Original Article

Intergranular corrosion evaluation of friction stir welded AISI 410S ferritic stainless steel

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Abstract

This study aimed to investigate the susceptibility of AISI 410S ferritic stainless steel to intergranular corrosion when friction stir welded (FSW) using the double-loop electrochemical potentiokinetic reactivation (DL-EPR) test. The highest values obtained for the ratio between the reactivation current (I\textsubscript{R}) and the activation current (I\textsubscript{A}) were found at the top of the advancing side for the two conditions tested. The steel for Condition 1, which was welded with a rotational speed of 800 rpm and high heat input, gave I\textsubscript{R}/I\textsubscript{A} peaks 60\% greater than Condition 2, which was welded with a rotational speed of 450 rpm and a lower heat input. These peaks were attributed to the presence of precipitates with high chromium content of about 21\%. In these FSW welds the sensitization of the AISI 410S steel was detected by the electrochemical test according to the intensity of the undesirable phases formed. The DL-EPR test was clearly able to quantify the different levels of sensitization in the FSW welds according to the energy used by the process parameters.

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1. Introduction

Friction stir welding (FSW) is a solid state welding process that was developed by The Welding Institute (TWI) in the 1990s [1]. This process is considered one of the most significant developments for the welding of metals in recent times [2]. The FSW process uses a rotating tool with a non-consumable tool-probe that is inserted into the butt joint to be welded, generating heat and intense plastic deformation [3]. The combination of the rotation and translation movement of the tool results in the movement of material from the front to the rear of the probe, consolidating the friction welding [4]. The FSW process was originally developed for low melting point materials, such as Al, Mg and Cu [5,6]. However, significant progress in the development of tool materials has enabled the welding
of materials with higher melting points [7,8]. Thus, a series of investigations have been made to find suitable parameters to develop FSW welds for different types of high quality steels [9].

Austenitic stainless steels are widely used by various industries [10]. Ferritic stainless steels provide approximately the same corrosion resistance as austenitic stainless steels in certain environments [11] and have less nickel in their composition [12], which is a great advantage since nickel is one of the most expensive alloying elements. This greatly reduces the manufacturing costs with ferritic alloys. However, these steels, when welded by the fusion welding processes, are subjected to temperatures high enough to cause excessive grain growth [13]. In addition, the formation of undesirable phases such as phase $\sigma$ and $\chi$ [14] and sensitization [15] of the material impair the corrosion resistance of ferritic steels.

However, this problem can be solved by applying and improving the FSW process for ferritic stainless steels. Some studies show a lower heat transfer to the material when compared to conventional fusion welding techniques [16,17]. These lower temperatures, achieved by the correct balance between rotational speed, axial force and welding speed, generate faster cooling rates and decrease the extent of the heat affected zone (HAZ). In addition, these lower temperatures may remain below sensitizing temperatures, thereby avoiding the formation of undesirable phases and precipitation of carbides, and thus avoiding intergranular corrosion [18,19]. This work aims to evaluate the effect of different FSW process parameters on the generation of heat, and their implications on the susceptibility to intergranular corrosion in AISI 410S ferritic stainless steel welds. In addition, a detailed evaluation of intergranular susceptibility was also performed.

### Materials and methods

The welds were made with 4 mm thick plates of AISI 410S ferritic stainless steel. The chemical composition of the material was obtained by optical emission spectroscopy (Shimadzu model PA7000 Japan) and is presented in Table 1. The welding in this study was carried out by the friction stir welding (FSW) process at the Helmholtz-Zentrum Geesthacht in Germany. All welded joints were made using the HZG Gantry System. An inert gas injection system with Ar was used to protect the material during the process, since at temperatures above 535 °C this stainless steel tends to react with oxygen or other elements present in the atmosphere. An integrated system to monitor the penetration depth, tool position, rotational speed, torque, axial force and welding time was used.

The tool has a conical diameter of 25 mm with a conical probe with a diameter of 9.2 mm and a length of 3.7 mm. The probe has a conical surface with the presence of negative recesses in the form of a spiral with respect to the axis of symmetry of the tool. The weld butt joints were performed in a single lap, in the flat position, as shown in Fig. 1.

To evaluate the effect of the process parameters on the generation of heat, two FSW welds were performed; one with a rotational speeds of 450 rpm and the other 800 rpm with axial forces of 22 kN and 20 kN, respectively, as shown in Table 2. The angle of inclination of the tool was maintained at 0° and the welding speed was 1 mm/s for both welds.

