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Control of Formation of Intermetallic Compound in Dissimilar Joints Aluminum-steel

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A B S T R A C T

The elimination of the Fe_xAl_y type phases was considered the solution to low ductility presented in aluminum-steel welded joints. Recently, the researches do not seek the suppression but the control of the thickness of these compounds. In this work, Al-Fe joints were manufactured by solid state and fusion welding, looking for controlling the formation of intermetallic compounds. Temperature measurements were carried out during the welding. The joints interface was characterized using optical and scanning electronic microscopy, aided by chemical composition measures with X-EDS. The microstructural characterization at the interface of aluminum-steel joints, in solid state welded joints, demonstrated the absence of intermetallic compounds, which is attributed to the low temperature reached during the process - less than 300 ° C. In the case of fusion joints, it has observed the permanent formation of intermetallic compounds whose thickness varies significantly with the heat input.

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NOMENCLATURE

IMC	Intermetallic compounds	MZ	Mixed zone
FSW	Friction stir welding	TMAZ	Thermo-mechanically affected zone
GMAW	Gas metal arc welding	*L*	Deformation length (mm)
CMT	Cold metal transfer	*t*	Holding time (s)
FW	Friction welding	*v*	Welding speed (mm/min)
OM	Optical microscopy	υ	Wire feed rate (mm/min)
SEM	Scanning electron microscopy	**Greek Symbols**	
EDS	Energy dispersive spectrometry	ω	Rotation speed (RPM)
IMC	Intermetallic compounds	MZ	Mixed zone

1. INTRODUCTION

The union of materials such as steel and aluminum was used in transport systems to reduce the weight of the structure, with the consequent decrease in fuel consumption. Therefore, it is necessary to develop techniques that allow welding these materials without compromising the structural integrity of the vehicles. In general, conventional welding processes, whose main mechanism is the fusion of one or both elements, result in the formation of Fe_xAl_y compounds [1, 2], which are responsible for the low ductility of these joints due to the fragility of the intermetallic compounds (IMC). It was considered that the solution was the reduction or elimination of the said deleterious phases. Processes such as laser welding, diffusion, and ultrasound gave promising results by decreasing the amount of IMC [3, 4]. The total elimination of Fe_xAl_y phases came with the emergence of the Friction Stir Welding (FSW), which is founded in the union in the solid state, with a significant reduction in the heat input [5, 6]. Several authors studied the effect of different parameters such as tool position [7] and the advance and rotation velocities [8, 9]. Soundararajan and Kovacevic [10] and Tanaka et al. [11] who found that the thickness of the IMC increases with the heat input. In a series of papers, Torres and Ramirez

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[12, 13] developed welding parameters for Al-Fe joints without the formation of IMC [14], with very low heat inputs, where the mechanical strength of the system was lower compared with works where a small layer of a compound was reported.

The controversy was accentuated because of the results achieved with a new welding process, The GMAW by cold metal transfer (GMAW-CMT), which is a fusion process that allows reducing the heat input due to the combinations of two transfer modes of the GMAW process: short-circuit and globular [15]. This technology allowed the successful control of the IMC, generating a thin and homogeneous layer with a thickness between 5 and 10 μm [16 -18], which gives it an excellent mechanical behavior.

These results agree with the statements of Xue *et al*. [19] and Zhang *et al*. [20] who claimed that the formation of a fine layer of IMC is necessary to guarantee the metallurgical union of both materials, with a thickness that does not substantially compromise the ductility.

Currently, the debate persists in recent works such as Wang *et al*. [21], they insist in the negative influence of Fe_xAl_y IMC type, although this statement is based on results obtained in joints with IMC with a thickness superior to 10 μm. The important aspect is to note that both processes -FSW and GMAW-CMT- allows controlling the thickness and the homogeneity of the IMC layer, although the way how this compound is produced is different.

For the Al-Fe system, the formation of IMC rich in Al and Fe are expected, the former is considered as "fragile" IMC, while the latter showed higher ductility, for that reason it is called "ductile". The former is commonly observed in solid-state joints due to the diffusion of the Fe in the Al, forming compounds such as Al_2Fe, Al_5Fe_2, and Al_3Fe. The formation of the latter is promoted from the liquid due to the dissolution of solid Fe in the liquid Al accompanied by the diffusion of the Al atoms in the Fe forming compounds rich in Fe [22], highlighting the formation of $AlFe_3$.

