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Hybrid joints manufactured by ultrasound enhanced friction stir welding (USE-FSW) - corrosion properties

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Abstract. To realize lightweight structures of material combinations like aluminum/magnesium and aluminum/steel an Ultrasound Enhanced Friction Stir Welding (USE-FSW) process was used. This process has a beneficial influence on the resulting microstructure (elimination of the brittle intermetallic phase Al3Mg2 as coherent layer) and the mechanical properties (increased tensile strength) of Al/Mg-joints and was now also applied for Al/steel-hybrid joints. Besides the mechanical properties the corrosion properties of the hybrid joints may play a significant role concerning the later use of the hybrid materials. Therefore, the corrosion properties of various hybrid joints have been investigated by different methods. With the Scanning Kelvin Probe (SKP) Volta potential differences between the base alloys and the welded area were investigated in air. The two-dimensional color-plots illustrate not only the Volta potential differences between the different phases but also their oxidation properties in air during the measurement time. Electrochemical measurements (open circuit potential and potentiodynamic polarization) have been carried out for the investigation of the corrosion properties of the FSW and USE-FSW hybrid joints in 0.5 molar NaCl solution. A three electrode setup within a mini-cell was used to enable measurements on different areas of the joints. This allows to observe the corrosion activity of the base alloys and the nugget phase separately. Differences between Al/steel-hybrid joints processed with and without ultrasound enhancement are discussed and compared with Al/Mg-hybrids.

1. Introduction

The rapid development of transport applications suggests adaptive and hybrid structures. Therefore, the employment of lightweight materials in steel structures is of great interest. In this regard a very advantageous material combination is aluminum/steel. The established method of fusion welding may not be used for aluminum/steel because of the formation of brittle intermetallic (IM) phases which decrease the joint strength [1]. Due to this, pressure welding methods which are characterized by welding temperatures below the melting points of the materials to join are an interesting alternative with the aim to avoid the development of brittle phases in the welding zone. A well established welding method in this group is friction stir welding (FSW) [2-4]. The research work on FSW to produce aluminum-to-steel joints is summarized in a review article by Hussein et al. [5]. The relatively low process temperature of the FSW results in a decreased amount of brittle intermetallic phases compared to fusion welded joints but cannot avoid the formation of intermetallics completely. To
minimize this negative effect a hybrid welding method called “Ultrasound Enhanced Friction Stir Welding” (USE-FSW) was developed. In this process additional ultrasonic energy is transmitted in the welding parts during FSW with the aim to crack interlayers and to support the stirring in the joining area. This process was successfully applied on joints of aluminum and magnesium alloys [6, 7]. The combination of different metals in hybrid structures may cause corrosion problems in the welding area because of the formation of a galvanic couple which leads to severe corrosion of the less noble element. In the case of Al/Mg-hybrids an accelerated corrosion of Mg in chloride containing electrolyte was found [8, 9]. This paper will focus on the corrosion investigation of FSW and USE-FSW Al/steel joints. With the Scanning Kelvin Probe (SKP) Volta potential differences between the base alloys and the welded area were investigated in air. Electrochemical measurements (open circuit potential and potentiodynamic polarization) have been carried out for the investigation of the corrosion properties of the FSW and USE-FSW hybrid joints in 0.5 molar NaCl solution using a minicell. Differences between Al/steel-hybrid joints processed with and without ultrasound enhancement are discussed and compared with Al/Mg-hybrids.

2. Experimental

2.1 Materials
For the investigations the commercially available aluminum wrought alloy EN AW-6061 T6 (AlMg1SiCu) and the deep-drawing steel DC04 (1.0338) were used in a sheet geometry of 280 mm length, 100 mm width and 3 mm thickness. The chemical composition of the materials according to the manufacturer’s specifications is given in Table 1.

| Material  | Si  | Fe  | Cu  | Mn  | Mg  | Cr  | Zn  | Ti  | Al  |
|-----------|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| EN AW-6061 T6 | 0.64 | 0.51 | 0.21 | 0.14 | 0.89 | 0.15 | 0.04 | 0.05 | bal. |
| DC04      | 0.041 | 0.015 | 0.3 | 0.01 | 0.0077 | 0.49 | 0.0007 | 0.0037 | bal. |

