Microstructure of arc brazed and diffusion bonded joints of stainless steel and SiC reinforced aluminum matrix composite

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Abstract. Joint interfaces of aluminum and stainless steel often exhibit intermetallics of Al-Fe, which limit the joint strength. In order to reduce these brittle phases in joints of aluminum matrix composites (AMC) and stainless steel, diffusion bonding and arc brazing are used. Due to the absence of a liquid phase, diffusion welding can reduce the formation of these critical intermetallics. For this joining technique, the influence of surface treatments and adjusted time-temperature-surface-pressure-regimes is investigated. On the other hand, arc brazing offers the advantage to combine a localized heat input with the application of a low melting filler and was conducted using the system Al-Ag-Cu. Results of the joining tests using both approaches are described and discussed with regard to the microstructure of the joints and the interfaces.

1 Introduction

In the field of light-weight materials AMC are the subject of current research work. The advantages of AMC in comparison to aluminum alloys are a high specific strength, an increased wear resistance and a low coefficient of thermal expansion (CTE) [1-3]. Numerous applications in aerospace and automotive industry as well as plant manufacturing can be found. Thermal expansion matched heat transfer components can be mentioned for example. To realize this application, an appropriate joining technology for dissimilar joints of AMC and stainless steel is required. In recent works, the suitability of different joining technologies and treatments was investigated. For example, joints of aluminum and steel are realizable by using the TIG (Tungsten Inert Gas) welding process. The TIG welding of AMC leads to an inhomogeneous welding area. Furthermore an unequal distribution of the reinforcing particles and the formation of intermetallics can occur. The properties of these joints are insufficient [4]. Brittle intermetallics like Al₅Fe₄, Al₂Fe₃, Al₂Fe and Al₂Fe₅ downgrade the properties of the joints. To reduce the formation of the intermetallics, a zinc coating is used [5]. Another approach is the decrease of the heat input while joining. This is also important because of the thermal induced damage of the AMC that leads to ineligible porosities and inhomogeneities [6]. This can be avoided by another process to join steel and AMC: Friction stir welding [7]. During the process the tool, however, is affected by the reinforcement particles, which leads to a considerable wear. Therefore, the process conditions change continuously, which affect the reproducibility of the process results [6]. Transient liquid phase bonding (TLP bonding) is another suitable process for joining AMC. One advantage is that the surface condition is not important due to the liquid state of the interlayer at joining temperature [8, 9]. Askew et al. investigated TLP bonding of AMC with nickel interlayer [9]. The results indicate, that the shear
strength increases by limiting the bond line width to 1 µm, but the joints were not free of defects and failures.

A high potential has arc brazing. The melting temperature of the filler is lower than the base materials one, which avoids thermal damage of the AMC. Due to the focused arc, the heat input is reduced in comparison to TIG welding. Another advantage is the degradation of the oxide layer on the AMC induced by the arc, which improves the wetting. The formation of intermetallics can be reduced by an adapted filler. For brazing of aluminum alloys, Al-Si based [10-12] and Zn-Al based [13, 14] fillers are commonly used. The disadvantages are the limited number of aluminum alloys brazable by Al-Si fillers and the poor corrosion resistance of Zn-based fillers. In this work, a filler based on the ternary system Al-Ag-Cu, designed for arc brazing, is investigated. The eutectic composition of 40 wt% Al, 40 wt% Ag and 20 wt% Cu (named Al40Ag20Cu) has a melting temperature of 506 °C, measured by differential scanning calorimetry (DSC) [14].

Diffusion bonding is another suitable joining technique to avoid the formation of intermetallics within joints of aluminum and stainless steel [15, 16]. Zhang et al. investigated diffusion joints of AMC, with different SiCp volume fractions and interlayer materials [17]. The tensile strength of these joints decreases with an increasing volume fraction [17, 18]. It is also limited by the surface preparation or surface roughness of the samples [17-19]. Furthermore, an interlayer was used to compensate the difference in surface roughness of the two samples and to improve the joint properties [17, 20, 21]. By using an active interlayer material, e.g. lithium, the surface oxides can be reduced. Also the formation of intermetallics could be avoided [21]. However, there is still a need for research for joints of AMC and stainless steel.

