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Influence of the Thickness of the Reaction Zone in Aluminum/Stainless Steel Brazed Joints on the Mechanical Properties

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Abstract: The study deals with the characterization of the relationship between the microstructure of the reaction zone and the mechanical properties in the brazed joints of aluminum alloy 3003 and stainless steel AISI 304 in order to determine the influence of the intermetallic layers on the tensile shear strength of the joints. The joints were produced by induction brazing using an AlSi10 filler in an argon atmosphere at a temperature of 600 °C. Due to the local heat input into the liquid brazing filler during a short brazing time, a thin reaction zone is formed in the brazed joints (~1 µm), which ensures good mechanical properties of the joints. In order to observe the growth kinetics of the reaction zone in the brazed joints and to investigate the influence of the thickness of the reaction zone on the mechanical properties of the brazed joints, the joints were aged at temperatures of 200 °C and 500 °C for 6, 48 and 120 h. The results have shown that the thickness of this layer increases to a maximum of 2 µm depending on the duration of the thermal aging at a temperature of 200 °C. The results of the tensile shear strength tests have shown that the brazed joints with this thin layer ensure a high strength. The thermal aging at a temperature of 500 °C influences the growth of the reaction zone in the brazed joints significantly. The total thickness of the reaction zone increases to a maximum of 12 µm during the thermal aging. The results of the tensile shear tests of these joints have shown that the thermal aging at a higher temperature leads to a decrease of the tensile shear strength of the brazed joints to 67% due to the growth of the existing intermetallic layer and the formation of a new intermetallic layer in the reaction zone.

Keywords: aluminum; stainless steel; Al-Si filler metal; induction brazing; intermetallic layer; tensile shear strength

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1. Introduction

Due to the potential in weight and cost reduction of various components, aluminum/stainless steel joints became more and more interesting in the automotive industry [1]. To produce a high-quality joint between aluminum alloys and steels, several joining processes have been investigated [2]. For example, conventional mechanical fastening processes like self-piercing riveting and flow drill screwing are limited when the materials to be joined have high strength and low ductility [3,4]. The novel mechanical fastening processes using nonconventional fasteners show high strength and corrosion resistance and low thermal expansion. However, they require a long process time [5,6]. Moreover, the geometry and dimensions of these components require sufficient mechanical properties as well as sealing and insulating properties. Therefore, the cohesive joining processes offer clear advantages compared to the mechanical fastening processes. The cohesive joining processes include gluing, welding and brazing. The joined components are commonly used in aggressive media and under thermomechanical stress. Hence, aging and creep processes occur in the joints. Therefore, gluing of these components is not possible.

For such applications, welded and brazed joints are of interest, because they show good mechanical properties (high temperature strength and fatigue strength) and a higher corrosion resistance in comparison to adhesive joints [7]. The welding techniques like resistance spot welding [8], friction stir welding [9], arc weld-brazing [10] and laser weld-brazing [11] offer a great potential for aluminum alloy/steel joining. However, these processes are often limited to special part geometries and designs of the welded joints. In comparison to welding, brazing offers the possibilities to manufacture several high-quality joints with a complex geometry in one step at lower temperatures. Especially, induction brazing allows a short brazing time and a local heat input. Consequently, good mechanical properties are achievable [12]. Nevertheless, the joining of aluminum to stainless steel is a great challenge due to the formation of hard and brittle Fe-Al intermetallic layers at the dissimilar interface [7]. Achar et al. found out that the thickness of the intermetallic layer has a great influence on the tensile strength of aluminum/steel joints. The results of monotonic tensile tests on aluminum/steel joints produced by fusion welding showed that an increase in the thickness of the intermetallic layer leads to an exponential decrease of the tensile strength of the joints [13]. In regard to the tensile strength of pure aluminum, a critical thickness of the intermetallic Fe-Al layer is about 15 μm for such mixed joints [13]. The influence of the intermetallic Fe-Al layers on the mechanical properties of the joints produced by laser welding was also investigated. It was found out that the mechanical properties of joints with a reaction zone of less than 10 μm are acceptable for technical applications [14]. The highest tensile strengths of the aluminum/steel joints produced by arc brazing were achieved with an intermetallic Fe-Al-Si layer of about 2 μm [15].

