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Formation of a diffusion-based intermetallic interface layer in friction stir welded dissimilar Al-Cu lap joints

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Abstract. The joining of dissimilar metals is an important issue in modern lightweight design. Friction Stir Welding (FSW) is suitable for this task since the solidus temperature is usually not exceeded during the process. As a consequence, dissimilar joints can be produced with a minimum of deteriorating intermetallic phases. The latest studies showed the formation of intermetallic layers at the bonding interface with no significant negative influence on the seam quality. In this study, those intermetallic nanolayers at the interface of aluminium / copper lap joints were analysed. For the experiments, the commercially pure alloys EN AW-1050 and CW008A were chosen. The process temperature changed with respect to the parameter setup and was measured at different locations of the seam. The intermetallic layers at the interface were analysed by scanning electron microscopy (SEM) and scanning transmission electron microscopy (STEM). The experiments show that the thickness of the interlayer clearly correlates with the process temperature using an Arrhenius equation. It is supposed, that the rotating probe removes the oxide layers of the metal surfaces and a metallic bonding between the Al- and the Cu-phase is formed. Due to the elevated temperature after the probe has passed, the intermetallic layer has emerged by interdiffusion.

1. Introduction

The use of materials adapted to the locally specified requirements of structures (e.g. load capability, thermal/electrical conductivity) is one of the most promising solutions in modern lightweight design to reach higher efficiencies in terms of energy, mass and cost reduction. This results in the necessity of combining dissimilar materials. The thermal and physical properties of these material combinations are often very different or even competing. Therefore, the joining of dissimilar materials remains challenging for traditional fusion welding technologies. However, innovative solid state welding technologies, such as Friction Stir Welding (FSW), are suitable for this task. The solidus temperature is usually not exceeded during this process, which reduces the formation of thin intermetallic phase layers. As a consequence, high quality joints can be produced.

1.1 Literature review

The use of FSW to join dissimilar materials was reported and confirmed in numerous studies [1]. Most studies focused on feasibility issues or the dependency between the welding conditions, the interface zone structure and the mechanical properties. Many studies also proved the existence of diffusion based intermetallic compounds (IMC) at the interface between the materials.

Giera et al. [2] reported on laser-assisted FSW of butt joints of aluminium (AA5182) and steel (ZStE340). The authors described a saw tooth shaped interface area, which provides mechanical interlocking on the one hand and enlarges the interface area for diffusion on the other hand. Marginal diffusion processes were also discussed by Uzun et al. [3] for butt joints of aluminium (Al 6013-T4) and stainless steel (X5CrNi18-10). Kostka et al. [4] assumed a positive influence of intermetallic phases (particularly Fe_2Al_5) on joints of lap-welded aluminium (Al6181) and steel (ZStE340).

Former studies analysed the interface of friction stir welded aluminium / titanium joints both in butt and in lap joint configuration. Aonuma & Nakata [5] observed mixed regions with the IMC $TiAl_3$ at the interface of butt welded aluminium (2024-T3 and 7075 T651) and titanium (pure and Ti6Al4V) joints. A continuous IMC, consisting of TiAl3 with a thickness of about $100\ nm$, was detected by Wu et al. [6] for butt joints of aluminium (Al6061) and titanium (Ti6Al4V). The authors stated a dependency between the layer thickness, which is also influenced by the rotational speed, and the tensile strength, since the joint strength was reduced for higher layer thicknesses. Dressler et al. [7] analysed butt welded joints of aluminium (AA2024) and titanium (TiAl6V4). It was suggested, that an IMC layer existing in small regions could positively influence the mechanical properties. The formation of $TiAl_3$ was also reported by Chen & Nakata [8] for lap joints of aluminium (ADC12) and titanium (commercially pure). The authors stated a strong dependency between the heat input during FSW and the diffusion based formation of $TiAl_3$, which influences the mechanical properties. Chen et al. [9] performed FSW on lap joints of aluminium (LF6) and titanium (TC1). The existence of IMCs at the interface zone was also indicated in their studies and decreasing feed rates lead to increasing shear strengths. Krutzlinger et al. [10] detected an interface layer with a constant thickness of about $20\ nm$ at the bonding area of lap joints of aluminium (EN AW-1050) and titanium (Ti grade 1). It was assumed that this layer consists of $TiAl_3$. In some experiments, also possibilities for macroscopic form-fit were provided by a recess of the titanium surface. The tensile shear strengths of such joints were not significantly increased compared to joints without any recess and macroscopic form-fit, respectively. Similar observations were reported by Chen & Yazdanian [11] for lap joints of aluminium (6060) and titanium (Ti6Al4V). The authors achieved the highest tensile shear strength when the probe was positioned close to the interface without penetrating the titanium surface. They assumed, that a very thin interface layer leads to high tensile shear strengths, since no interface layer could be detected with the used resolution of $250\ nm$.