These parameters were related to the energy of the process. The heat input generated during the FSW welding was calculated according to Deqing et al. [20], as shown in Eq. (1):

$$E = \pi \cdot \mu \cdot Ps \cdot VR \cdot \frac{D^2 + D \cdot d + d^2}{45 \cdot (D + d)}$$

(1)

where $E$ is the heat input in J/m, $\mu$ is the friction coefficient of the material, $Ps$ the pressure exerted by the tool on the material in Pa, $VR$ is the speed of rotation in rad/s, $D$ is the diameter of the shoulder and $d$ the diameter of the probe in mm.

For the macroscopic analysis, the welds were initially cut with a diamond abrasive disc in a Struers Discotom-6 cutter. Sandpapers with granulometry between 120 and 2500 mesh were used for the grinding. The polishing step was performed in a universal polishing machine (Buehler Phoenix 4000) with diamond pastes of 3μm, 1μm and 1/4μm, and a rotational speed of 150 rpm. Subsequently, electrolytic etching with 40% nitric acid was carried out, using a voltage of 3 V for 2 min for analysis by optical microscopy (MO) with a Carl Zeiss optical system integrated with the Axio Vision SE64 software. A more detailed investigation of the possible precipitates and

### Table 1 – Chemical composition of the material (% weight).

<table>
<thead>
<tr>
<th>Material</th>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>P</th>
<th>S</th>
<th>Cr</th>
<th>Ni</th>
<th>Mo</th>
<th>Cu</th>
<th>Co</th>
<th>N</th>
<th>Fe</th>
</tr>
</thead>
<tbody>
<tr>
<td>410S</td>
<td>0.025</td>
<td>0.37</td>
<td>0.30</td>
<td>0.023</td>
<td>&lt;0.010</td>
<td>12.8</td>
<td>0.21</td>
<td>0.01</td>
<td>0.21</td>
<td>0.02</td>
<td>0.033</td>
<td>Bal.</td>
</tr>
</tbody>
</table>

### Table 2 – Welding parameters applied to the AISI 410S steel.

<table>
<thead>
<tr>
<th>Condition</th>
<th>Rotation speed (rpm)</th>
<th>Axial force (kN)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>800</td>
<td>22</td>
</tr>
<tr>
<td>2</td>
<td>450</td>
<td>20</td>
</tr>
</tbody>
</table>
deleterious phases formed was carried out using a FEI Quanta 250 scanning electron microscopy (SEM).

The double loop electrochemical potentiokinetic reactivation (DL-EPR) tests of the welds produced by FSW were performed at room temperature, about 30 °C, using a portable electrochemical cell. This cell is formed by a platinum counter electrode and a reference electrode consisting of a silver wire immersed in KCl solution using a working solution containing 0.1 mol/l H₂SO₄ + 0.4 mol/l Na₂SO₄ + 1000 mg/l KSCN. The cell was placed in contact with the sample surface using a flexible nozzle adhering to the material and the area of contact was approximately 0.8 mm². A total of 24 different points were used for the analysis. This allowed all the different welding zones, from the advancing side where the direction of travel is the same as the direction of rotation of the tool to the retreating side that has the opposite directions, to be evaluated as shown in Fig. 2. The current versus the potential curve was acquired with the Palm SensFc software loaded with the parameters, after stabilizing the potentiostat and using a constant scan speed of 3 mV/s.

3. Results and discussion

3.1. Heat input

The process parameters directly affect the heat input, which strongly influences the heating and cooling rates of the thermal cycle and consequently the resulting microstructure. However, the energy calculated based on the process parameters corresponds to an energy equivalent and not exactly to the energy produced during the process, since there are losses that are not considered, and the main ones are conduction and convection in the region of the weld.

The rotational speed is the parameter related to the frictional force and friction at the interface between the material and the tool, and is closely linked to the generation of heat during the welding process [21]. The engagement of the tool surface with the butt joint governs the heat input and the rotation of the tool results in agitation and mixing of the material around the probe. Thus, the higher the rotational speed, the higher the temperature reached during the process, due to friction [1,22]. The rotational speed of FSW welds has a strong influence on the generation of heat in AISI 410S ferritic stainless steel. This is clearly shown in Fig. 3 by the reduction of around 400 J/mm in the heat input when the rotational speed was reduced from 800 rpm to 450 rpm.