Consequently, the growth of the intermetallic Fe-Al layers can be controlled and avoided by reducing the joining temperature and duration as well as adding certain alloying elements to the filler material [7]. With regard to the modification of the filler metal, Akdeniz et al. analyzed the effect of alloying elements on the formation and growth of the reaction zone after the interaction between iron and liquid aluminum. It was found out that the alloying elements Mg, Si, Ca, Ti, Cr, Cu, Ge, Ag, Cd and Sb reduce the activation of aluminum atoms. This leads to a decrease in the thickness of the Fe-Al intermetallic layer [16]. Cheng et al. investigated the influence of the addition of silicon on the thickness of the reaction zone during the interaction of the steel with liquid aluminum. A mild steel was coated in a bath of pure aluminum and the alloys AlSi0.5, AlSi2.5, AlSi5 and AlSi10 at a temperature of 700 °C for 180 s. It was found out that the minimum thickness of the reaction zone could be achieved at a Si content of 10 wt.% [17]. With regard to the reduction of the joining temperature and duration, Roulin et al. reported that the brazing time has an important influence on the thickness of the intermetallic layer formed in the reaction zone during furnace brazing. It was possible to produce aluminum/steel joints with a minimum thickness of the reaction zone of 10 μm [18]. Induction brazing offers the possibility to reduce the brazing time to very short values of some seconds in the liquid state. This results in a reaction zone thickness of about 2 μm. Hence, the risk of the formation of the brittle intermetallic layers is minimized because of the reduction of the time for the diffusion processes [19].

However, the relationship between the characteristics of the intermetallic layers of the reaction zone (thickness and composition) and the mechanical properties of the joints with regard to the joining process have not been systematically investigated yet. The main aim of the present work is to investigate the relationship between the microstructure of the reaction zone and the mechanical properties of aluminum/stainless steel joints produced by induction brazing using an AlSi10 filler metal in order to determine the influence of the intermetallic layers on the tensile shear strength of the joints.

2. Materials and Methods

Aluminum alloy 3003 (AA 3003) sheets with dimensions of $5 \times 20 \times 1.5$ mm^3 and austenitic stainless steel (AISI 304) sheets with dimensions of $40 \times 20 \times 1.5$ mm^3 were used as base materials. The AlSi10 filler metal (AA 4045) was applied as a paste. The thickness of the produced brazed joints was adjusted at 100 μm. The chemical compositions of the used materials are presented in Table 1.

Table 1. Chemical composition of the used materials.

Alloy	Alloy Composition (wt.%)									
	Al	Fe	C	Cr	Ni	Si	Mn	Cu	Zn	Mg
AA 3003	bal.	0.7	-	0.05	0.05	0.6	1	0.2	0.1	0.05
AISI 304	-	bal.	0.07	18	9	0.4	1.4	-	-	-
AlSi10	bal.	0.8	-	-	-	10	0.05	0.3	0.1	0.05

In previous work [19], it was found out that single lap joints of stainless steel and aluminum failed in the Al base material. Hence, no information about the properties of the joint was possible. The optimized sample geometry uses a double lap joint of stainless steel with aluminum in between, as shown in Figure 1. The overlap length is 5 mm. This sample geometry allows the accurate determination of the mechanical properties of the brazed joint, because the influence of the mechanical properties of the aluminum base material was reduced [20].

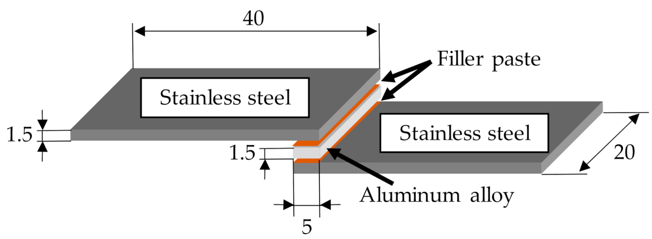

Figure 1. Investigated material combination, produced using a filler paste (mm).