Besides that, the combination of aluminium and copper materials is of increasing interest, especially in the electrical industry. A detailed review on friction stir welded aluminium / copper joints is given by Galvão et al. [12]. The studies presented in the following also discuss the occurrence of IMCs (e.g. CuAl, $CuAl_2$, or Cu_9Al_4). Abdollah-Zadeh et al. [13] and Saeid et al. [14] performed FSW on joints of aluminium (1060) and copper (commercially pure). Both studies suggest that a sufficient joint strength results neither from high nor from low heat input conditions. While Abdollah-Zadeh et al. [13] observed an increasing amount of IMCs for an increasing rotational speed or a decreasing feed rate, Saeid et al. [14] reported on cavity defects for increased and on microcracks for reduced feed rates. Continuous and uniform IMC layers with thicknesses of about $1\ \mu m$ were detected by Xue et al. [15; 16] for butt joints of aluminium (1060) and copper (commercially pure). Similar results were reported by Tan et al. [17], who welded butt joints of aluminium (5A02) and copper (T2), and by Muthu & Jayabalan [18], who welded butt joints of aluminium (AA1100-H14) and copper (commercially pure). It is concluded, that thin IMC layers provide high joint strengths. To maintain such thin layer thicknesses, a probe offset from the interface can be used as described by Genevois et al. [19] and Galvão et al. [20], who welded butt joints of aluminium and copper (1050-H16 / Cu-bl and AA6082-T6 / Cu-DHP, respectively). With the probe only stirring in the aluminium matrix, the thickness of the interface layer was reduced to $200\ nm$ in the first study. It was not reported, if there was any influence on the joint strength. In Galvão et al. [20], the IMC build-up could be inhibited with a probe offset. However, there was no improvement of the mechanical properties of the joints. FSW of aluminium (Al99.5) and copper (Cu-DHP) was performed with a stationary shoulder tool by Doehner et al. [21]. It was observed that a probe length causing a small distance between the probe tip and the interface leads to a minimal recess of the copper sheet and results in increased joint strengths. A deeper recess of the copper sheet by an increased probe length causes a drastic reduction of the joint strength due to the occurrence of cavities at the interface zone.

Summarizing the above studies, the build-up of IMC layers during FSW of aluminium and copper is evident. These layers have to be very thin to improve the mechanical properties, especially the strength of the joints. There is a strong dependency between the layer thickness and the heat input on the interface zone, which can be influenced by the welding conditions. However, a detailed description of the mechanisms during welding, which lead to the final joint properties, is still subject of research. In the following, these dependencies are discussed concerning process temperature, joint strength and a detailed analysis of the bonding interface.

2. Experimental and analysis setup

2.1 Experimental setup

A schematic diagram of a lap welded aluminium / copper joint is shown in Figure 1. The sheets had dimensions of $245\ mm\ x\ 100\ mm$ with a thickness of $4.0\ mm$ for the commercially pure aluminium EN AW-1050 and $2.0\ mm$ for the commercially pure copper CW008A. A constant lap width of $40\ mm$ was set for all experiments with the aluminium sheet on the advancing side (AS) and the copper sheet on the retreating side (RS) of the tool. The tool dimensions were $14\ mm$ for the shoulder diameter, $5.0\ mm$ for the base and $3.6\ mm$ for the tip diameter of the conical probe and $3.5\ mm$ for the probe length. In combination with a shoulder plunge depth of $0.1\ mm$, this resulted in an effective probe-tip-to-interface distance of about $0.3\ mm$ for a tool tilt angle of 2 °. The probe was threaded and furnished with three equally distributed flats. A CNC milling machine (Heller MCH250) was used in position controlled mode for the experiments.