2.2 Joining procedure
For the process of friction stir welding a universal four axis machining center DMU80T from DMG MORI got extended with a pneumatic clamping for the metal sheets as well as four load cells from Kistler, so that a force-controlled process-sequence was possible. A butt-joint configuration was chosen for the joints, whereas steel was placed on the advancing side and aluminum on the retreating side. The welding direction was perpendicular to the rolling direction of EN AW-6061 and parallel to the rolling direction of DC04. For the hybrid-joining method of USE-FSW the machining center was additionally equipped with a Lab VIEW-controlled ultrasonic roll seam module of Schunk Sonosystems, which runs synchronously and parallel to the tool. The instrument and the used joining parameters are described in detail in the papers of Thomä et al. [10, 11].

2.3 Corrosion investigations
Prior to all measurements the surface of the samples was ground with wet SiC-paper (up to P1200), rinsed with ethanol and dried to achieve comparable conditions for all samples. One method that gives space-resolved information about the corrosion properties of materials is the Scanning Kelvin Probe (SKP). The Kelvin Probe uses a contactless vibrating condensator technique which allows to measure the changes in the Volta potential or work function of electrons from metals, semi conductors or at phase boundaries of liquids. The method is described in detail in [12-14]. For our investigations of Volta potential differences between the Al alloy and steel and the formed phases after the FSW process a SKP-setup with height control (SKP_KM, Wicinski-Wicinski GbR) was used.
The measuring probe was a Cr-Ni-wire with a diameter of 100 μm. The Volta potential of the probe was not calibrated. Therefore the measured values are no absolute values and only Volta potential differences are discussed which also allow to draw first conclusions about the corrosion properties. Measurements were carried out in air with a relative humidity of about 33-35 %.

Electrochemical measurements have been carried out for the investigation of the corrosion properties of the FSW and USE-FSW Al/steel-joints in chloride containing electrolyte. A three electrode setup within a mini-cell (measurement area 0.071 cm$^2$) was used to enable measurements on different areas of the joints. The investigated samples were used as working electrode, a platinum electrode as counter electrode and a standard calomel electrode (+ 246 mV vs NHE) as reference electrode. The recorded values were converted to values against the normal hydrogen electrode (NHE).

The open circuit potential (OCP) measurements and the potentiodynamic polarization have been done with a Zahner IM 6 measurement unit. The open circuit potential was recorded for 60 min prior to the polarization experiment. The potentiodynamic curves were started -0.1 V relative to the open circuit potential and recorded up to +0.5 V relative to it with a scan rate of 1 mV/s. The electrochemical measurements were carried out in 0.5 molar NaCl solution.

3. Results and Discussion

3.1 SKP measurements

Figure 1 shows the measured Volta potential differences of the welding zones of an Al/steel FSW-joint (a) and an USE-FSW-joint (b) as two-dimensional colour-plots. The recording of the whole plot of one sample takes about 36 h. Therefore the plots illustrate not only the Volta potential differences between the different phases but also their oxidation properties in air during the measurement time. At the beginning of the measurements the Volta potential difference between the Al alloy and the steel is about 1.0 V. Both materials exhibit a shift of the Volta potential by about 0.2 to 0.3 V to more positive values within the measurement time due to surface oxidation processes, but the difference between the materials is nearly constant.

![Image of Volta-Potential maps of a FSW-joint (a) and an USE-FSW-joint (b) measured in air (relative humidity 33-35 %); time of each measurement: 36 h starting from the top of the plot.](image-url)

Figure 1. Volta-Potential maps of a FSW-joint (a) and an USE-FSW-joint (b) measured in air (relative humidity 33-35 %); time of each measurement: 36 h starting from the top of the plot.

The values of Volta potential differences of the main phases of FSW- and USE-FSW-joints are nearly the same. Differences between the two joints are obvious directly at the welding area. The FSW-joint shows a 1-2 mm broad seam with Volta potentials that are 0.1 to 0.3 V more negative than the steel. In addition there is a broader area in the nugget region (Al side) with a potential which is 0.1 to 0.2 V
more positive than the Al alloy but considerably more negative than the steel. For the USE-FSW-joint the 1-2 mm seam was not observed. In the nugget area some kind of hook is visible which has a more positive Volta potential than the surrounding metal surface.

