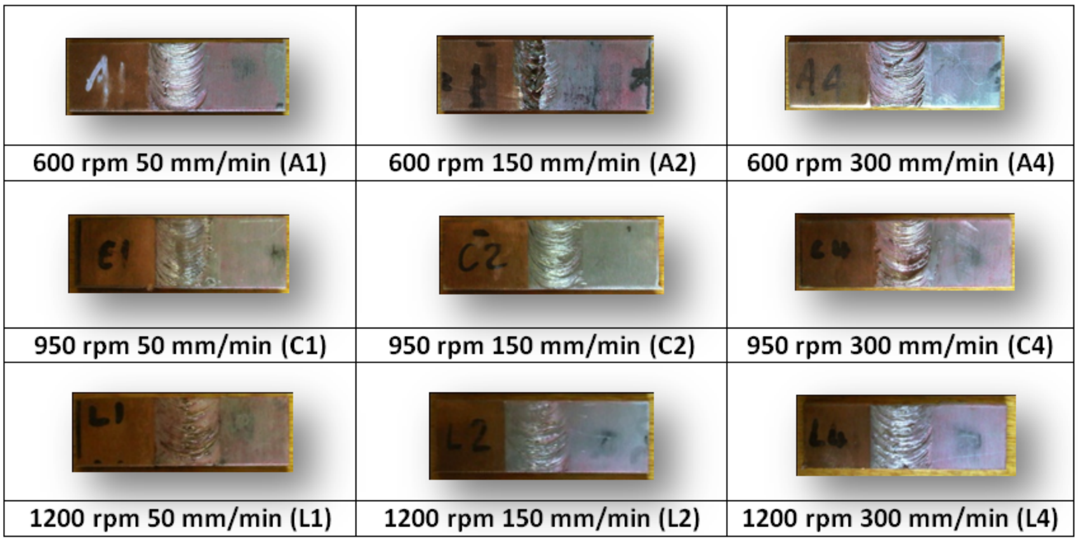


Figure 3

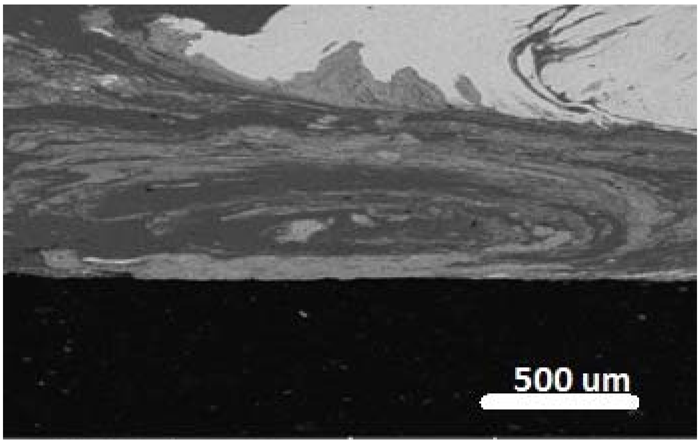


Figure 4

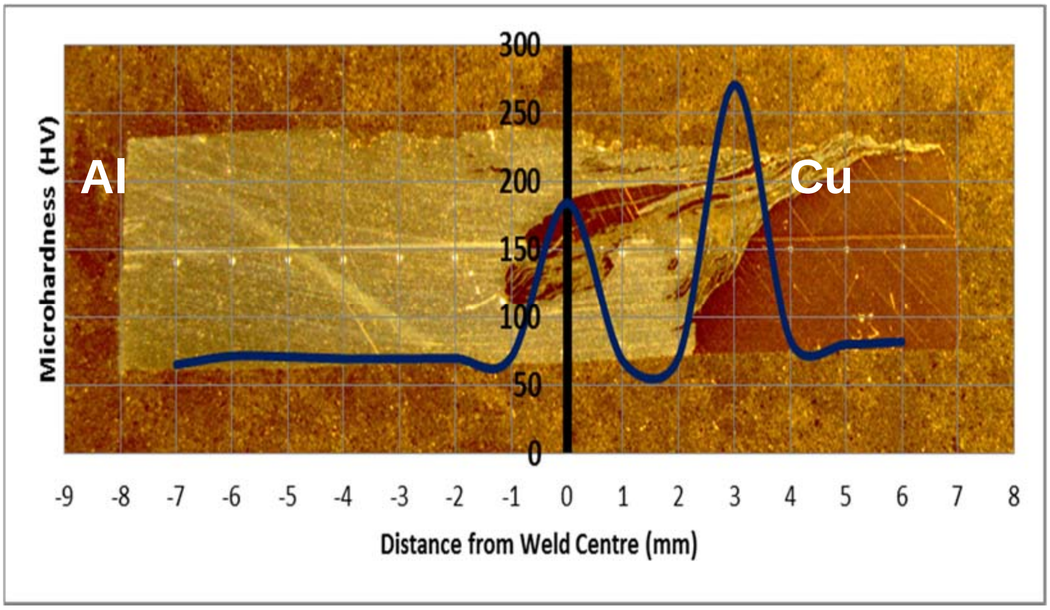
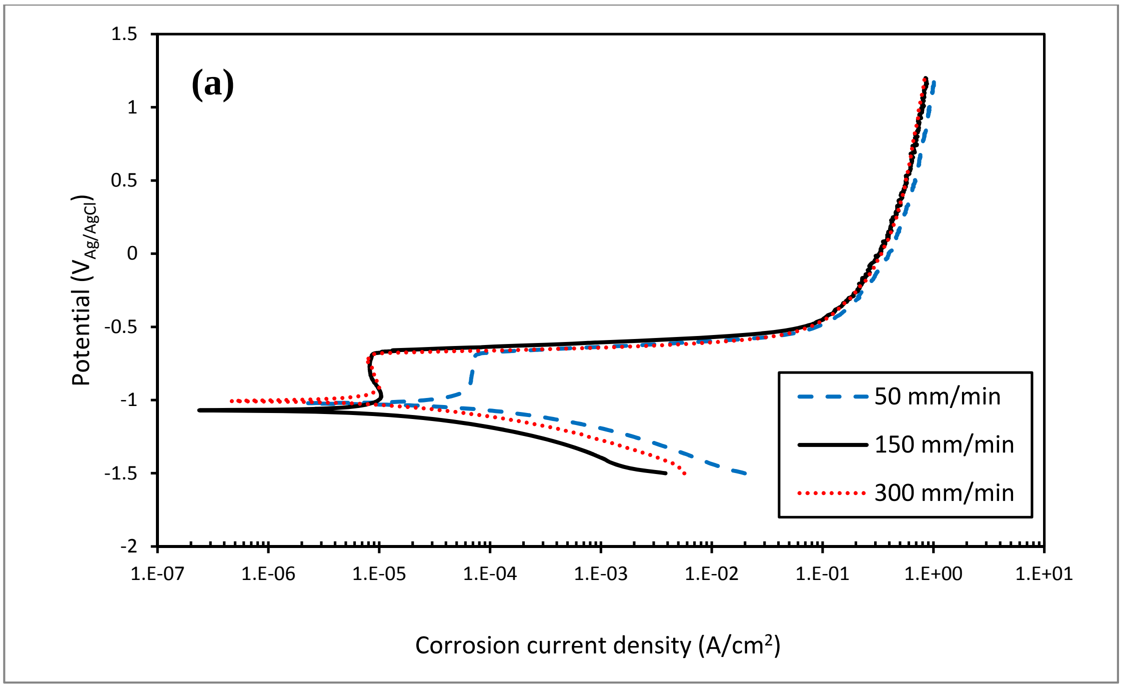

300
250
200
150
100
50
0
Microhardness (HV)
Al
Cu
-9
-8
-7
-6
-5
-4
-3
-2
-1
0
1
2
3
4
5
6
7
8
Distance from Weld Centre (mm)

Figure 5


(a)
Potential (V$_{Ag/AgCl}$)
1.5
1
0.5
0
-0.5
-1
-1.5
-2
1.E-07
1.E-06
1.E-05
1.E-04
1.E-03
1.E-02
1.E-01
1.E+00
1.E+01
Corrosion current density (A/cm^2)
50 mm/min
150 mm/min
300 mm/min