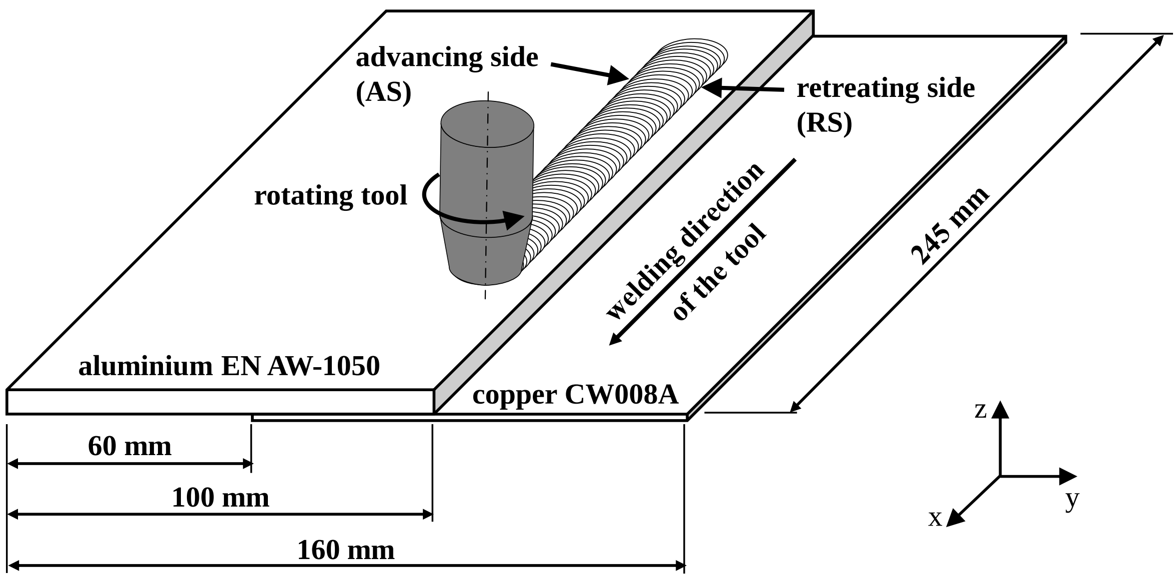

Figure 1. Schematic diagram of the sample dimensions and of the lap welded aluminium / copper joints.

In order to measure the process temperature, each of the sheets (aluminium and copper) was outfitted with three type K thermocouples at various positions along the welding path. The thermocouples were placed $60\,mm$, $122.5\,mm$ and $182.5\,mm$ away from the edge where the process began. The thermocouples with a diameter of $0.5\,mm$ were positioned in drill holes with a depth of $13\,mm$. The holes were placed at the face side of the RS for the aluminium sheets with a distance of $0.55\,mm$ to the interface and at the face side of the AS for the copper in the middle of the sheets (figure 2).

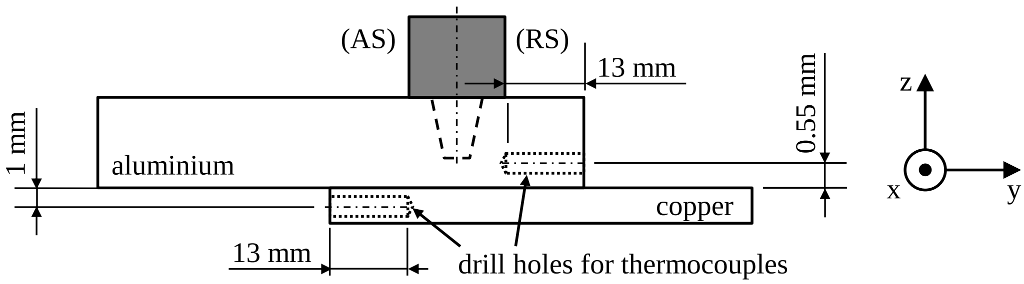

Figure 2. Positions of the drill holes for temperature measurements via thermocouples.