Therefore, the union processes for this kind of system should promote the formation of a controlled and homogeneous thin layer of IMC, which guarantees the metallurgical union of both metals, looking for the generation of ductile compounds. For this reason, the present work looks to establish welding parameters through welding processes by friction welding and by gas metal arc welding (GMAW) that allows obtaining IMC in a controlled way.

2. METHODOLOGY

The joining process looks to generate IMC through two processes: one in solid state and the other by fusion. The processes of friction welding (FRW) and GMAW were considered; in both cases using parameters that leads to different heat inputs, with the intention of controlling the form of the IMC by means of the heat input.

2. 1. Obtention of Welded Joints by Friction Welding

The friction process is like direct or continuous drive friction welding. For the manufacturing of these joints aluminum-based alloys AA1100 and stainless steel AISI 304, both in cylinder shape of 3/4 in (19 mm) of diameter were used as base metals. The metal compositions are given in Table 1.

The welds were made in a butt joint, with the plug manufactured in stainless steel while the nut was made-up in aluminum, as it shows in Figure 1. The process was made with the nut mounted in a rotating element end the plug was fixed in the tailstock.

To control the heat input, variables such as rotational speed (ω), holding time (*t*) and the deformation length (*L*), that corresponds to the penetration of the plug in the nut, controlled through the difference between the plug length and the hole depth. The variation of the parameters is shown in Table 2.

From these values, significant parameters which allow comparing their effect in the formation of IMC were determined.

TABLE 1. Chemical composition (% weight) for AA1100 alloys and the stainless steel AISI-SAE 304

	Fe	Si	Cu	Mn	Mg	Zn
AA1100	0.95	0.95	0.15	0.05	-	0.10
	C	**Si**	**Cu**	**Mn**	**P**	**S**
SAE 304	0.08	0.70	0.20	0.15	0.04	0.03

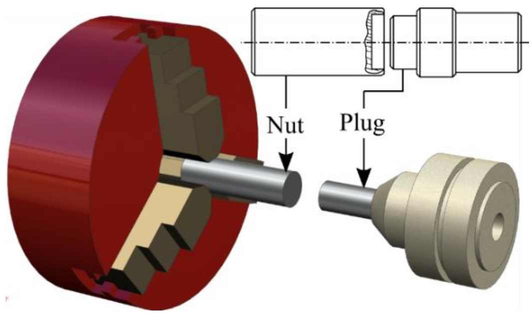

Figure 1. Montage and configuration of the joint for the solid-state welding (FW)

TABLE 2. Preliminary parameters to obtain IMC by FW

Parameter	P1	P2	P3
L, deformation (mm)	3 & 5	3 & 5	3 & 5
t, holding (s)	5	10	15
ω, rotation (RPM)	835	1320	2000

2. 2. Obtention of the Welded Joint by Gas Metal Arc Welding Aluminum alloy and structural steel ASTM A36 were used as the base metal, with the chemical composition shown in Table 3.

The elements were welded using the overlap configuration with a fillet welding bead, as it is shown in Figure 2. The plates use the present measures of 40 x 125 mm, with a thickness of 3/16 in (4.8 mm) for the AA6061 and 5/64 in (2.0 mm) for the A36. The distance overlapped is 1.0 in (25.4 mm). For the deposit wire filler ER4043 of 0.035 in was used, and welding made in drag direction.

Preliminary joints were manufactured to determinate suitable parameters, establishing that the minimum voltage to join both metals is 18 V, while the maximum is 30 V. the welding parameters are shown in Table 4.

2. 3. Temperature measures, microstructural and mechanical characterization In the case of FRW, temperature measures were developed using a type k (cromel-alumel) thermocouple, fixed in the plugs in a drilled hole, of 3/8 in (10 mm) of depth and at 3/8 in from the interface of welding. For data acquisition, a signal processing system with amplifier MAX6675 and an Arduino Mega 2560, with a response time of 180 ms was used.