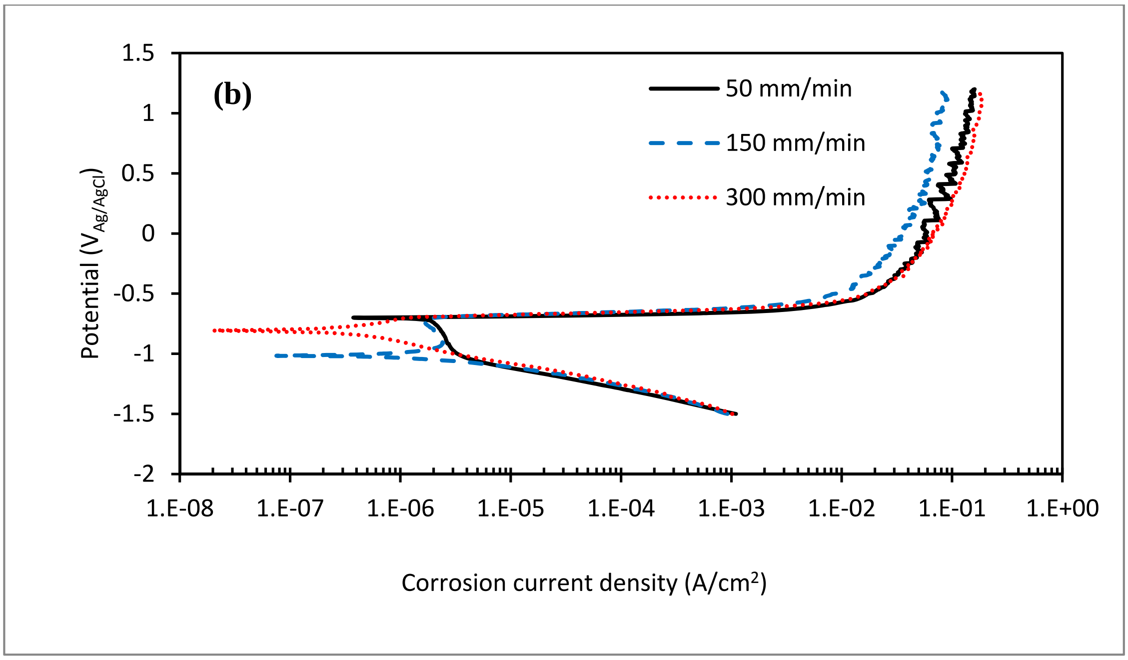

(b)
Potential (V$_{Ag/AgCl}$)
1.5
1
0.5
0
-0.5
-1
-1.5
-2
1.E-08
1.E-07
1.E-06
1.E-05
1.E-04
1.E-03
1.E-02
1.E-01
1.E+00
Corrosion current density (A/cm^2)
50 mm/min
150 mm/min
300 mm/min