3.2. DL-EPR test

The sensitization of stainless steels is related to the segregation of certain elements to the grain boundaries of the material, mainly in the form of complex chromium carbides. In some regions, the material presents less than 12% chrome and is therefore susceptible to intergranular corrosion [23]. The research to determine the degree of sensitization of stainless steels led to the development of a simple, non-destructive and quantitative technique called the double loop electrochemical potentiokinetic reactivation test (DL-EPR). In this test, the polarization curve in the anodic direction, called the activation loop, promotes the formation of a passive layer in the material. This curve is compared with the reactivation loop, which evaluates the integrity formed when applying the reverse potential. The presence and intensity of the reactivation peak in the reverse polarization and the relationship between the reactivation current (Iᵣ) and the activation current (Iₐ) are a direct response to the susceptibility of the material to intergranular corrosion [24]. The DL-EPR test has been consolidated over the years as a simple and efficient technique.
to analyze the susceptibility of stainless steels to intergranular corrosion after they have been subjected to different types of processing, such as heat treatments and welding processes [25,26].

The DL-EPR tests are able to show whether the welding thermal cycle may promotes microstructural changes capable to make some areas of the weld susceptible to corrosion. The test also showed that the difference in the heat input observed in the welds of the AISI 410S ferritic stainless steel produced by FSW caused the formation of different levels of susceptibility to intergranular corrosion. Although the reactivation peaks in the current versus voltage graphs obtained by the technique (DL-EPR) were present for both Condition 1, welded with a rotational speed of 800 rpm, and Condition 2, welded at 450 rpm, the ratios of ($I_r/I_s$) were different.

In the FSW welding of the AISI 410S ferritic stainless steel the highest relation between the reactivation and activation peaks ($I_r/I_s$) occurred on the advancing side, and was higher for the welded condition with higher energy (Condition 1, Fig. 4). Normally chromium carbide precipitation in stainless steels requires some time to occur, due to the kinetics of nucleation and growth. However, it is possible that the effect of plastic deformation and recrystallization can act to accelerate this precipitation, especially on the advancing side, where this phenomenon occurs with greater intensity as reported by Park et al., 2004 [27], who verified this effect on the sigma phase precipitation on the advancing side of FSW welds of austenitic stainless steels.

For both conditions the $I_r/I_s$ ratio was more critical for the upper line, with values close to 0.22. Studies show there is a greater generation of heat on the advancing side of the FSW welds. The heat intensity produced in the upper part of the FSW weld is high, mainly due to the greater contact of the material with the tool shoulder, promoting a greater generation of heat [17,28]. Thus, the advancing side becomes a region critical to the occurrence of precipitates, as it is subjected to the effects of recrystallization and higher heat input.

When the rotational speed was reduced to 450 rpm, the heat input reduced from 998.5 J/mm to 562.6 J/mm. Consequently, there was a significant reduction in the temperatures reached and, furthermore, there was an increase in the cooling rate. These changes directly affect the results of corrosion resistance, as can be seen in Fig. 5 showing $I_r/I_s$ peaks around 0.08 and 60% lower than Condition 1. However, reactivation peaks were still found in the graphs obtained by DL-EPR, both in the stir zone and on the advancing side. Moreover, these reactivations were more critical in the upper line, as seen in Condition 1.
Vasconcelos and Silva [29] using the DL-EPR test evaluated the modifications caused by the autogenous TIG welding process with AISI 410S ferritic stainless steel. The DL-EPR tests showed that all weld regions were susceptible to intergranular corrosion due to the presence of fine Cr2N precipitates dispersed in the ferritic matrix. These authors determined that the presence of reactivation peaks in all current versus voltage graphs along the cross section of the weld, for all different levels of energy tested, had higher $I_r/I_a$ values than those found in the most susceptible regions to corrosion of the FSW welds evaluated in this study.

Lakshminarayanan and Balasubramanian carried out a study on the susceptibility to intergranular corrosion of AISI 409M ferritic stainless steel, immersed in a solution of copper sulfate and sulfuric acid [30]. The results of the analysis showed that, although this material presents better resistance to intergranular corrosion when welded by the FSW process in comparison to the TIG process, the use of a rotational speed of 400 rpm and an axial force of 22 kN still shows sensitization in regions subjected to higher temperatures. The same behavior was observed in the FSW welds produced in our study for the AISI 410S ferritic stainless steel welded under Condition 1.

3.3. Microstructural examination

The cross-section of an FSW joint shows an asymmetric arrangement of the different zones formed. The welding side where the direction of travel is the same as the direction of rotation of the tool is called the "advancing side". Likewise, the side that has the opposite directions is called the "retreating side" [31]. The cross-section of welded joints by FSW is divided into four different regions. The first region is the base metal (MB) which is unaffected by the heat or strain rate induced by the welding tool during the FSW process. The second region is the heat affected zone (HAZ), where the grains deform according to the processing and heat treatment of the alloy. This region is affected by the thermal cycle during welding, which leads to modifications of the microstructure and mechanical properties, but without any residual plastic deformation in the microstructure.