A microstructural characterization of the joints obtained in both processes was carried out by optical microscopy (OM) and scanning electron microscopy (SEM); in the latter, chemical composition profiles were also carried out, using X-ray energy dispersive spectrometry (X-EDS).

Finally, hardness measures were carried out by micro-indentation with different loads and times of 15 s (HRV), for those joints that showed IMC formation. To reveal the microstructure in the welded joints, metallographic attacks were performed using 1% hydrofluoric acid for 5 minutes, for the joints by FRW and 2% Keller for 15 s, for the joints by GMAW. The mechanical properties of the joints were evaluated using tensile streght tests; the geometry of the samples and the parameters for the test were selected based on the ASTM E8 standard.

TABLE 3. Chemical composition (% weight) by rule for AA6061-T6 alloy and the A36 steel

	Fe	Si	Cu	Mn	Mg	Zn
AA6061	0.35	0.65	0.30	0.10	0.95	0.04
	C	**Si**	**Cu**	**Mn**	**P**	**S**
SAE A36	0.25	0.40	0.20	0.80	0.04	0.05

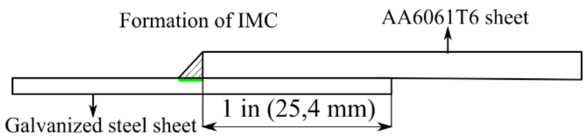

Figure 2. Configuration of the joint between AA6061-T6 and galvanized steel for GMAW process

TABLE 1. Preliminary parameters to obtain IMC by GMAW

Parameter	P1	P2	P3	P4	P5
Voltage (V)	18	20	22	25	30
ν (mm/min)			372		
υal (mm/min)			700		

3. RESULTS

3. 1. Welded Joints by FRW Welded joints of AA1100-304 were obtained, by means of solid-state employing FRW. The microstructural evaluation of the welded joints presented in Figure 3, reveals the formation of two clearly defined regions in the aluminum side: (1) the mixed zone (MZ) and (2) the thermo-mechanically affected zone (TMAZ).

The first one is generated by the high plasticization of the material, as consequence of the temperature and agitation of the material; the second one is produced by the deformation, without plasticization of the material, resulting in the deformation bands. The MZ is characterized by an accentuated refining in the grain size, as a result of the aluminum dynamic recrystallization, while in the TMAZ, the deformation bands are preserved, due that recrystallization of metal is not produced, similar to another process based on friction [23, 24]. On the other hand, in the region of the interfaces corresponding to the stainless steel, any microstructural transformation, promoted by the temperature or the deformation, which indicates that the temperatures and the stress are relatively low, were observed [25].

Ashfaq and Rao [26] determined that the grow in burn-off length increase the amount of extruded material, in which the difference in the shape of the MZ-flat or cone (Figure 3), is attributed to large and small burn-off, respectively. Li et al. [27] define that the conical shape of the MZ is obtained by using high forge pressure or a low rotational speed. The results shown in Figure 3 match with those described by Ashfaq and Rao [26], in which the low amount of plasticized material produces a cone in the center of the cylinder, where the welding speed is lower, which prevents homogenization of the plasticized material.

Despite the evident change in the MZ and the TMAZ, the variation in the parameters does not result in the formation of IMC (Figure 4).

One of the causes of the absence of the IMC phase is attributed to the maximum temperature.