Figure 6

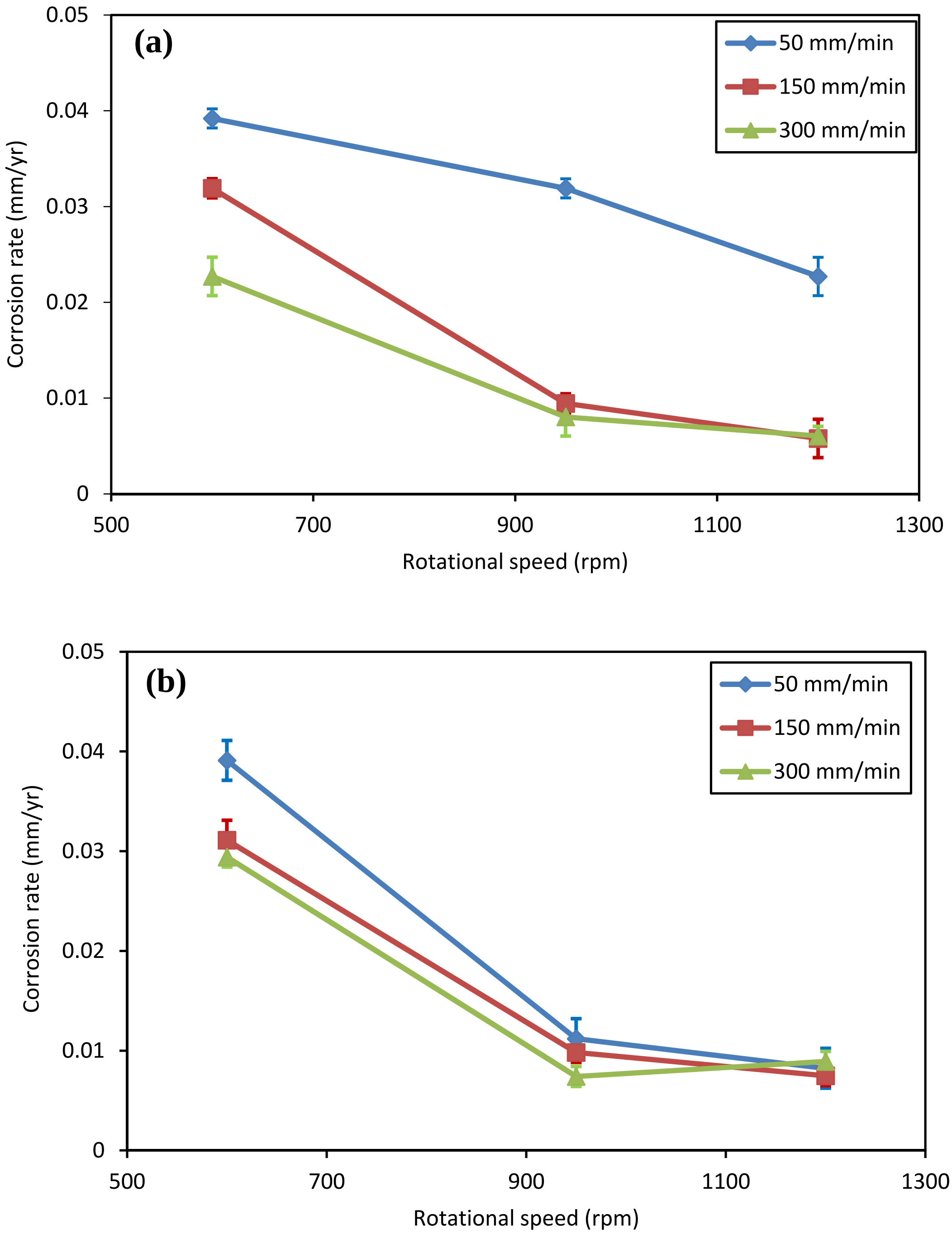
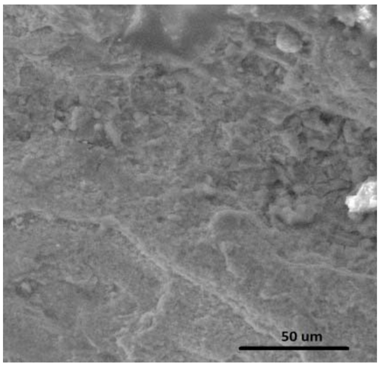
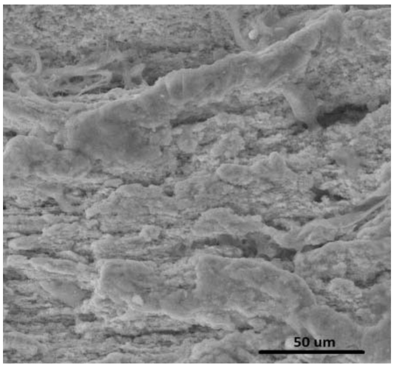

(a)
50 mm/min
150 mm/min
300 mm/min
0.05
0.04
0.03
0.02
0.01
0
Corrosion rate (mm/yr)
500
700
900
1100
1300
Rotational speed (rpm)
(b)
50 mm/min
150 mm/min
300 mm/min
0.05
0.04
0.03
0.02
0.01
0
Corrosion rate (mm/yr)
500
700
900
1100
1300
Rotational speed (rpm)