The Volta potential maps clearly show an influence of the ultrasound on the welding area of Al/steel hybrid joints but it is much less pronounced than in the case of the Al/Mg-hybrid joints [9]. This can be explained with the different joining parameters for the different joints. For the Al/Mg-hybrid joints the pin-offset was 1 mm to the Mg which results in a mechanical mixing of both materials during the process. In case of the Al/steel-hybrid joints, because of the high difference in the melting points of the materials, the pin only rotates within the Al with a distance of 0.3 mm to the faying surface of the steel. Therefore the influence of the ultrasound introduced via the steel sheet is less visible. Only the shoulder of the rotating tool has mechanical contact to the steel and may transport material from one side to the other.

The SKP measurements result in about 1 V more negative Volta potential values for the EN AW-6061 Al alloy compared to the DC04 steel. This is a similar difference to that measured between AZ80 Mg alloy and AC-48000 Al alloy where the Al alloy has shown the more positive values [9]. From that result one would expect that the EN AW-6061 Al alloy will show stronger corrosion in corrosive media than the steel particularly if both phases are in simultaneous contact to an electrolyte.

3.2 Open circuit potential (OCP)

The measurements in air are not directly transferable to that in a liquid electrolyte, as a comparison of the potential values for the steel and the Al alloy shows. During the SKP measurements in air the Volta potential of the steel was always about 1 V more positive than that of the Al alloy. The values change significantly in the sodium chloride electrolyte (figure 2).

The OCP of the EN AW-6061 alloy exhibits a shift from about -0.7 V to -0.47 V within the first 5 to 10 min and then stabilises around that value. The OCP of the steel declines continuously from about -0.23 V to -0.45 V within the first 10 min and then further to -0.52 V. After 15 to 20 min the OCP value of the DC04 steel is more negative than that of the EN AW-6061 Al alloy. This behaviour is different to that of the Al/Mg-hybrid joints where an OCP difference of 0.9 V was measured between the base materials in the sodium chloride electrolyte similar to the Volta potential difference in air (SKP measurement) [9].

The nugget area and the Al/steel transitional area show nearly constant OCP values immediately after the immersion in the electrolyte. The values are similar to that of the EN AW-6061 Al alloy. The OCP values of the base materials and the nugget phase within the hybrid joints are independent of the joining conditions (with or without ultrasound enhancement) as a comparison of figures 2a) and 2b) shows. Only the OCP value of the Al/steel transitional area seems to depend on the processing conditions, because the value for the FSW-joint shows a little shift to more negative values (-0.49 V after 60 min). But this may also be an effect of the area ratio Al/steel in the measured area, because of the uneven weld seam and positioning of the measurement cell. The OCP value of the DC04 measured near the weld shows within the first minutes the same decline as the base material but then reaches an OCP value between the DC04 and the EN AW-6061 after 60 min.

After the immersion time of 60 minutes the OPC difference between the materials is only about 50 mV or less. Therefore, the risk of an enhanced corrosion caused by a galvanic element is much less pronounced than in the case of the Al/Mg-hybrid joints.
3.3 Potentiodynamic polarization

Figure 3 shows the results of the potentiodynamic polarization measurements on different positions of a FSW (figure 3a) and an USE-FSW (figure 3b) Al/steel-hybrid joint in 0.5 molar NaCl solution. The measurements were started 60 minutes after immersion at open circuit potential. The EN AW-6061 Al alloy exhibits a free corrosion potential ($E_{oc}$) around -0.46 V and a pitting potential around -0.42 V.
which is only slightly more positive than the free corrosion potential. The corrosion current density measured for the Al alloy at the $E_{\text{oc}}$ is around 1-2 μA/cm$^2$ indicating a low corrosion rate. At the pitting potential the current density increases immediately up to values of several hundred μA/cm$^2$.

![Figure 3](image)

**Figure 3.** Potentiodynamic polarization curves measured on different positions of a FSW-joint (a) and an USE-FSW-joint (b) in 0.5 molar NaCl solution.

The free corrosion potential of the DC04 steel is around -0.54 to -0.53 V. The polarization curve of the steel shows a continuous increase in the anodic region. The corrosion current density at the $E_{\text{oc}}$ is slightly higher than that of the EN AW-6061 Al alloy (2-3 μA/cm$^2$). The steel surface near the weld (figure 3b) only shows a small shift of the $E_{\text{oc}}$ otherwise the polarization curve is nearly identical to that of the DC04 base material (greater distance from the weld).