Three specimens were cut out of each joint perpendicular to the welding seam to test the shear strength, tensile tests were carried out on a Zwick Z020 testing machine. The extraction positions were at $62.5\,mm$, $102.5\,mm$ and $142.5\,mm$ from the starting edges of the sheets and the nominal width of the specimens was $20\,mm$.

The rotational speed n was varied during the welding study (see Table 1), while the feed rate remained constant at $v = 300\,mm/min$.

Table 1. Rotational speed settings for the experiments.

Sample	Rotational speed in min^{-1}
1	800
2	1300
3	1800
4	2300
5	2800

2.2 Analysis setup

Two specimens of each sample were extracted perpendicular to the welding seam at $87\,mm$ and $127\,mm$ from the starting edges of the sheets for electron microscopy. The cross sections were prepared metallographically for scanning electron microscopy (SEM) with a final thermomechanical polishing using $40\,nm$ silica particles with hydrogen peroxide. The specimens for transmission electron microscopy (TEM) were electrolytically thinned and finalised by a brief ion polishing. High resolution SEM was executed using a ZEISS MERLIN microscope. A JEOL JEM-2100F allowed TEM with a resolution up to atomic scale.

3. Results and discussion

3.1 Process temperature

Figure 3 illustrates the average of the measured peak temperatures in the aluminium and in the copper sheet for different rotational speeds. The error bars show the minimum and the maximum peak temperature for each rotational speed. The mean temperature in both materials increased with an increasing rotational speed and saturated at a temperature of about $390\,°C$. However, the difference between the minimum and the maximum peak temperatures in the aluminium sheet significantly rises for rotational speeds $n > 1300\,min^{-1}$. This was not observed for the temperatures in the copper sheet.

3.2 Tensile shear strength

The results of the tensile tests are shown in Figure 4. The symbols represent the average and the error bars of the minimum and maximum tensile shear strength of each joint. For a rotational speed of $n = 800\ min^{-1}$, the sheets were not bonded over the entire welding length and only one specimen could be cut out and tested. Therefore, the peak temperatures at $n = 800\ min^{-1}$ seem to be very low for a sufficient bonding. The minimum rotational speed for a sufficient bonding and joint quality should be above $n = 800\ min^{-1}$ for a feed rate of $v = 300\ mm/min$. Comparing the tensile shear strengths to the temperature data, three similar trends can be concluded. First, an increasing rotational speed basically leads to an increased joint strength. Second, the joint strength is reduced at a rotational speed of $n = 2800\ min^{-1}$. As a consequence, the maximum achievable joint strength seems to occur at a rotational speed between $n = 1800\ min^{-1}$ and $n = 2800\ min^{-1}$. Further experiments have to be carried out to prove this. Third, the variations between the maximum and the minimum shear strengths extend above a rotational speed of $n = 1300\ min^{-1}$.