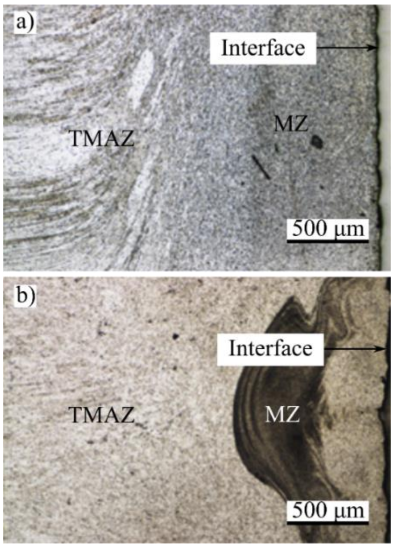

Figure 3. Micrographs by OM for the FW joints: a) 500 RPM, 5 s; b) 1150 RPM, 5 s

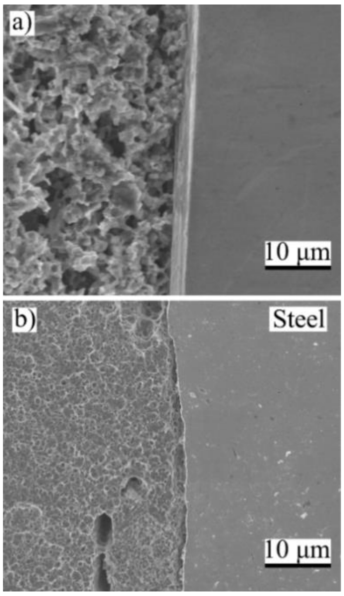

Figure 4. Micrographs by SEM of the interface for the joints of FW: a) 500 RPM, 5 s; b) 1150 RPM, 5 s

Although the increase of the rotational speed results in an increase in the temperature in the joint, in the point of measurement, it does not exceed the 300 °C, as it is shown in Figure 5. It is evident, the direct relation between the holding temperature and the rotational speed. The influence of the holding time is not as substantive as the rotation. Inferring that in the interface, the temperature is high, but it is considered insufficient to promote the diffusion, imperative for the formation of IMC.

The no formation of IMC in this work contrast with the results of Yilmaz et al. [28], who obtained metallurgical joints, thanks to the presence of $FeAl_3$ compounds. In this case, the weldings were manufactured with pressure control and pre-heating of the steel shaft up to 1000 °C, allowing the formation of a liquid layer in the interface, promoting the formation of the intermetallic.

The temperature reached to 300 °C which is lower than 500 °C reached by Chen and Kovacevic [3]; should also be pointed the lack of forging pressure during the welding and the continuous erosion of the steel surface by the plasticized aluminum as well as elements that prevent the formation of the compounds, which can have repercussions in the mechanical behavior of the joints, when the metallurgical joint is not guaranteed.

Torres and Ramirez [14] obtained IMC free joints, with temperatures equally low 350 °C in MZ during the FSW of Al-Steel joints, as consequence of the low heat input, in comparison to the other works addressed by the same authors.

3. 2. Welded Joints by GMAW. Because of the characterization by OM, it was evident the formation of IMC. The phase diagram of Al-Fe system shown in Figure 6 allows to establish the types of IMC generated.

The Al-steel interface is present in all the welded joints, and they were fabricated using GMAW as it is shown in Figure 7. A proper way to identify the IMC is because of their form and growth direction [19], where the Al_3Fe is identified by its elliptical form with growth in the aluminum direction, while that the Al_5Fe_2 looks like trapezoidal grains growing in the steel direction.

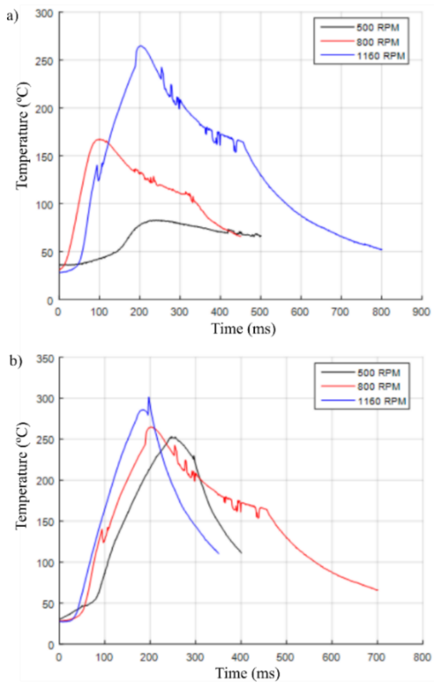

Figure 5. Temperature during the FW for different rotational speeds and holding times: a) 5 s, b) 20 s

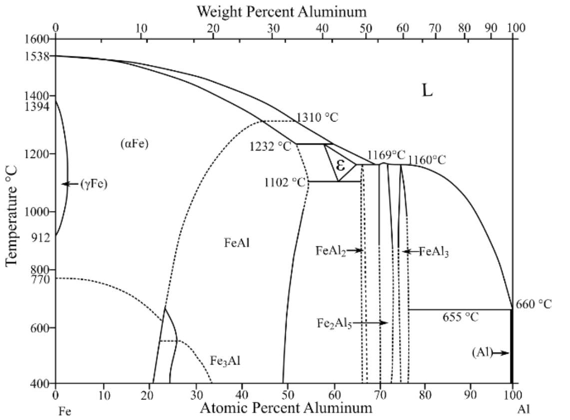

Figure 6. Phase diagram for Al-Fe system. Adapted from Bouche et al. [29]