The free corrosion potential of the nugget area shows during the potentiodynamic polarization a small shift to the anodic direction compared to the OCP measurement. The pitting potential of the
The nugget area is identical to that of the EN AW-6061 Al alloy. The corrosion current density of the nugget area of the FSW-hybrid joint at \( E_{OC} \) is somewhat higher than that of the USE-FSW-hybrid joint and higher than that of the EN AW-6061 Al alloy. This may be attributed to the higher amount of intermetallic phases within the measured area. These phases show a higher corrosion tendency (figure 4).

The \( E_{OC} \) value of the Al/steel transitional area of the FSW-joint is about 50 mV more negative than that of the USE-FSW-hybrid joint, which may be a result of a different Al to steel ratio or different phases within the measured area. However, both polarization curves exhibit in the anodic range first a continuous increase of the current density - comparable to the DC04 base material - and then a pitting potential and further progress comparable to that of the EN AW-6061 Al alloy. The corrosion current density at the \( E_{OC} \) is lower than that of the DC04 base material. This is a distinct difference to the Al/Mg-hybrid joints where an increase of the corrosion current density of several orders of magnitude was observed at the Mg/nugget transitional area [9].

3.4 Microscopic investigations

The measured areas were investigated by light microscopy after the polarization experiment. The pictures are presented in figure 4.

![Microscopic pictures of the EN AW-6061 Al alloy (a), the nugget on the Al side of the FSW-joint (b) and the USE-FSW-joint (c), the Al/steel transitional area of the FSW-joint (d) and the USE-FSW-joint (e) and the DC04 steel (f) after the polarization measurements.](image)

It is obvious that the EN AW-6061 Al alloy (4 a) shows typical signs of pitting corrosion whereas the DC04 steel (4 f) surface exhibits a whitish oxidation layer after the polarization experiment. In the welding area the corrosion properties change. The nugget area seems to consist of two phases which differ not only in colour but also in their corrosion properties (figures 4 b and c). The darker area shows a significantly stronger corrosion attack than the brighter one. There are not only small pits like in the case of the EN AW-6061 alloy but larger corroded areas. The corrosion properties at the Al/steel transitional area (figures 4 d and e) also differ significantly from that of the base materials. The steel in
this area does not show the whitish oxidation layer. Especially in the case of the FSW-joint (4d) most parts of the steel area seem to be corroded. The first 1-2 mm at the boundary to the Al side show a different colour than the rest of the steel. This may be explained with the formation of intermetallic layers of Al and iron. This is in accordance with the SKP results which show a small seam at the weld area of the FSW-joint with a more negative Volta potential than the steel (figure 1 a). This more corroded area is not observed for the USE-FSW-joint (figure 4 e). The additional ultrasound seems to prevent the formation of this phase.

Detailed analyses of the corrosion products and the phases that show a preferred corrosion attack are still ongoing.

4. Summary
The innovative hybrid joining process of ultrasound enhanced friction stir welding was applied on EN AW-6061 and DC04 Al/steel-hybrid joints. The corrosion properties of the joints have been investigated in air (SKP) as well as in an aqueous electrolyte. It turned out that the two materials behave very different in the two media. The SKP measurements in air result in a difference of the Volta potentials of 1 V whereas the difference of the OCPs in 0.5 molar NaCl solution was only about 50 mV. Moreover the DC04 steel exhibits a more negative potential than the EN AW-6061 Al alloy after 60 min immersion in the electrolyte whereas it has shown the more positive Volta potential in air. As a result of the low potential difference in the NaCl solution no enhanced galvanic corrosion was observed for the EN AW-6061/DC04-hybrid joints as it has been the case for the Al/Mg-hybrid joints. Differences in the corrosion properties of the welded area (nugget, Al/steel transitional area) compared to the base materials may be attributed to the formation of intermetallic phases in this region. The amount of the intermetallic phases seemed to be reduced by the introduced ultrasound as a comparison of FSW- and USE-FSW-hybrids showed.

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Acknowledgements
The investigations were supported by the DFG Priority Program 1640: "Joining by plastic deformation" which is gratefully acknowledged.