Evaluation of the diffusivity and susceptibility to hydrogen embrittlement of API 5L X80 steel welded joints

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ABSTRACT
This paper presents a study of susceptibility to hydrogen embrittlement of API 5L X80 steel welded joints by SMAW and GTAW processes. By varying the consumables used and the use of the same interpass temperature three different welded joints were obtained. Tests of hydrogen embrittlement susceptibility were performed according to ASTM G129-2006 with an aqueous solution (Solution A - TM0177/2005 NACE) sodium thiosulfate ($Na_2S_2O_3$) replacing the bubbling of H_2S. From the elongation values was observed that the joint obtained in all welding conditions showed susceptibility to hydrogen embrittlement, which was determined by the elongation ratio. The joints that showed higher levels of hardness showed higher susceptibility to hydrogen embrittlement. The joints obtained with higher welding speeds for the same amount of heat input presented a reduction in the rate of hydrogen embrittlement. All joints tested in solution showed fracture surfaces with quasi cleavage zones.

Keywords: API 5L X80 steel, Welded joints, Hydrogen embrittlement

1. INTRODUCTION

High Strength Low Alloy (HSLA) steels, like API 5L X80 steel, were developed to meet the needs of high strength and toughness required to produce pipes for oil and gas transport. In Europe and Japan the use of API 5L X80 steel is already quite significant. In Brazil this steel is being used for new distribution lines, but it is still subject of research, especially for evaluation of its weldability [1].

The major concern in studies of weldability is its behavior in relation to the degradation of properties caused by hydrogen. In the steels, particularly in welded joints, damage induced by hydrogen can occur during: the manufacture and construction as Cold Cracking; operation in a corrosive environment as Hydrogen Embrittlement (HE), Hydrogen Induced Cracking (HIC), Hydrogen Enhanced Stress Corrosion Cracking (HESCC) [2].

The HE is a type of failure that occurs in presence of atomic hydrogen and residual or applied stress. Since the hydrogen builds up in the crystal lattice, dislocations and other defects of the alloys potentially increases the possibility of cracking, blistering, and can

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consequently lead to catastrophic failure of the material [3]. The HE can occur when the steel is subjected to H_2S environments with atomic hydrogen produced on the surface by a corrosion reaction. In this case, it is called Sulphide Stress Cracking (SSC). This name is related to the type of environment in which the phenomenon occurs [4]. This type of degradation can be rapid and catastrophic; yet it is known that hard microstructures are more susceptible to this phenomenon. So, Heat Affected Zone (HAZ) of HSLA steel welded joint obtained with high cooling rate from low heat input can be more susceptible to this problem [5].

These processes are intensified in welded joints by metallurgical changes undergone by Base Metal (BM) when subjected to thermal cycles of welding. Thus, the challenge is therefore to maintain high strength without prejudice the toughness of the ZTA, especially considering the processes of corrosion and embrittlement [6].

Thus, the objective of this study is to evaluate the susceptibility to HE in H_2S environment, i.e. SSC, of three API 5L X80 welded joints with different welding parameters using Slow Strain Rate Testing (SSRT), according to ASTM G 129-2006 [7]. The solution employed was the solution A recommended by the standard NACE TM077/2005 [8], substituting the H_2S bubbling by addition of sodium thiosulfate. To assess the susceptibility to SSC the values of the elongations obtained in the tests in solution were compared with the values obtained in tests conducted in air.

2. EXPERIMENTAL PROCEDURE

In this work plates of API 5L X80 steel of 120 mm × 360 mm, 17 mm thickness and groove angle of 60° were used as base metal (Figure 1a). Three different welded joints were executed using different welding processes and welding parameters. Table 1 shows the welding processes and consumables to weld the three joints (T1.0, T2.0 and E1.0). The SMAW and GTAW process using Argon as shielding gas were used. Eight weld passes of welding, as