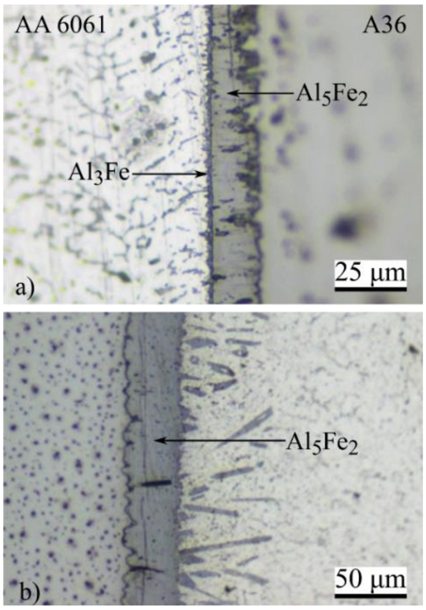

Figure 7. IMC Al_3Fe y Al_5Fe_2 for samples welded by GMAW for: a) 18 V, b) 30 V

Therefore, as described by Agudo *et al*. [30], It is possible to identify the formation of two types of IMC: Al_5Fe_2 and Al_3Fe; the first one with trapezoidal form growing in the steel direction while the second presents an elliptical form growing in the aluminum direction.

It is evident that the change in the thickness of the IMC layer along the Al-steel interface is due to the shape of the fillet bead, the heterogeneous distribution of the heat by the arc, and the differences in the thermal conductivity of both materials. To establish the relationship between the welding parameters and the IMC generated, tree measures of thickness were made in the points shown in the scheme of Figure 8, where the tree locals are referenced as left (L), central (C), and right (R).

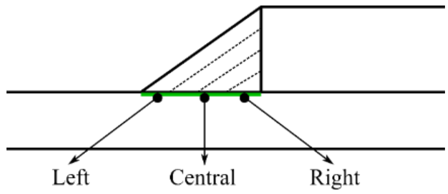

Figure 8. Scheme for the measure of the IMC thickness

These measures were carried out using the micrographs obtained by SEM, as shown Figure 9. The results of these measures are presented in Table 5. The size of the IMC along the Al-steel interface, in the fillet welding, is not uniform; this is attributed mainly to the heterogeneous transference of heat. As a consequence, it is observed that the thickness of the Al_5Fe_2 and the Al_3Fe becomes thinner in the extremes, growing near the center. The compound of larger thickness is the Al_5Fe_2, being the first in forming and present great growing kinetics, thanks to the high diffusivity of the aluminum atoms in a liquid state [31]. The Al_3Fe has less thickness because this compound grows from the Al_5Fe_2 [24].

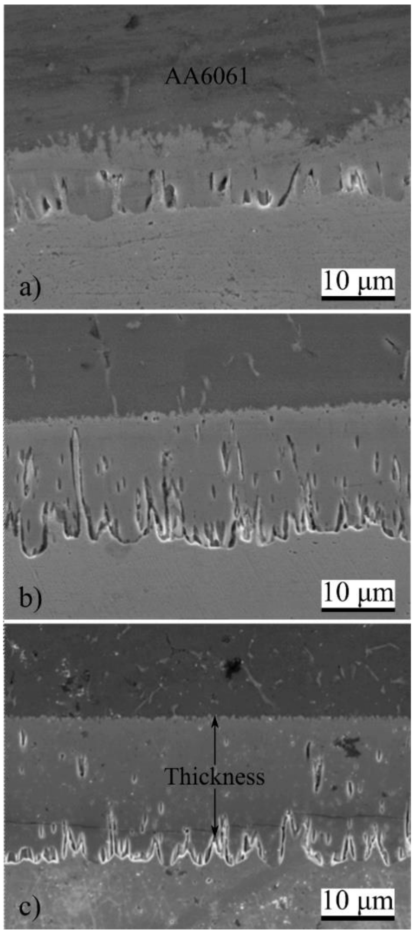

Figure 9. IMC for the welded joints with a) 18 V, b) 20 V and c) 30 V

TABLE 5. Thickness of the IMC along the interface: Left (L), Central (C), Right (R)