Before brazing, the surfaces of the base materials were cleaned by abrasive paper and ethanol. The aluminum/stainless steel joints were produced by induction brazing in an argon atmosphere at a temperature of 600 °C. This temperature was chosen with regard to the melting temperature of the filler of 575 °C and the liquidus temperature of the aluminum base material of 640 °C. The brazing temperature was measured by a twin-channel pyrometer Impac® (IMPAC Electronic GmbH, Frankfurt, Germany). The brazing process including cooling time takes about 2 min. After brazing, the samples were prepared by mechanical grinding and polishing. Long-term thermal exposure experiments were done in a muffle furnace (Linn High Therm GmbH, Eschenfelden, Germany) at temperatures of 200 °C and 500 °C for 6, 48 and 120 h to observe the growth kinetic of the reaction zone in the brazed joints. The exposure temperature of 200 °C corresponds to the maximum application temperature of aluminum/stainless steel brazed joints. The exposure temperature of 500 °C was chosen to produce a high thickness of the reaction zone in order to evaluate its influence on the mechanical properties of the brazed joints.

Cross sections of the brazed samples were used to control the thickness of the reaction zone before and after the long-term thermal exposure experiments. The microstructure and thickness of the reaction zone in the brazed joints was characterized using a scanning electron microscope Zeiss Leo 1455VP (Carl Zeiss Microscopy GmbH, Jena, Germany). The chemical composition of the microstructural constituents was analyzed by energy-dispersive X-ray spectroscopy Ametek Genesis MK2 (AMETEK GmbH, Meerbusch, Germany) in SEM. The mechanical properties were determined by monotonic tensile shear tests at ambient temperature. The tensile tests with a test speed of 0.01 mm/s were carried out in a material testing machine Zwick Allround-Line 20 kN (ZwickRoell GmbH & Co., KG, Ulm, Germany). Five samples for the brazing process and for each long-term thermal exposure experiments were tested. Furthermore, the fracture behavior was observed and discussed.

3. Results

3.1. Long-Term Thermal Exposure Experiments at 200 °C and Tensile Shear Tests

The microstructure of the aluminum/stainless steel joints brazed using AlSi10 filler was investigated in the previous work [19]. It was reported that the resulting braze metal consists of a primary Al solid solution and an Al-Si eutectic. At the interface to the stainless steel, an Al_7Fe_2Si layer is formed. During the short-time induction brazing, no rapid growth of the layer occurs. The Al_7Fe_2Si layer is about 1 µm thick. According to the results of the work [20], the Al-Fe-Si precipitates formed in the braze metal also correspond to Al_7Fe_2Si phases. The results of the long-term thermal exposure experiments conducted at 200 °C for 6, 48 and 120 h were presented in [19]. The SEM images of the reaction zones after the brazing process and after the aging experiments can be seen in Figure 2a–d. It was found out that the long-term thermal load influences the growth of the Al_7Fe_2Si layer respectively in the reaction zone. Nevertheless, a good mechanical strength of the brazed joints can be expected, because the thickness of the layer is less than the critical thickness of 10 µm [14].