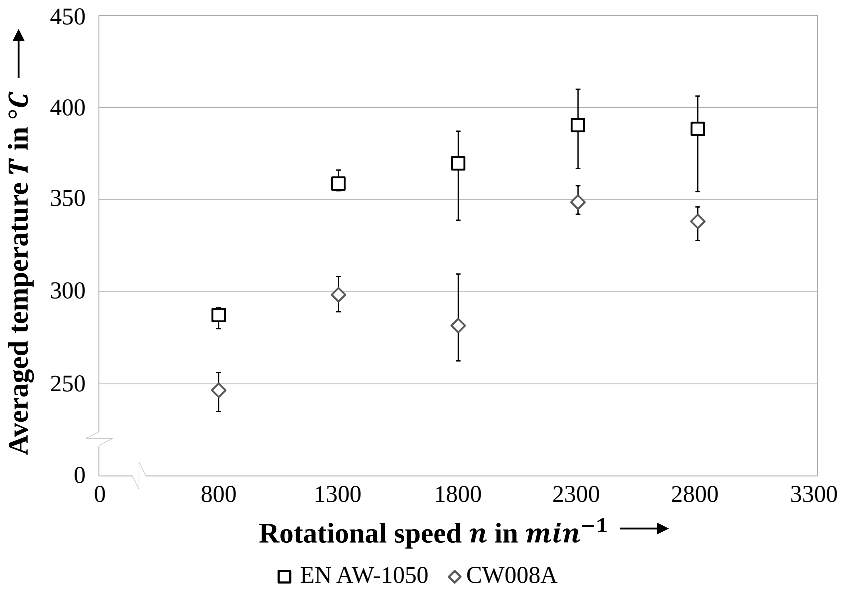

Figure 3. Averaged peak temperatures for different rotational speeds.

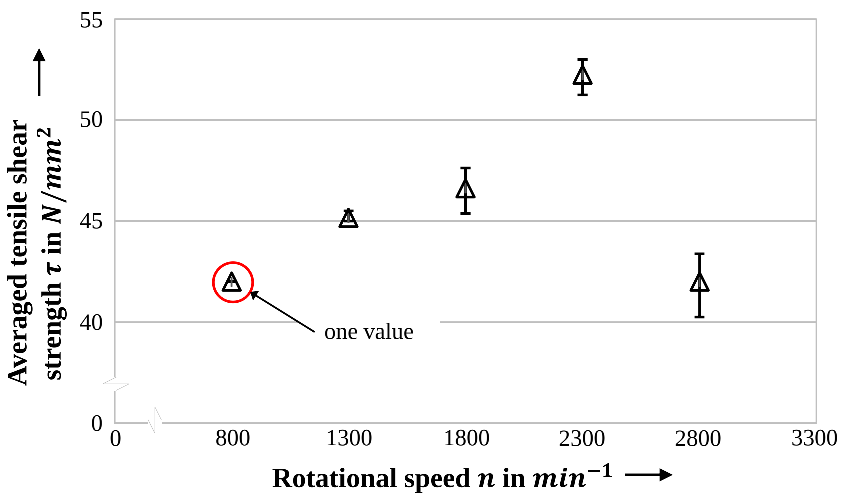

Figure 4. Averaged tensile shear strength for different rotational speeds.

The results seem to indicate a significant dependency between the temperature and the achievable tensile shear strengths of the joints. A further examination of the bonding area was carried out to determine how this dependency relates to the metallographic structure.

3.3 Interface analysis

For all analysed samples, the analysis of the interface area revealed an intermetallic compound (IMC) layer causing the bonding (see Figure 5 a). This layer discernibly consists of two sublayers. Xue et al. [15] observed similar double layers of Al_2Cu and Al_4Cu_9 for intermixed butt joints. In contrast, TEM-EDX analysis distinguished the stoichiometry of the phases as AlCu and $AlCu_3$ in this study. However, the shape of the interface layer, which is smooth and plane on the aluminium side but waved on the copper side, is in good agreement with Xue et al. [15]. The TEM image also shows that the crystals of both phases are columnar on the interface plane. Figure 5 b) shows a high resolution TEM (HRTEM) image of the interface between the aluminium matrix and the AlCu phase layer. It is clearly visible, that the AlCu sublayer consists of a long range crystalline structure with a semi coherence of the atomic plains at the imaged interface. This semi coherence is presumably crucial for the mechanical properties of this phase boundary and therefore of the joint strength. EDX did not detect residual oxides or the occurrence of any elements other than aluminium or copper at the interface, which could influence the bonding.