2 Experimental Procedure

In the present study the used base materials are a stainless steel and a powder-metallurgically produced aluminum matrix composite (AMC). The stainless steel is grade X2CrNi18-10 (material no.: 1.4301 / AISI 304). The matrix of the AMC is similar to EN AW-2017, reinforced by 10 % SiC particles with an average particle size of 1.5 µm. The feedstock of the AMC is mechanically alloyed in a high energy ball mill. A rod is produced by hot isostatic pressing, extruding is carried out afterwards. Different sample geometries are necessary for arc brazing and diffusion bonding experiments, figure 1. The generated rods with a cross-section of 15 x 15 mm² are cut in half lengthwise and divided into 50 mm sections for arc brazing. The cut surfaces are ground and polished (Rz < 1 µm) prior joining tests. The stainless steel sheets with a thickness of 1.5 mm have dimensions of 20 x 50 mm². The surface is polished using a hand grinder and cleaned by ethanol afterwards. For diffusion bonding the same materials in a smaller sample size were used. The dimensions of the AMC samples are 14 x 14 x 10 mm³, that of stainless steel samples are 14 x 14 x 1.5 mm³. Important parameters for diffusion bonding are in addition to the temperature-time-load-regime the surface condition or the surface roughness. Therefore, the joining surfaces of the samples were analysed in accordance with ISO 25178 and in dependence of the machining process. The stainless steel surface was used in the as-delivered condition with a surface roughness Sz = 15.4 µm. The AMC joining surface was turned to a surface roughness of Sz = 14.2 µm and additionally sand-blasted to Sz = 32.3 µm. The higher surface roughness of the sand-blasted samples causes micro surface deformations during the application of the load, which increases the number of lattice defects at the interface and thus increases the diffusibility. The plane parallelism of each sample was measured to ensure a deviation of less than 10 µm, so that a tight interface contact and hence defect-free joint can be reached. To remove the oxide layer or impurities on the joining surfaces, which limit the diffusivity, all samples were chemically treated in a glove box in inert atmosphere. The AMCs were etched in 5% NaOH for 45 s and the stainless steel samples were cleaned with acetone. After the chemical surface treatment, both materials were batched and moved under inert atmosphere protection into the joining chamber.
Arc brazing

For arc brazing a new developed filler alloy of the system Al-Ag-Cu-Si is used [22]. The base composition is the eutectic with 40 wt% Al, 40 wt% Ag and 20 wt% Cu, named Al40Ag20Cu. In a previous work the wettablility of stainless steel was improved by additional alloying with 1.5 wt% Si [23]. Al40Ag20Cu + 1.5 wt% Si has a melting temperature of 498 °C, measured by DSC. All filler alloys are prepared by melting and continuous casting. Therefore B-Ag72Cu28 as master alloy is molten in the continuous caster with additions of Al (purity 99.99 vol%), Cu (purity 99.9 vol%) and Si (purity 99.9999 vol%). The induction melting under Ar atmosphere (purity 99.996 vol%) takes place in a graphite crucible of the quality GM-125i. A wire of 2 mm diameter is produced by continuous casting. The cast specimens are analyzed by X-ray fluorescence analysis (voltage 30 keV) to ensure a good homogeneity. All brazing tests are carried out in an inert atmosphere (Ar). A TIG welding source with a maximum current of 250 A which enable alternating current (AC) is employed for this purpose. The applied current intensity of 40 A is a result of previous wetting tests [23].

Diffusion bonding

All experiments are carried out in a diffusion bonding machine with a maximum joining temperature of 1000 °C, 80 t max. load and a chamber size of 300 x 300 220 mm³. Additionally, a glove box is directly connected with the diffusion bonding machine, so that it is possible to treat the joining surfaces mechanically or chemically in defined atmosphere and move the samples into the chamber under inert atmosphere. The joining tests are carried out under inert atmosphere 700 hPa Argon (99.999% Ar) at different temperatures (380-560 °C) over a time of 60 min. A constant load of 10 N/mm² is used in the present study. Only the joining temperature is varied to examine the microstructural changes of the stainless steel-AMC interface as a function of the temperature. The parameter sets were selected to avoid the formation of the critical Al-Fe intermetallics by a systematic temperature reduction. 560 °C as highest joining temperature is chosen, since the intermetallic compound CuAl and Cu11,5Al9 form at higher temperatures and the possible interaction of the Cu-Al and Al-Fe intermetallics can be investigated. The characterization of the joined samples after arc brazing and diffusion bonding is performed by light microscopy (LM) and scanning electron microscopy (SEM). The composition of the phases and the diffusion zones are examined by energy dispersive X-ray spectroscopy (EDX) and X-ray diffraction analysis (XRD). The hardness is determined as Martens hardness (HM) profiles across the joint with a measuring force of 50 mN. The values in HM are transferred in Vickers hardness (HV) to compare the hardness results with typical literature values.