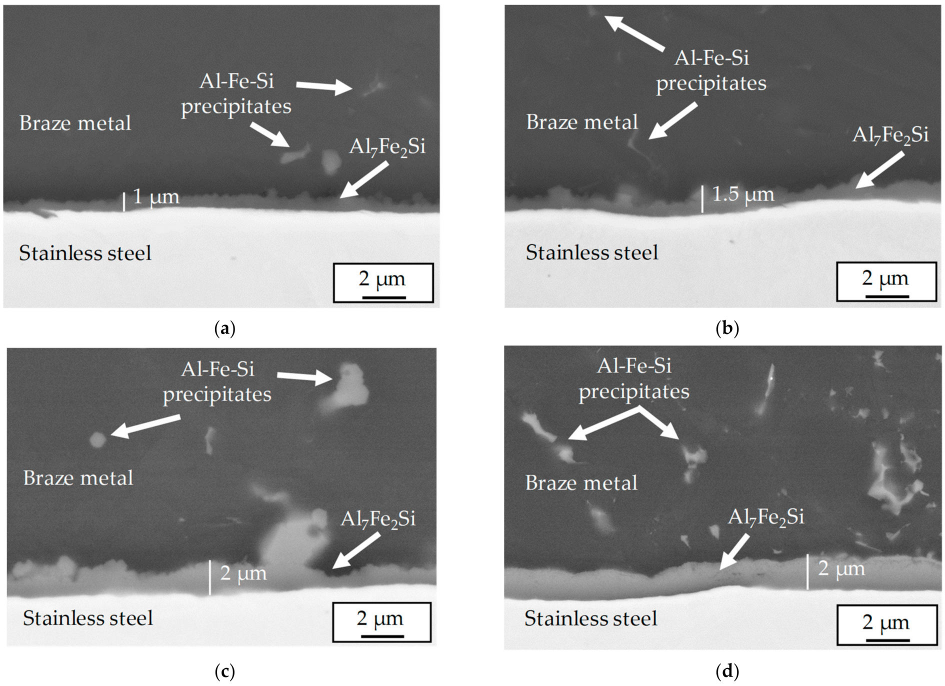

Figure 2. SEM images of the reaction zones after brazing (**a**) and after long-term thermal exposure experiments at a temperature of 200 °C for 6 h (**b**), for 48 h (**c**) and for 120 h (**d**).

In the present work, tensile shear tests of the brazed joints after long-term thermal exposure experiments were carried out in order to investigate the influence of the thermal load during the application of the joints. The results of the tests are presented in Figure 3. It was determined that the average value of the tensile shear strength is 54 MPa for the joints after brazing. It can be seen that the tensile shear strengths of the brazed joints after long-term thermal exposure experiments are in the average value range of 54 MPa with a non-essential difference in the standard deviation. The negligible decrease of the strengths of the joints can be explained by no significant growth of the reaction zone respectively in the Al_7Fe_2Si layer during the long-term thermal aging. All investigated joints show the same fracture mechanism. The fracture occurs in the Al_7Fe_2Si intermetallic layer in all cases [20]. Consequently, thermal aging at a temperature of 200 °C for 6, 48 and 120 h does not significantly influence the fracture mechanism as well as the tensile shear strength of the brazed joints. If it is necessary for the application, the joints can be thermally treated at a temperature below 200 °C after the brazing process. Moreover, it was proven that the brazed joints with a thin reaction zone (2 µm) ensure a good mechanical strength. It can be summarized that a reaction zone with a thickness of 2 µm does not negatively affect the tensile shear strength of the brazed joints.

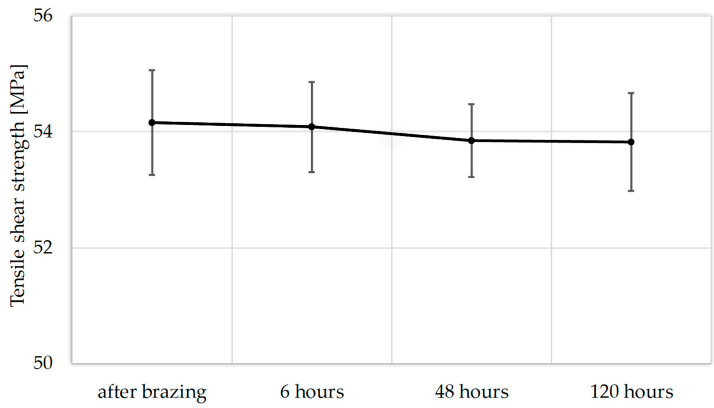

Figure 3. Tensile shear strength of brazed joints depending on the thermal exposure (200 °C)**.**

3.2. Long-Term Thermal Exposure Experiments at 500 °C and Tensile Shear Tests

In order to investigate the influence of a high thermal stress on the growth kinetics of the reaction zone in the brazed joints, long-term thermal exposure experiments were carried out at a temperature of 500 °C for 6, 48 and 120 h. Analogous to Figure 2, the joint after the brazing process in comparison to thermally stressed brazed joints is presented in Figure 4. During the thermal aging at 500 °C for 6 h, the diffusion of Fe atoms into the braze metal and their reaction with the Al and Si atoms from the braze metal causes the formation of a new intermetallic layer, Figure 4b. As reported in [21], the diffusion of Fe atoms into the aluminum is easier than that of Al atoms into the iron. With an increase of the thickness of the existing Al_7Fe_2Si layer, the diffusion of Fe atoms from the stainless steel into the aluminum-containing braze metal declines.