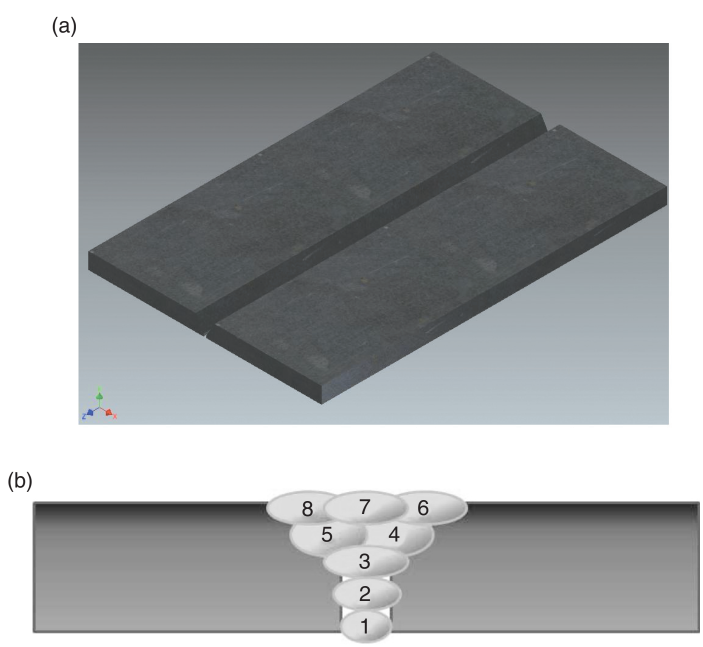

Figure 1 Welded joint; (a) sheet and groove representation and (b) welding sequence.

Table 1 Consumables used in welding conditions.

	Consumables				
	Bead number				
Sequence	**1 (root)**	**2**	**3, 4 and 5**	**6, 7 and 8**	**Welding processes**
T1.0	ER70S -3 - GTAW	E8010	E8018-G	E8018-G	GTAW+ SMAW
T2.0	ER70S -3 - GTAW	E9010	E9018-G	E9018-G	GTAW+ SMAW
E1.0	E6010 - SMAW	E8010	E8018-G	E8018-G	SMAW

illustrated in Figure 1b, were performed. The interpass temperature was 175 °C and the welds were performed without restriction. The spacing between the joints was the measurement of the diameter of the electrode used. The thermal efficiency was 0.8 used for SMAW process and 0.65 for the GTAW process according to Machado [9]. Table 2, 3 and 4 shows the welding parameters employed in the three welded joints.

Hydrogen permeation experiments were conducted with a Devanathan–Stachurski [10] assembly that was designed especially for these tests. The exposed surface area was 0.785 cm^2. The experimental set-up consisted of two identical electrolytic cells separated by the steel membrane. The charging surface was at the left of the corrosion potential in the test solution. The exit surface was held in a 0,1 N NaOH solution at a potential of + 300 mV vs. the saturated calomel electrode (SCE). Thus, all the hydrogen atoms diffusing through the membrane were oxidized. These tests were performed in order to determine the diffusivity, the permeability and solubility of hydrogen in the welded joints. The tests were conducted according to ASTM

Table 2 Welding parameters for T1.0 joint.

Bead number	I (A)	V (V)	H (KJ/cm)	v (mm/s)
1	152.4	12.04	9.55	1.25
2	68.8	33.38	12.22	1.50
3	110.9	20.88	16.13	1.15
4	130.6	20.11	9.24	2.27
5	130.9	20.52	12.38	1.73
6	130.8	20.85	10.29	2.12
7	130.8	20.16	8.88	2.37
8	130.7	19.73	9.87	2.09

Table 3 Welding parameters for T2.0 joint.

Bead number	I (A)	V (V)	H (KJ/cm)	v (mm/s)
1	156.6	12.56	13.23	0.96
2	86,5	34.56	15.13	1.58
3	113.4	23.49	17.62	1.21
4	116.6	21.68	10.82	1.87
5	116.5	21.74	9.57	2.12
6	116.5	21.42	10.83	1.84
7	116.7	22.29	8.68	2.39
8	118.5	21.85	7.90	2.62

Table 4 Welding parameters for E1.0 joint.

Bead number	I (A)	V (V)	H (KJ/cm)	v (mm/s)
1	54.2	35.76	15.50	1.00
2	81.8	33.17	14.09	1.54
3	160.5	21.78	15.71	1.78
4	165.5	21.20	10.31	2.72
5	165.3	22.19	10.70	2.74
6	165.1	22.40	11.83	2.50
7	165.4	22.19	10.15	2.89
8	165.3	22.52	9.48	3.14

G148-97 (2003) [11]. The charging solution was the Solution A (NACE TM0177/2005) [8] used with an addition of sodium thiosulfate (10^{-3} mol.l^{-1}). The pH solution was 3.1–3.4. This solution was proposed by Tsujikawa *et al*. [12] to simulate low levels of H_2S as an alternative to the solutions that are proposed by NACE that are quite aggressive and highly toxic [13]. When the specimen is immersed in this solution, H_2S (5.10^{-4} M) is generated on its surface in a significant range for 24 hours [12].