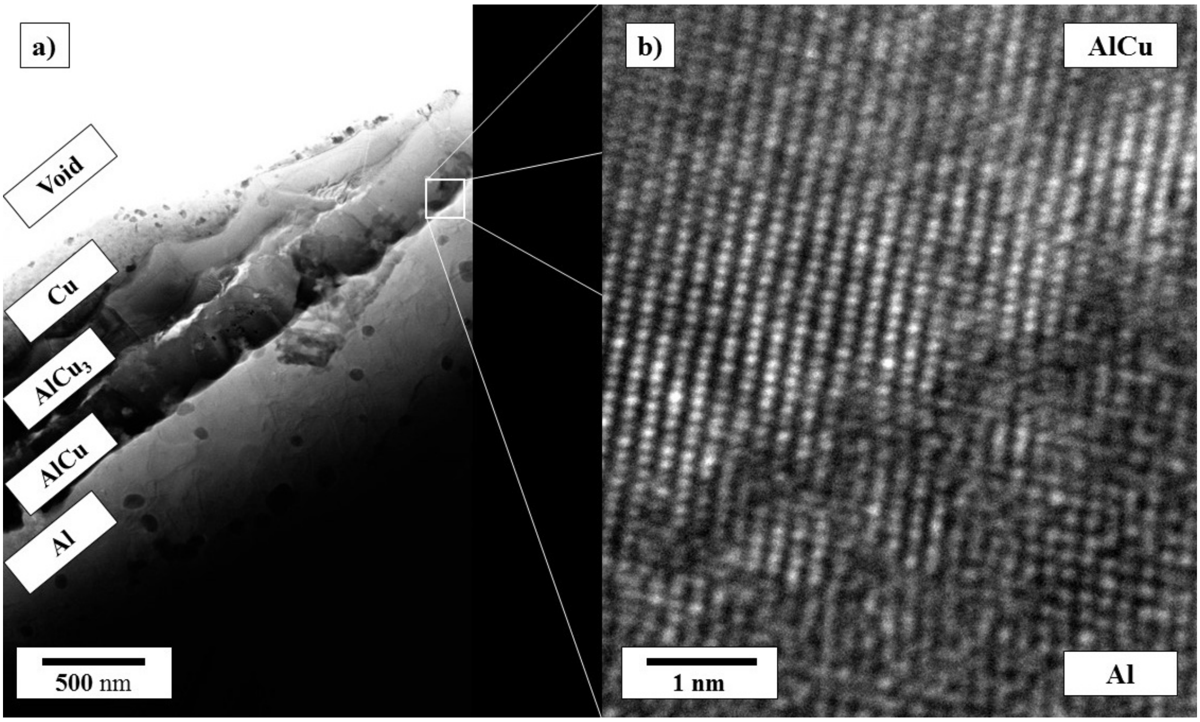

Figure 5. a) TEM image of the IMC interface layer b) HRTEM image of the semi coherence of the atomic plains at the Al to AlCu interface.

TEM and SEM were used to measure the thickness of the IMC interface layer of samples, which were welded with different rotational speeds. As shown in Figure 6, the layer thickness reaches a maximum of $704.6\ nm$ at the rotational speed of $n = 2300\ min^{-1}$. At $n = 800\ min^{-1}$, the formed layer is not continuous and the fragments are only $47.4\ nm$ thick. The error bars represent the standard deviation for at least 5 thickness measurements. The thickest IMC layer was detected for the sample welded at a rotational speed of $n = 2300\ min^{-1}$. The highest peak temperatures as well as the highest tensile shear strengths were also observed at this rotational speed. Lower temperatures lead to a thinner IMC layer and to lower joint strengths. Based on the observed results, the highest joint strengths can be obtained with a layer thickness of $600 - 700\ nm$.