The third region is the area affected by both heat and plastic deformation and is called the thermomechanically affected zone (TMAZ); here the material is plastically deformed by the tool and the resulting heat flow exerts some kind of influence on the material, resulting in its recovery and/or recrystallization. The fourth region is known as the stir zone (SZ) and
corresponds to the central region of the weld. Here, the original grains and subgrain boundaries favor the formation of new fine equiaxial grains from the recrystallization due to the combined action of the shoulder and probe of the tool, as sources of heat generated by friction and plastic deformation simultaneously [31,32].

In spite of the different parameters, the two welding conditions presented very similar HAZ, TMAZ and SZ. The base metal of both conditions consists only of equiaxial ferritic grains, while in the thermally affected zone there are ferritic grains surrounded by martensite. Both conditions showed intense grain refining in the stir zone with coarser grains in the upper part and more deformed and refined grains in the lower region. The main microstructural difference between Conditions 1 and 2 is found in the TMAZ. In Condition 1, welded at 800 rpm and 22 kN, the increase of the rotational speed and the axial force generated an increase of the heat input and of the plastic deformation favoring the formation of the precipitates and of a greater grain refining in the advancing side.

In this work, the grain size changes, along the transition from the base metal, through HAZ, TMAZ and SZ, were mainly due to recrystallization. This resulted in a grain refining in TMAZ and SZ. However this grain refining was not the determining factor for the reactivation peaks, because for the same area with the same grain size the peaks of reactivations were different. The variation of parameters and the consequent change in the heat input, between Condition 1 and Condition 2, were the determining factors in the appearance of these precipitates and consequent reactivations. Therefore, it is not possible to make a direct correlation between grain boundary density and intergranular corrosion in these samples.

Although TMAZ is composed of recrystallized and deformed grains, the main microstructural changes observed in the weld zone were the martensitic transformations presented in the SZ, TMAZ and HAZ, as shown in Fig. 6 (a) and (b). This behavior is due to the low chromium content of the alloy, causing instability of the ferrite. During the heating, ferrite will partially transform to austenite at a temperature above 870 °C. At this temperature, most carbides can be dissolved and the carbon migrates to the austenite where it is absorbed and maintained in solid solution in the interstitial sites of the FCC lattice, due to the higher carbon solubility of the austenite. When the cooling rate is fast enough, the austenite will transform into untempered martensite supersaturated with carbon in solid solution, avoiding the Cr-rich carbide precipitation [33]. Thus, despite this important phase transformation occurring for both the conditions evaluated, the martensite formation along the SZ and HAZ, as a general rule, was not

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**Fig. 6** – Micrographs obtained by Scanning Electron Microscopy of Condition 1 of AISI 410S steel (a) Martensitic transformation in the stir zone and (b) Martensitic transformation in the heat affected zone, (c) Cross-sectional macrograph of Condition 1 of AISI 410S steel, (d) Probable formation of deleterious phases (10,000×), (e) precipitation of carbides (5000×).
Fig. 7—Analysis performed by EDS (energy dispersive X-ray detector) showing precipitates with 16.44 wt% of chromium.

directly associated with the corrosion results, since regions with a high volumetric fraction of martensite are not sensitive to DL-EPR.

The scanning electron microscopy analysis of Condition 1 for the E and F regions indicated in Fig. 4, are in the stir zone close to the advancing side. The results of this analysis indicated the presence of fine precipitates in the ferrite grain boundaries, which may indicate the formation of possible deleterious phases, as shown in Fig. 6(d) and (e).

The analysis by EDS detected high levels of chromium in these precipitates with values of around 16% (Fig. 7) and 21% (Fig. 8). These values were well above the 12.8% of chromium present in the base metal. This suggests that there is a chromium depleted zone in the regions surrounding these precipitates. Consequently, an electrochemical potential difference between the Cr-rich compounds and Cr depleted zone of the matrix develops. Thus, the particles act as a cathode and the Cr depleted matrix turns into an anodic region, as indicated in Fig. 6(d), rendering the steel susceptible to intergranular corrosion. This well-known mechanism of intergranular corrosion susceptibility is commonly observed for the precipitation of several Cr-rich compounds such as nitrides, carbides, α-phase and others [34,35].

Since chromium is the essential element in the corrosion resistance of stainless steels, these chromium depleted regions are susceptible to attack and thus the corrosion proceeds intergranularly, as shown in Fig. 9. The width of the chromium depleted zone normal to the grain boundaries increases with increasing annealing time and/or temperature. In the fully sensitized state it is found to be in the order of a few hundred nanometers [36,37]. This is illustrated in Fig. 6(d) where one of the arrow indicates the region that was strongly corroded around the precipitate due to chromium depletion.