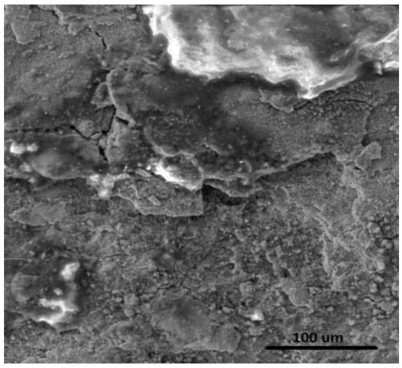
Figure 7



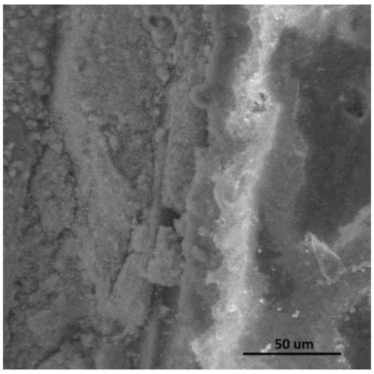
A1 (600 rpm : 50 mm/min)



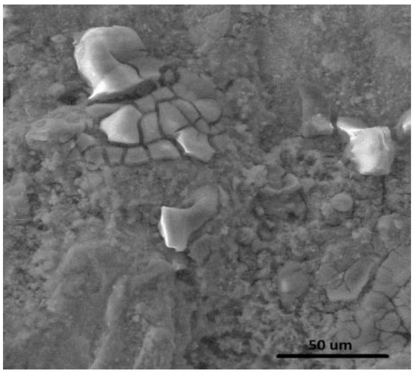
C1 (950 rpm : 50 mm/min)



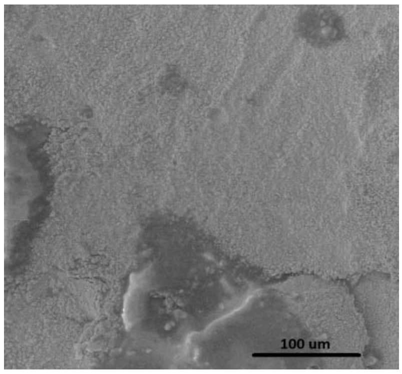
L1 (1200 rpm : 50 mm/min)



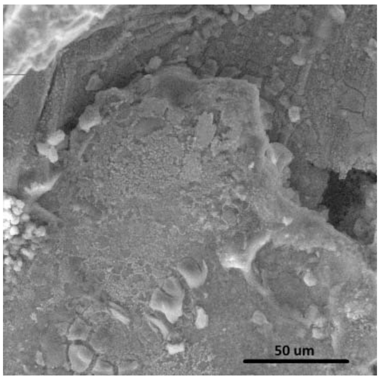
A2 (600 rpm : 150 mm/min)



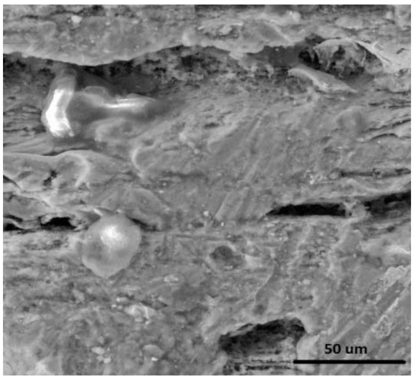
C2 (950 rpm : 150 mm/min)



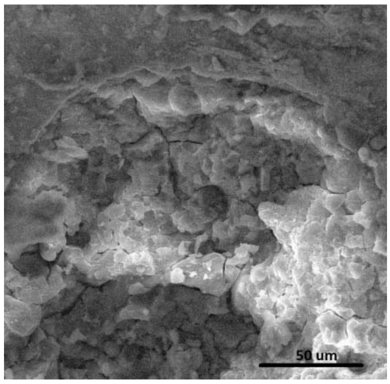
L2 (1200 rpm : 150 mm/min)



A4 (600 rpm : 300 mm/min)

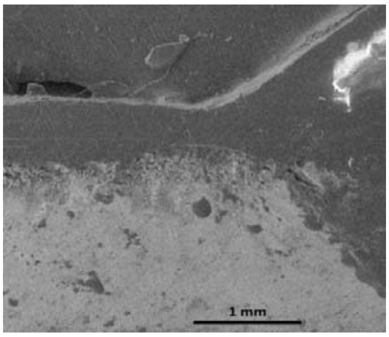


C4 (950 rpm : 300 mm/min)