The SSC tests were carried out at a strain rate of $2.5 \times 10^{-5}.s^{-1}$, according to ASTM G 129-2006 [7], using cylindrical specimens according to ASTM E8/E8M-09 [14]. All tests were performed at room temperature. This test, termed SSRT, is an important tool to evaluate materials to be employed in the oil and gas industry, and can be satisfactory carried out a number of tests within a relatively short time [15]. The solution test was the same solution of hydrogen permeation tests.

Vickers hardness testing was performed on welded joints in accordance with standard N-133 [16] using a 5 Kgf load. Hardness measurements were performed at more points of middle of joints, as proposed by Rocha [17]. For the current analysis it was necessary, because when the specimens are machined to SSC tests, the regions containing the passes 1, 6, 7 and 8 are eliminated. Therefore it was necessary to analyze the central region of the weld and the results can be analyzed later with the SSC tests. The regions analyzed (Lines 2, 3 and 4) in the hardness testing are presented in Figure 2. Table 5 shows the average values of heat input of bead numbers 2, 3, 4 and 5.

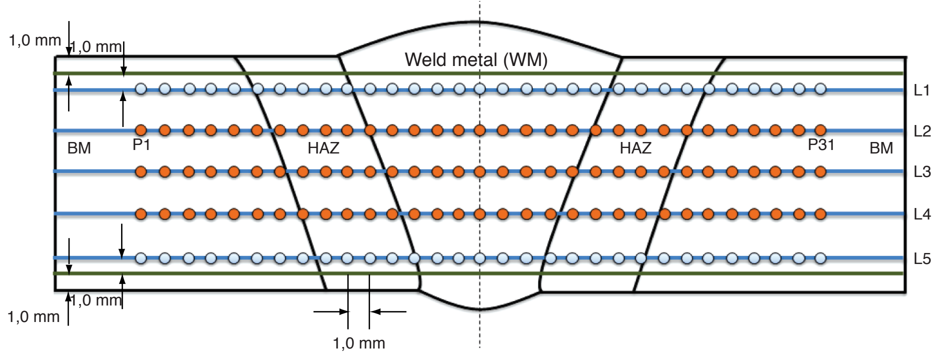

Figure 2 Points analyzed in Vickers hardness testing.

Table 5 Average values of heat input.

Welded joint	Heat input (KJ/cm)
T1.0	12.49
T2.0	13.28
E1.0	12.70

3. RESULTS AND DISCUSSION

Figure 3 shows hydrogen permeation curves for the BM. Table 6 shows the results of parameters obtained from the three hydrogen permeation curves. The parameters in the table represent an average from results of the three hydrogen permeation curves.

Evaluating the hydrogen permeation in API 5L X80 steel welded joints, Han *et al*. [18], found that for API 5L X80 (BM) steel the diffusivity was around 0.27 ∞ $10^{-10} m^2.s^{-1}$. The

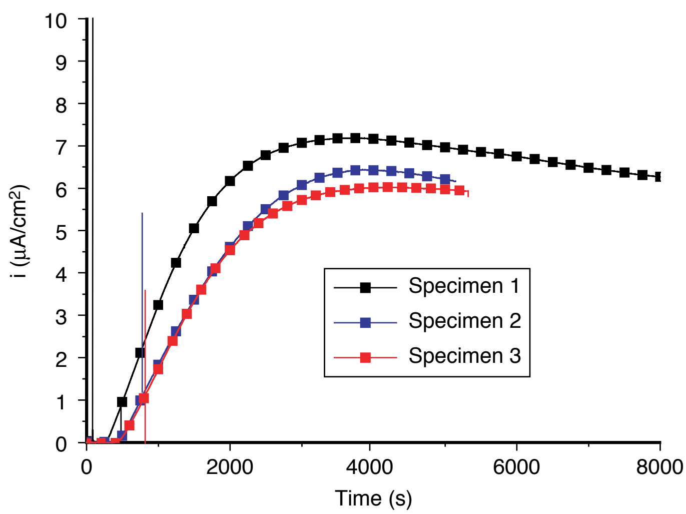

Figure 3 Hydrogen permeation curves to the API 5L X80 steel.

Table 6 Parameters obtained from hydrogen permeation curves.