An important finding in this study is related to the difference in microstructure and precipitation related to the welding parameters. Condition 1, which was welded with a higher rotational speed and, consequently, greater heat input, had a greater apparent precipitation along the grain boundaries. Thus, it is believed that the susceptibility to intergranular corrosion is, in fact, directly associated with the microstructural changes caused by the thermal cycle, especially with the precipitation of Cr-rich compounds formed along grain boundaries, which depends on the selected welding parameters. Decreasing the heat input, as in Condition 2, also reduces the tendency to sensitization, because of the higher cooling rate. In fact, higher cooling rates tend to suppress Cr-rich carbide precipitation. The time to precipitate will be shorter than those for slower cooling rates. Li et al. [38] evaluated the influence of rotational speed on the mechanical properties and corrosion sensitivity of friction stir welded joints. They showed that the rotational speed has a remarkable influence on the microstructure, with the best results found in the FWS joints for conditions welded with lower rotational speeds.

Lakshminarayanan and Balasubramanian [39], also analyzed the susceptibility of AISI 409M ferritic stainless steel to intergranular corrosion, through DL-EPR. These authors attributed the susceptibility to intergranular corrosion of welds produced with high heat input to the precipitation of Cr23C6 chromium carbides in the ferrite grain boundaries.

Recently, Kim et al. [26] reported another possibility for the intergranular corrosion mechanism when they analyzed 409L ferritic stainless steel subjected to temperatures of around 600 °C. These authors observed fine precipitates of Ti, Cr and C nucleates intergranularly, which favored the formation of regions with low Cr contents, compromising the resistance to intergranular corrosion of the material, as shown by the DL-EPR results. In another study, Kim et al. [40] studied the susceptibility to intergranular corrosion in welds of low Cr ferritic stainless steels. A depletion of the chromium content along the grain boundary was suggested, due to the TiC carbide precipitation, in which the presence of Cr was found to be associated with these particles. Although, the susceptibility to intergranular corrosion was not observed in the as-welded condition, it was seen after an aging treatment at 500 °C for at least 1 h.

Since the steel investigated in this work was not stabilized with an addition of Ti, the behavior in terms of corrosion resistance for the FSW tested conditions was different from that observed by Kim et al. [40]. The susceptibility to
Fig. 8 – Analysis performed by EDS (energy dispersive X-ray detector) showing precipitates with 21.06% wt of chromium.

Fig. 9 – Micrographs obtained by Scanning Electron Microscopy of Condition 1 of AISI 410S steel after DL-EPR test. (a) Intergranular corrosion behavior (5000×). (b) Cr-rich precipitates along grain boundaries (10,000×).
intergranular corrosion, observed by DL-EPR even in the as-welded condition, was only found in some regions in which precipitations were detected along the grain boundaries, as indicated in Fig. 7. In another region, in which the martensitic microstructure together with ferritic grains was found, but without any evidence of Cr-rich particle precipitations, the formation of a reactivation peak was not observed in DL-EPR. Thus, it is believed that a depletion of Cr is in fact associated with probable M23C6 carbide precipitations, evidencing the importance of the carbon stabilization effect produced by the addition of Ti, to suppress the M23C6 precipitation.

4. Conclusions

1. Welding of AISI 410S steel by the FSW process, using a rotational speed of 450 rpm and low heat input, produced joints with excellent susceptibility to intergranular corrosion, with low I/IS values and showing no evidence of microstructural changes detrimental to corrosion resistance.

2. Increasing the rotational speed to 800 rpm, regions susceptible to intergranular corrosion became more evident showing the presence of precipitations of Cr carbides. However, the values of I/IS are inferior to those presented by the same material when welded by the fusion process, as presented in the literature.

3. For both welding conditions tested, reactivation peaks were observed in the current versus potential graphs of the DL-EPR test. However, the highest I/IS ratios were observed in the stir zone of Condition 1, welded with the higher rotational speed and, consequently, a greater heat input.

4. Differences of the I/IS ratios were also observed between the advancing and retreating sides, due to differences in terms of the generation of heat, peak temperature and cooling rates.

5. The highest values obtained for the I/IS ratios were found at the top of the advancing side for both conditions.

6. The analysis by scanning electron microscopy detected the precipitation of chromium carbides and possible deleterious phases in the regions of the weld where the highest I/IS ratios were observed.

Conflicts of interest

The authors declare no conflicts of interest.

Acknowledgments

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