Results and Discussion

Arc brazing

For arc brazing the AMC/braze metal interface and the stainless steel/braze metal interface are considered. The first one shows a diffusion zone in the direction of braze metal near to the AMC and a zone of alloying afterwards, figure 2. The alloying results in an increase in hardness at the interface and the alloying zone, 280 HV0.005 to about 350 HV0.005 at the interface. The hardness of the pure filler was measured at 306 HV1, previously. The difference in the hardness values is a result of the alloying pro-
cesses between the filler and the base material. Additionally, the rising influence of defects like porosities at small test loads may cause the differences in the values.

![Microstructure (SEM) of AMC/braze metal interface, position of indents (LM) and hardness](image)

**Figure 2.** Microstructure (SEM) of AMC/braze metal interface, position of indents (LM) and hardness

Before brazing the eutectic filler consists of the phases Ag$_2$Al, Al$_2$Cu, a solid solution of Al and additional Si, analyzed by XRD. The peak in hardness is a result of alloying processes between the filler and the base material and the development of a solid solution of aluminum and intermetallics. After brazing the phases Ag$_2$Al, Al$_2$Cu and a solid solution of Al are detected using XRD analysis. The EDXS shows an increased content of Ag (up to 5 at%) in the solid solution of Al. This amount is higher than the solubility in Al in equilibrium state (< 2 at%). Therefore, Ag rich intermetallics are precipitated and lead to an increased hardness. Subsequently the hardness slowly decreases to about 280 HV0.005 towards the AMC. The hardness of the AMC doesn’t drop to lower values, which indicates a diffusion zone or a section of heat treatment near the joint. The EDXS mapping in this section shows that no element of the filler enriches the AMC near the interface. A diffusion zone is not the reason for the hardness profile in AMC. It can be seen that the distribution of Cu within the AMC matrix metal is finer as in the untreated base material. Due to the heat treatment during brazing, a precipitation hardening effect is one possibility to explain the increase in the hardness of the aluminum alloys. No cracks can be detected at the interface to AMC.
At the interface to stainless steel the diffusion and reaction zones appear differently in comparison to the AMC, figure 3. The morphology of the braze metal and the filler is dissimilar. The reaction zone is large in its areal extent. The hardness of the braze metal (550 HV0.005) increases after brazing due to the formation of intermetallics. The hardness values reach a maximum of about 870 HV0.005 at the interface between the phase seam and the braze metal. In the direction of the braze metal the hardness decreases faster as in the direction of the stainless steel. The hardness value of the braze metal at the interface to the reaction zone near the stainless steel (570 HV0.005) is approx. two times higher than at the interface to the AMC, figure 2.

Figure 3. Microstructure (SEM) of stainless steel/braze metal interface, position of indents (LM) and hardness

The EDXS shows a reduction of the content of Al in the reaction zone from braze metal to the stainless steel. The increased hardness is probably a result of the formation of intermetallics of the system Fe-Al. Using XRD analysis, the different phases AlFe, Al-Cr-Fe and Al-Fe-Ni are determined. The correlation of the pattern to the phases is difficult due a marked texture. The highest hardness (870 HV0.005) indicates the phase FeAl3 (892 HV1) [24]. In some places, cracks can be observed in the joining zone, figure 4 and figure 5. Cracks in joints of aluminum and stainless steel mainly occur in the brittle intermetallics. The zone of reaction and diffusion consists of different layers, figure 5. Cracks occur at the interface between the Al rich phases of the system Al-Fe and the Fe rich phases. The reason is the difference in hardness, FeAl3 (892 HV1) and AlFe (470 HV1) [24].
3.2 Diffusion welding