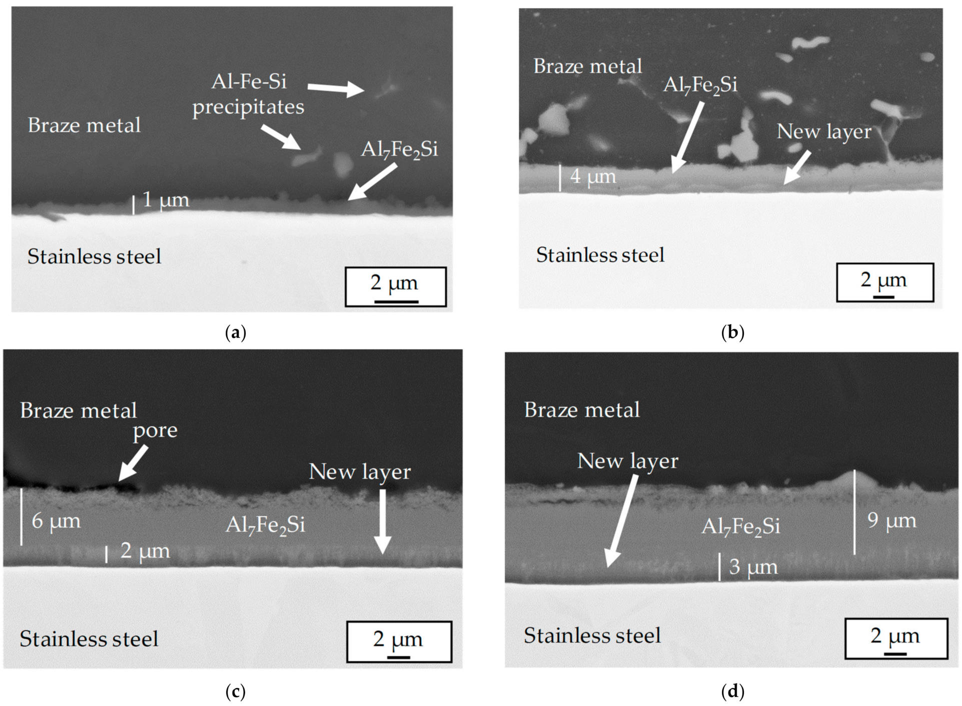

Figure 4. SEM images of the reaction zones after brazing (**a**) and after long-term thermal exposure experiments at temperature of 500 °C for 6 h (**b**), for 48 h (**c**) and for 120 h (**d**).

Hence, a new—compared to the Al_7Fe_2Si layer, energetically more favorable—iron-rich intermetallic layer is formed in the reaction zone. This layer consists of 54 at. % Al, 28 at. % Fe, 10 at. % Cr, 8 at. % Si. The chemical composition corresponds to the stoichiometry Al_2Fe (Cr, Si). Compared to the reaction zone with a thickness of 1 µm formed after the brazing process, the total thickness of the reaction zone increases to 4 µm due to the thermal aging for 6 h. During the further thermal aging, the diffusion of the Fe atoms proceeds. The total thickness of the reaction zone increases from 8 µm (48 h) to 12 µm (120 h), as shown in Figure 4c,d.

Additionally, it can also be seen that an Al_2Fe (Cr, Si) layer occurs in the form of columnar crystals, which preferably grow in the direction of the braze metal. The results of the thermal aging of the brazed joints show that a high thermal load causes the growth of the Al_7Fe_2Si intermetallic layer and the formation of a new intermetallic layer in the reaction zone. Moreover, with an increase of the duration of exposure, the presence of pores can be observed in the reaction zone along the Al_7Fe_2Si intermetallic layer, as shown in Figure 4c. This can cause the mechanical failure of the brazed joints [22,23]. It can be summarized that the total thickness of the reaction zones in the brazed joints significantly increases with an increase of the exposure duration during the thermal aging at 500 °C.

The results of the tensile shear tests of these joints are presented in Figure 5. It can be seen that the measured strengths of the joints after the long-term thermal exposure experiments decrease with an increase of the exposure duration in comparison to that of the joints after brazing process (54 MPa). This tendency can be explained by the growth of the existing Al_7Fe_2Si intermetallic layer and the formation and growth of the new intermetallic layer in the reaction zone. After thermal aging at 500 °C for 6 h, the strength decrease is not too sharp in comparison to the values of joints after further thermal aging for 48 and 120 h. Consequently, it was found out that the thermal aging at 500 °C for 6, 48 and 120 h influences the tensile shear strength of the brazed joints significantly. This difference in the strengths of these joints can be explained by a difference in the total thickness of the reaction zones.