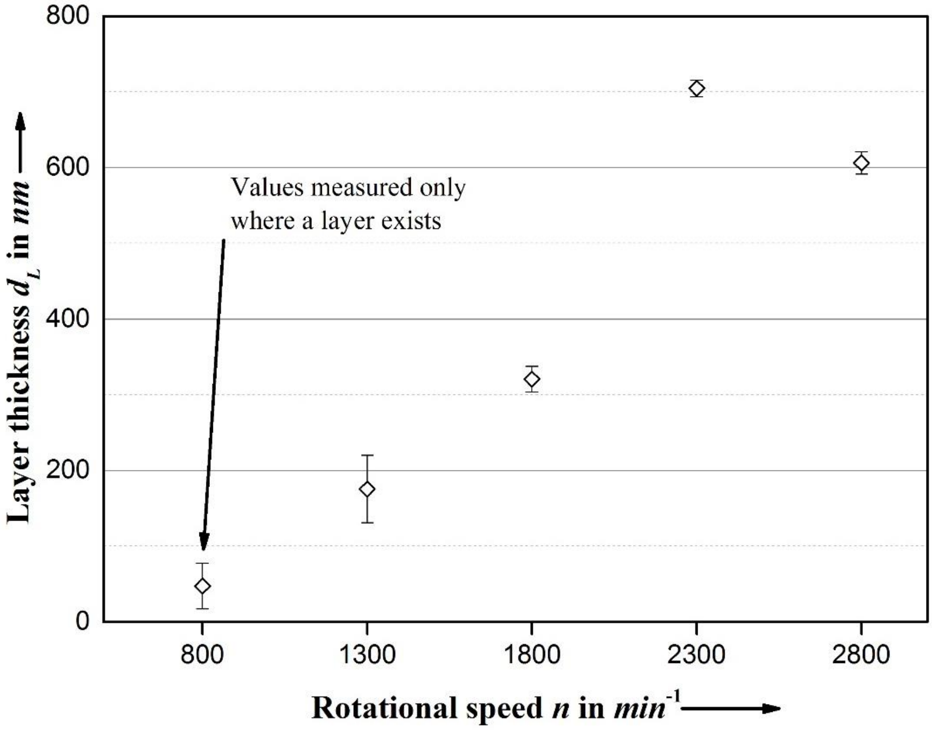

Figure 6. Thickness of the IMC layer for different rotational speeds.

3.4 Correlation between bonding double layer and process temperature

The measured temperature-time curves are equally shaped for all process parameters and differ only in the value of the peak temperatures. The short time interval at the peak temperature primarily controls the thermal activation of the interface region. Figure 7 shows an Arrhenius diagram where the thickness of the IMC interface layer is plotted logarithmically against the inverse peak temperature. The thickness of the barely bonded joint at a rotational speed of $\boldsymbol{n = 800\ min^{-1}}$ is also illustrated in Figure 7 but not used for the calculations since the layer is only partially formed. Hence, the measured thicknesses for this rotational speed only represent maximum values and the average cannot be compared quantitatively to the other thicknesses. The remaining valid layer thicknesses exhibit a linear dependency of the inverse temperatures. As a consequence, the layer thickness of friction stir welded aluminium / copper joints is a result of a thermal activation. Although not taken into account, this linear correlation would also be valid for the broad error margin of the weld at $\boldsymbol{n = 800\ min^{-1}}$. Assuming a thermal activation for diffusion the Arrhenius equation follows

$$\boldsymbol{D(T) = D_0\, e^{\frac{-E_{act}}{k_B\, T}}}$$

$\boldsymbol{D}$ is the diffusion coefficient, $\boldsymbol{T}$ the temperature, $\boldsymbol{D_0}$ is the prefactor of diffusion, $\boldsymbol{E_{act}}$ is the activation energy and $\boldsymbol{k_B}$ is the Boltzmann constant. Further assuming a diffusion controlled and time dependent growth of the IMC layer thickness $\boldsymbol{d(t)}$ as

$$\boldsymbol{d(t) = \sqrt{2Dt}}$$

a linear function for the fit in the Arrhenius plot can be defined as

$$\boldsymbol{ln(d)} = \frac{\mathbf{1}}{\mathbf{2}}\boldsymbol{ln\left(2t_{eq}D_0\right)} - \frac{\boldsymbol{E_{act}}}{\boldsymbol{2k_B}}\frac{\mathbf{1}}{\boldsymbol{T_{peak}}} \ .$$