Thickness IMC						
IMC (μm)	**Zone**	**Voltage (V)**				
		18	**20**	**22**	**25**	**30**
Al_3Fe	L	4	4	5	5	8
	C	2	2	3	4	5
	R	3	4	4	4	5
Al_5Fe_2	L	15	9	10	8	20
	C	20	30	21	55	54
	R	5	12	11	26	12

From these results, final welding parameters are established; the values 18, 20, and 30 V, which correspond to the joints carried out with low (2.9 kJ/cm), medium (4.2 kJ/cm) and high (8.9 kJ/cm) heat input.

Measurements of micro-hardness were made in the specimens, in the IMC Al_5Fe_2 area near the center zone, using different loads to adjust the size of the indentation to the thickness of the IMC. Five measures were made, the results are illustrated in Table 6.

The average hardness of the IMC layer formed by the Al_5Fe_2 and The Al3Fe varies between 834-1294 HV. These values can be compared with the ones reached by Kobayashi *et al*. [32], who could measure in a separated way the Al_5Fe_2 and the Al_3Fe, reporting values of 1000 and 320 HV. For the Al_5Fe_2, Tomida and Nakata [1] established measures of micro-hardness around the 800 HV, while for the Al_3Fe the value was 200 HV.

In the final joints (18, 20, and 30 V), chemical composition measurements were made by X-EDS. The results are presented in Figure 10. These profiles show how in the region corresponding to the thickness of the IMC, limited by dotted lines in the figure, the chemical composition remains constant [14], showing the coherence of the result for being a phase with a fixed chemical composition [11, 20]. All the measures coincide in presenting this phase composed of aluminum and steel, with more content of the former, being that the measure adjusts with the Al_5Fe_2 compound.

Rathod and Kutsuna [22] report similar results, in which Al_5Fe_2 is registered due to its higher proportion, compared to the low percentage of Al_3Fe.

TABLE 6. Hardness of the IMC Al_5Fe_2

Voltage (V)	Load (g)	Hardness (HRV)
18	10	887 ± 110
20	25	871 ± 36
22	25	1294 ± 97
25	25	834 ± 88
30	25	1056 ± 199

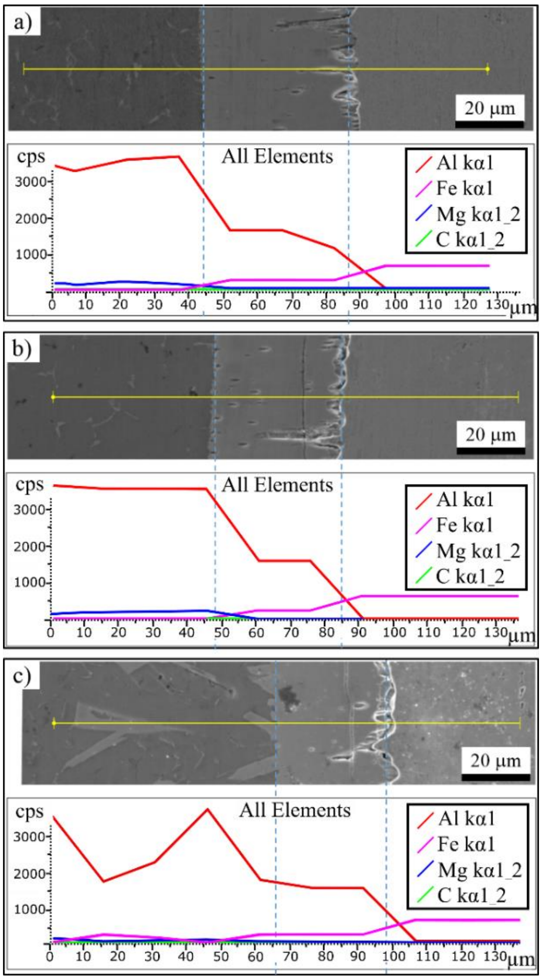

Figure 10. Chemical composition profile along the IMC in the welded joints: a) 18 V, b) 20 V y c) 30 V and c) 30 V