	Diffusivity $x10^{-10}(m^2.s^{-1})$	Permeability $x10^{-10}(molH.m^{-1}.s^{-1})$	Solubility $(molH.m^{-3})$
API 5L X80	1.63	8.43	5.14

value obtained for steel in the present study is higher than that obtained by the respective authors. An important observation is that the steel is not the same in the two studies. The steel used in the present study showed a microstructure consisting of Ferrite and Bainite. In the study described by Han *et al*. [18], the microstructure consisted of Ferrite, Bainite and degenerate Perlite. The proportion between these microconstituents can influence directly the parameters obtained in hydrogen permeation tests, because it is known that the efficiency of hydrogen trapping, and consequently, decreased diffusivity, increase in the order of degenerate Perlite, Ferrite/Bainite and Acicular Ferrite microstructure (Park *et al*. [19]. Consequently, even the steel being cataloged as API 5L X80 steel, if they have different proportions between microconstituents may have different values of diffusivity, solubility and permeability.

The stress-strain curves of the welded joints obtained from tests are shown in Figure 4. The comparative analysis, based on data obtained is presented in Table 7, where it is possible to observe the values of yield strength (σ_{YS}), ultimate tensile strength (σ_{UTS}), elongation (*El*) and reduction in area (*RA*). Susceptibility to SSC joints tested can be evaluated according to the Elongation Ratio (*ER*), according to Equation 1, where the loss of ductility indicates the susceptibility to SSC between hydrogenated and non-hydrogenated samples [7].

$$ER = \frac{El_H}{El_{nH}} \tag{1}$$

where, E_{lH} and El_{nH} are the elongations of the hydrogenated and non-hydrogenated samples. The values of hardness obtained are shown in Figure 4.

From Figure 5 we see that the joint E1.0 had the lowest hardness values . The points of hardness assessed in Figures 5a, 5b and 5c are the points on the lines 2, 3 and 4, as illustrated in Figure 2. As mentioned earlier, only those lines were evaluated due to the machining process of specimens for testing hydrogen embrittlement.

Considering the passes number 2, 3 ,4 and 5 of the respective joints, it appears that due to the use of a higher heat input, the welded joint E1.0 showed a lower hardness values across the region of WM. The heat input average values was 12.49 KJ/cm to the T1.0 welded, 13.28 KJ/cm to the T2.0 welded joint and 12.70 KJ/cm to the E1.0 welded joint. The joints T1.0 and E1.0 were performed with the same consumables throughout almost the entire joint, differing only in root passes (Pass number 1), as shown in Table 1. Considering these two welded joints, although the average heat input used in the two conditions have had only a small difference, the fact that the joint E1.0 had been executed with a higher current contributed significantly to the hardness values presented and consequently in lower susceptibility to hydrogen embrittlement. The T1.0 and T2.0 welded joints were performed with different consumables in the passes number 2, 3, 4 and 5. For T1.0 welded joint, the pass number 2 and passes number 3, 4 and 5 were executed with E8010 and E8018-G consumables. In the T2.0 welded joint theses passes were executed with E9010 and E9018-G. It was expected that the T2.0 welded joint had higher hardness due to the use of consumables with greater mechanical strength, but the effect of the greater heat input used in T1.0 welded joint promoted approximate hardness values between the two joints.