t_{eq} is set to the equivalent time interval around the temperature peak T_{peak}, during which considerable thermal diffusion takes place. It is of the magnitude of a few seconds. Determined from the slope of the Arrhenius plot, the activation energy is $E_{act} = 2.33\,eV$ and the prefactor D_0 ranges from $10\ ^{8}cm^2/s.$ to $10^9\ cm^2/s.$ The activation energy is in reasonable agreement with data for Al-Cu interdiffusion in Funamizu & Watanabe [22]. Although the diffusion prefactor is higher than the values found by Funamizu & Watanabe [22], the value reported here is considered still plausible due to the complex diffusion system generated during the FSW process.

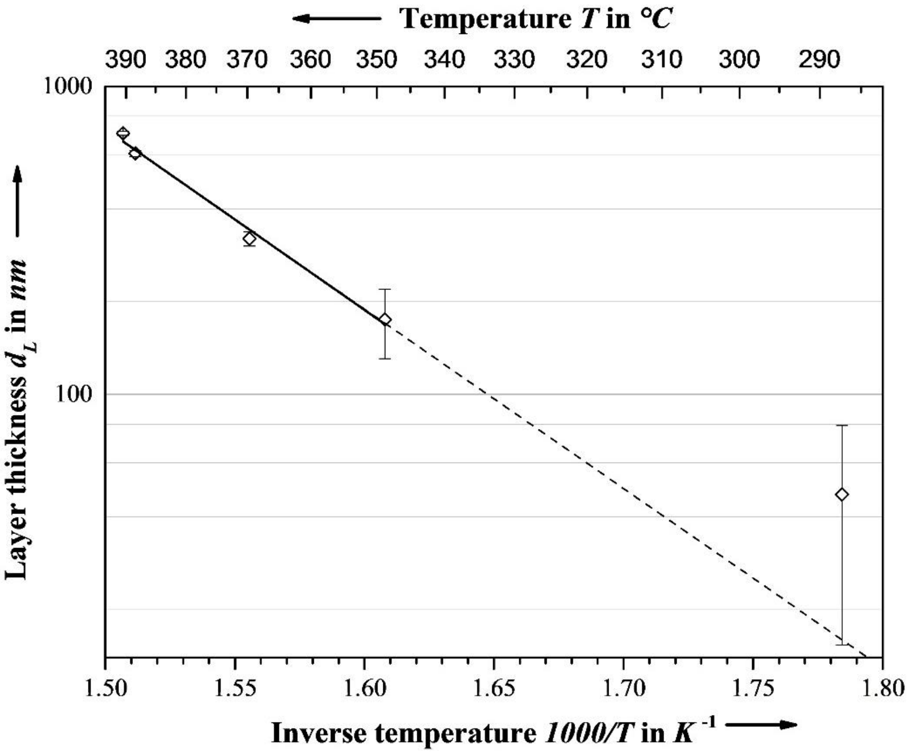

Figure 7. Arrhenius plot of the thickness of the double layer against the process peak temperature with a linear fit for calculations.

4. Conclusions and outlook

Since the material is heated by the friction during FSW, the process temperature is directly affected by the rotational speed. The temperature rises with an increasing rotational speed and saturates at about $390\ °C$ as a result of a decreasing friction based heat input caused by material softening. Similar tendencies could be observed for the dependencies between the rotational speed and the tensile shear strength of the joints. The ideal process conditions seem to exist shortly before the process temperature saturates. During FSW, a nanoscaled intermetallic double layer consisting of AlCu and $AlCu_3$ forms. The structure of the IMC layer at the bonding interface is columnar grown with a well-defined orientation. The layer thickness is controlled by thermal activation. Hence, this layer is formed by interdiffusion after the oxide layers at the interface are removed by the FSW process. This leads to a direct contact of the aluminium phase and the copper phase under the elevated thermal conditions.

The thickness of the double layer can be predicted by measuring the temperature during FSW. This predictability is the first step to a defined and reliable application of FSW for the commercial joining of aluminium and copper.

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