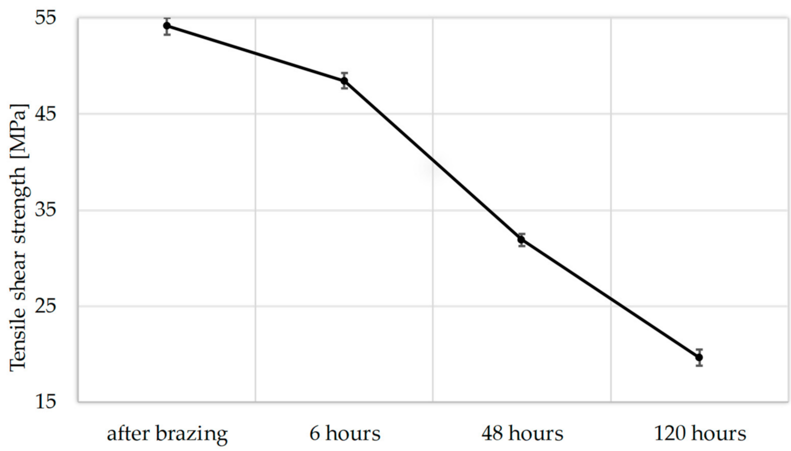

Figure 5. Tensile shear strength of brazed joints depending on the thermal exposure (500 °C)**.**

The fracture surfaces of the aged samples after the tensile shear tests were investigated using the top view and the cross sections. All tested joints show the same fracture mechanism. As an example for all investigated samples, the cross section of the fracture surface of the sample aged at 500 °C for 6 h after the tensile shear test is presented in Figure 6. It can be seen that the tested sample fails inside the Al_7Fe_2Si intermetallic layer. No delamination between the braze metal and the intermetallic layer was observed. According to Figure 6, the rough surface of the Al_7Fe_2Si intermetallic layer indicates a brittle fracture behavior in the brazed joint, as shown in Figure 7. Hence, it can be summarized that the reaction zone with a thickness above 2 µm affects the tensile shear strength of the brazed joints significantly.

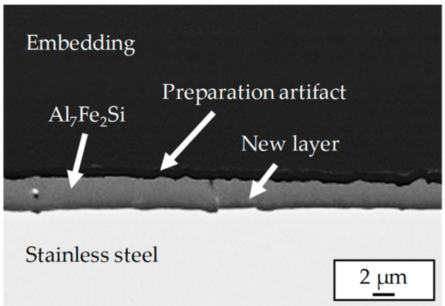

Figure 6. Cross section of the fracture surface of the sample aged at 500 °C for 6 h after the tensile shear test.

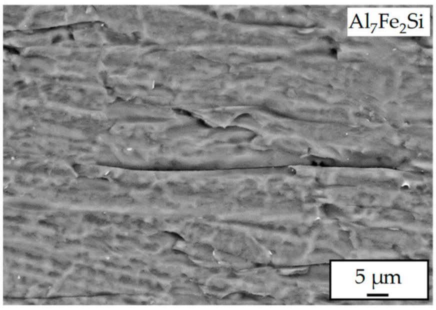

Figure 7. Fracture surface of the sample aged at 500 °C for 6 h after the tensile shear test.

4. Conclusions

Aluminum/stainless steel joints produced by induction brazing using an AlSi10 filler show a thin Al_7Fe_2Si intermetallic layer (~1 µm) at the interface to the stainless steel. The thickness of this layer increases to 2 µm depending on the exposure duration of the thermal aging at a temperature of 200 °C. The results of the tensile shear tests show that the brazed joints with this thin intermetallic layer ensure a good mechanical strength. Thermal aging at a temperature of 500 °C has a significant influence on the growth of the Al_7Fe_2Si intermetallic layer in the reaction zone of the brazed joints. In addition, a new Al_2Fe (Cr, Si) intermetallic layer is formed at the interface to the stainless steel. As a result, the total thickness of the reaction zone increases to a maximum of 12 µm during the thermal aging. Moreover, with an increase of the aging time, the presence of pores was observed along the Al_2Fe (Cr, Si) layer. The results of the tensile shear tests of the joints show that thermal aging at a higher temperature affects the tensile shear strength of the brazed joints significantly due to the formation of the thick reaction zones. Consequently, it can be summarized that the aluminum/stainless steel brazed joints with a thickness of the reaction zone above 2 µm cannot ensure a high tensile shear strength.

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