Figure 6 shows a cross-section of the stainless steel-AMC joint with an almost 20 µm wide diffusion zone across the interface. No pores have been detected in this zone, as the AMC-material at 560 °C gets so soft that both surfaces align very well to each other. Furthermore, no differences between turned and sand-blasted AMC joining surfaces were detected. In some joints, cracks, probably caused by the thermal stresses during cooling, were formed in-between the two intermetallic compounds of the diffusion zone. Areas of potential lower thermal stresses showed no cracks. The backscattered SEM-micrograph shows an area of the joint which is free of cracks, figure 7. EDXS have shown that the two intermetallic compounds are most probably Fe2Al5 and Fe4Al13 and the white pattern inside the AMC base material are Al-Cu precipitations. To limit the formation of the critical intermetallic Al-Fe compounds, the joining temperature was systematically reduced to 380 °C. The joints with turned surfaces, shown in figure 8, exhibit no pores, gaps or cracks along the interface. In contrast to that, the interface of the joints with sand-blasted AMC surfaces is interrupted by gaps, due to the limited load of 10 N/mm² at 480 °C and 380 °C and thus the lower surface deformation. To avoid these gaps one approach will be to increase the surface pressure, so that higher micro surface deformation occurs and thus a defect-free joint can be ensured. However, it can be seen that there is no diffusion zone at 480 °C and 380 °C. Also the SEM analysis of the joints showed no formation of the intermetallic Al-Fe compounds.
Figure 6. Interface of a diffusion bonded AMC-stainless steel joint formed at 560 °C with approx. 20 µm wide diffusion zone along the interface; turned joining surface

Figure 7. SEM micrograph of the AMC-stainless steel joint without cracks inside the diffusion zone; turned joining surface, joining temperature 560 °C

Only an increase of preparation artefacts, caused by the reaction of the base materials and the polishing suspension, along the interface and inside the AMC base material at 380 °C could be observed. Figure 9 shows, that these artefacts contain just some cracks with a dimension of less than 4 µm in length and 1 µm in width. The experiments with sand-blasted joining surfaces and a joining temperature of 380 °C resulted in weak joints and broke apart during cutting.
Figure 8. Influence of joining temperature on interface formation of diffusion bonded stainless steel-AMC joints with turned joining surfaces at a) 480 °C and b) 380 °C

Figure 9. SEM micrograph of the stainless steel to AMC diffusion bonded joint with some cracks on the AMC side along the interface

Figure 10 summarizes the results of the micro-hardness measurements. Especially the hardness profile across the diffusion zone of the joint at 560 °C confirms the expected critical behavior: an increase of hardness of approx. 800 HV0.01 in the Fe₄Al₁₃ intermetallic compound compared with the AMC base material. Due to its small dimension, the hardness of the Fe₂Al₅ phase could not be measured directly. The approximation of the hardness profile shows, that the Fe₂Al₅ phase has a hardness of around 580 HV0.01. Thus, both intermetallic compounds form a continuous metallurgical notch at the interface. This is the reason why cracks will be formed along the interface, promoted by thermal stresses or other (external) loads, figure 7. However, the diffusion bonding experiments at 480 °C and 380 °C did not show such a metallurgical notch with an increase of hardness at the interface. From figure 10, a much more remote change in hardness between the AMC material and the stainless steel, based on the lower joining temperatures and thus the limited formation of brittle intermetallic compounds, can be seen.
4 Conclusion
Both processes show new promising approaches for joining particle reinforced aluminum matrix composites (AMC) to stainless steel. The investigation of the microstructures of the joints leads to the following results:

1) During arc brazing of AMC and stainless steel, alloying of the filler material occurs, and a diffusion/reaction zone is recognizable. The slope of the hardness profile at the interfaces of braze metal to the AMC is more remote than that to the stainless steel. That is a result of the formation of Al-Fe intermetallics at the interface to stainless steel. The hardness values and XRD analyses indicate the phases FeAl$_3$ and AlFe. The reaction/diffusion zone at the stainless steel side obviously consists of different phases.

2) Diffusion bonding of AMC to stainless steel at 560 °C results in brittle intermetallic phases in the joint area that comprise Al and Fe (Fe$_2$Al$_5$ and Fe$_4$Al$_{13}$), so that cracks will form along the interface most likely. Al-Cu intermetallics were not detected along the interface. Micro hardness measurements across the diffusion zone showed that the Fe$_2$Al$_5$ phase exhibits an approx. 6 times higher and the Fe$_4$Al$_{13}$ phase an approx. 10 times higher hardness than the AMC.

3) By reducing the joining temperature to a range of 380 °C to 480 °C in combination with surface treatments, Al-Fe intermetallics are avoided and thus a continuous change in hardness can be attained.

Arc brazing and diffusion bonding are suitable to join AMC and stainless steel. The presented results of both approaches show the potential and differences of the investigated processes. The determination of the mechanical properties with respect to process parameters and the resulting microstructure are the purpose of the future work.

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