The largest thickness of the Al_5Fe_2 is explained through the formation sequence of the intermetallic compounds, which depends on how the joint is produced: in solid state or by fusion. In the first case, the IMC is generated in processes such as diffusion welding and friction welding; in the second one, the IMC is the result of processes with fusion such as arc welding. For the IMC produced from liquid, Agudo *et al*. [30] propose the following sequence for its formation: 1) Partial fusion of the aluminum, 2) wetting the surface of the steel, 3) dissolution of the steel (in solid state) in the molten aluminum, 4) atom diffusion of the aluminum in the steel, 5) formation of the Al_5Fe_2 phase from the steel matrix rich in aluminum and 6) heterogeneous solidification of the Al_3Fe compound from the Al_5Fe_2 due the Fe diffusion.

On the other hand, Chen *et al*. [33] suggest a different sequence starting with the formation of Al_3Fe that later reacts with the liquid aluminum forming the Al_5Fe_2. The beginning of the transformation from the Al_3Fe seems coherent, but results inconsistent with two microstructural pieces of evidences: The first one is related with the growth of several grains of Al_3Fe from a single crystal of Al_5Fe_2 (Figure 11), which indicates the heterogeneous nucleation of the first from the second. The second characteristic in relation to the wavy shape of the Al_3Fe interface and the matrix of aluminum, which indicates that the Al_3Fe is formed in the Al_5Fe_2-liquid interface.

A fundamental element for the manufacturing of the welded joints, and hence the formation of IMC, is the layer of galvanized in the A36 steel. This layer, with a thickness of 50 µm, is crucial because the zinc plays a role important due that acts as a flux, increasing the wettability of the steel, which favors the dissolution of the steel and the diffusion of Al atoms in the solid Fe [34].

The effect of zinc in the formation of IMC is also verified by Chen and Nakata [35] in solid-state welding by FSW, in Al-steel joints. Despite that the maximum temperature reached during the process is 479 °C -lower to the fusion temperature of the aluminum (660 °C) and steel (1538 °C)-, this is superior to the fusion temperature of the Zn (420 °C), forming a thin layer of liquid between both metals, increasing the diffusion, which promotes the compound formation.

The tensile strength tests' results of the samples welded by FRW and GMAW are presented in Table 7.

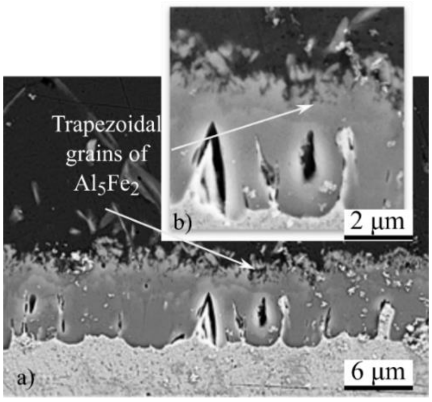

Figure 11. a) Growth of the IMC Al_3Fe from the trapezoidal grains of Al_5Fe_2. b) Detail showing the change in contrast due the presence of the Al_3Fe/ Al_5Fe_2 interface

TABLE 7. Results of tensile strenght test in FRW and GMAW welded joints

FRW		GMAW	
Sample	**σ (MPa)**	**Sample**	**σ (MPa)**
P1	94.7	P1	69.2
P2	102.3	P2	-
P3	94.2	P3	59.6
		P4	55.9
		P5	49.5

In the case of the joints welded by FRW the failure occurs in the aluminum, away from the Al-steel interface, as shown in Figure 12. As several authors indicate, the presence of IMC would guarantee the metallurgical joint between the welded elements [36]. The joining in the samples result non-metallurgical, due to the absence of IMCs; according Taban et al. [38] the mechanism involved is known as mechanical bonding, because of the molecular and mechanical nature forces due to the contact between surfaces [39]. In this case, the effect on the strength of the welded joint is not conclusive. Similar results were achieved by Sammaiah et al. [40] and Kimura et al. [41, 42] obtained welded joints with similar behavior, which they denominated as joints, with "high joint efficiency".