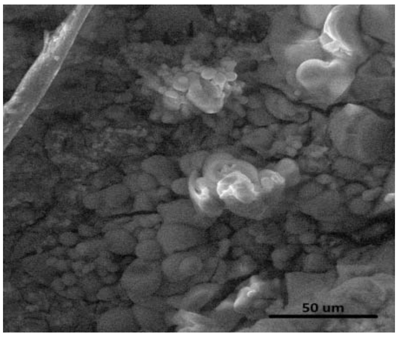


L4 (1200 rpm : 300 mm/min)

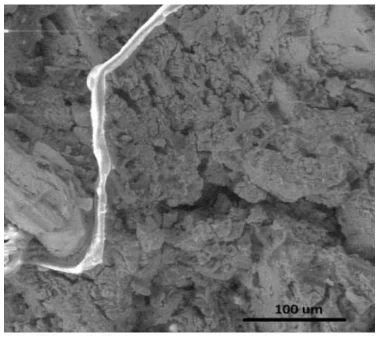
Figure 8



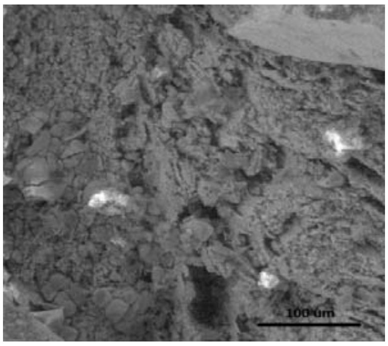
A1 (600 rpm : 50 mm/min)



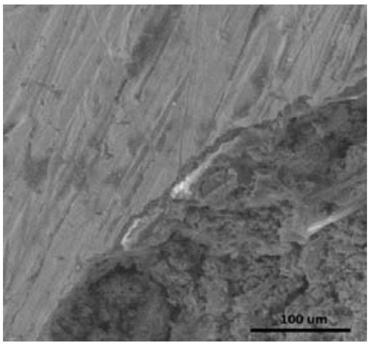
C1 (950 rpm : 50mm/min)



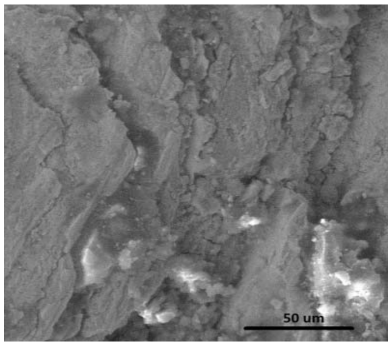
L1 (1200 rpm : 50 mm/min)



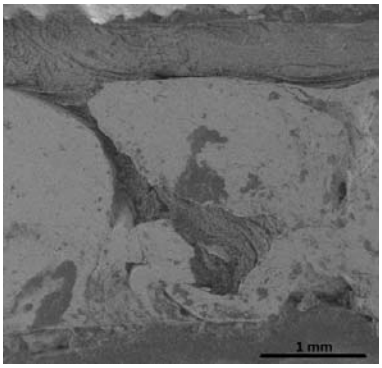
A2 (600 rpm: 150 mm/min)



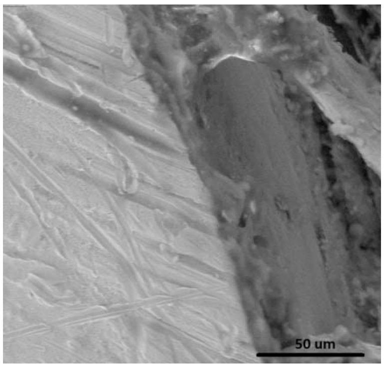
C2 (950 rpm : 150 mm/min)



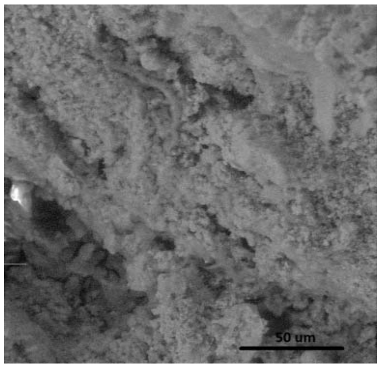
L2 (1200 rpm : 150 mm/min)



A4 (600 rpm : 300 mm/min)



C4 (950 rpm : 300 mm/min)



L4 (1200 rpm : 300 mm/min)

Figure 9