For joints welded by GMAW, the fracture is generated in the Al-steel interface, without indication of plastic deformation in the joint. The fracture is fragile, and the reached values of strength are inferior to those reached with AA6161-T6 (270 MPa). This is entirely due to the presence of IMCs. As it is indicated by Li et al. [27], when IMCs are formed, a significant reduction in fracture strength is observed.

The strength of the welded joint decreases as the thickness of the IMC increases [43, 44], which is confirmed in Table 2. Springer et al. [45] establish that the loss of strength as the thickness of the IMC increases is due to the increase in the number of porosities resulting from the Kirkendall effect in the IMC. Some of these porosities are observed in Figure 11, which weaken or even promote the formation of microcracks in the compound.

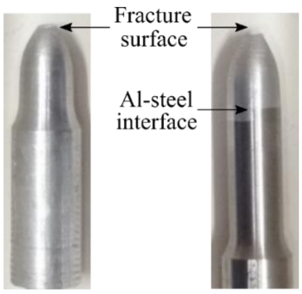

Figure 12. FRW sample after tensile strength test, showing the fracture region and ductile behavior of the joint.

4. CONCLUSIONS

Welding joints were manufactured using the FRW and GMAW processes to evaluate the formation of IMC. The process of FRW did not show the formation of IMC, which is attributed to the low temperature developed during the process, close to 300 °C.

All the samples welded by GMAW produced two types of IMC: Al_5Fe_2 and Al_3Fe, properly identified by their shape, as well as by the results of chemical composition and micro-hardness.

The Al_5Fe_2 presents significantly higher thickness compared to Al_3Fe, due to its primary formation during solidification, as well as the highest growth kinetics due to the high diffusivity of aluminum in the metal matrix of the steel.

The tensile behavior in the FRW samples is higher than in the GMAW, which implicate that even a metallurgical joining is not enough to guarantee of strength, in dissimilar welded systems. A mechanical bonding could have a better performance in the mechanical context, for joints of metals so different in chemical, mechanical, and physical characteristics, such as aluminum and steel.

5. RECOMENDATIONS

For the case of friction welding (FRW), it is recommended to use an alloy of highest strength, which allows the highest generation of heat during the application, trying to encourage the diffusion and thus the formation of IMC. Under this premise, it is important to apply longer holding times, in the same way as the previous measure. Likewise, it would be important to carry out solid-state joints, using a steel with a small layer of Zn, to promote the presence of liquid and with this promote the formation of IMC.

To perform temperature measurements in systems welded by GMAW, which allow linking the welding parameters with temperature peaks and holding times, and thus to try to establish a relationship that allows defining parameters for welded joints with less thickness to those presented in this job. To perform tensile tests, both in solid state welds and in those developed by fusion, which allows linking the thickness of the IMC with the mechanical behavior of the system.

6. ACKNOWLEDGMENTS

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Control of Formation of Intermetallic Compound in Dissimilar Joints Aluminum-steel

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چکیده

حذف فازهای FexAly به عنوان راه حل برای کم کاری ورق در اتصالات جوش داده شده با آلومینیوم در نظر گرفته شد. به تازگی، تحقیقات به دنبال مناسب بوده، بلکه کنترل ضخامت این ترکیبات است. در این کار، مفاصل Al-Fe توسط جوشکاری جامد و جوشکاری تولید می شوند، به دنبال کنترل ترکیب ترکیبات بین فلزات هستند. اندازه گیری دما در طی جوشکاری انجام گردید. رابط مفاصل با استفاده از میکروسکوپ الکترونی اپتیک و اسکن، با اندازه گیری ترکیب شیمیایی با X-EDS مشخص شد. خصوصیات میکروساختار در رابط مفاصل آلومینیوم و فولاد، در اتصالات جوش داده شده در حالت جامد، نشان دهنده عدم وجود ترکیبات متالولیک است که به دمای پایین رسیده است - کمتر از ۳۰۰ درجه سانتی گراد. در مورد اتصالات فیوژن، آن را مشاهده کرده است تشکیل دائمی ترکیبات intermetallic که ضخامت متفاوت با ورودی گرما را دارد.

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