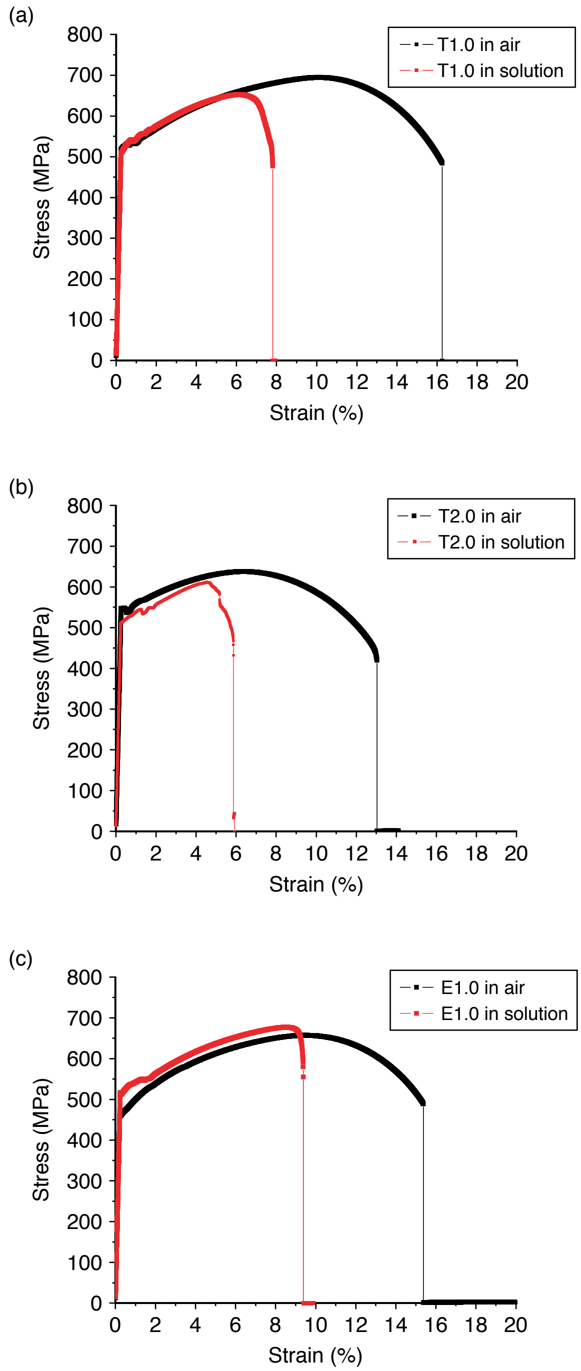

Figure 4 Stress-strain curves of the welded joints; (a) T1.0, (b) T2.0 and (c) E1.0.

Table 7 Parameters obtained in SSC tests.

	T1.0 in air	**T1.0 in solution**	**T2.0 in air**	**T2.0 in solution**	**E1.0 in air**	**E1.0 in solution**
σ_{YS} (Mpa)	527 ± 8.4	538 ± 4.2	531 ± 14.7	529.3 ± 18.5	509 ± 22.3	538 ± 3.0
σ_{UTS} (Mpa)	669 ± 35.3	657 ± 7.7	637 ± 0.5	603.3 ± 61.2	665 ± 20.2	675 ± 8.0
El (%)	18.5 ± 4.0	8.8 ± 0.2	16.6 ± 2.8	8.1 ± 2.3	21.2 ± 1.57	11.8 ± 1.0
RA (%)	49.6 ± 8.3	18.9 ± 6.3	57.7 ± 14.2	21.9 ± 5.1	66.5 ± 5.1	20.2 ± 3.7
ER	–	0.47	–	0.48	–	0.55

The parameters showed in Table 7 show that both welded joints had a significant loss of ductility when tested in solution. The elongations values to T1.0, T2.0 and E1.0 welded joints, when tested in air, were 18.5%, 16.6% and 21.2%. When tested in solution, these values were 8.8%, 8.1% and 11.8%, respectively. The *ER* values indicate a lower susceptibility of the E1.0 welded joint.

As it is known, the heat input is a parameter whose measure is relatively simple, being widely used in technical papers and standards to specify the conditions of welding. But its use should be done with care. There is not always a direct relationship between the heat input and its effects on joint because the welding parameters (current, voltage and welding speed) affect differently the intensity of the arc and the thermal efficiency of the process. Thus, while using the same process and welding energy, it is possible to obtain different welds formats by individual variation of the welding parameters [20]. Consequently, even employing the same or similar consumable and heat input in the passes number 2, 3, 4, and 5 in the T1.0 E1.0 welded joints, the fact that the E1.0 welded joint has been performed executed using a higher welding speed, can have generated a different microstructure in the WM. Whereas the studied phenomenon depends strongly on parameters of diffusivity, solubility and permeability, the microstructure presented by joints can contributes to the different results.

As mentioned earlier, the HE process in H_2S environment it is called SSC. The requirements for SSC based on the HE mechanism include a microstructure, a threshold level of HIC and an applied or residual stress [21]. For Arafin and Szpunar [22] and Koh *et al*. [23], the type of test used in this work, ie, SSRT tests, also indicates the susceptibility to Hydrogen Induced Cracking (HIC) and Sulfide Stress Cracking sulfides (SSC). Numerous studies have already presented, such as those performed by Park *et al.* [19] and Chang *et al*. [24] suggested that the microstructure of the steel plays a dominant role in HIC. Unfortunately, however, several studies have shown conflicting results on the role of the various microstructural constituents in respect of HIC. Chang *et al*. [24] showed that microstructures containing more hydrogen sites traps are more effective in reducing the susceptibility to HIC due to the fact that less hydrogen is available to participate in the process of cracking. Same conclusion was made in a study by Hardie *et al*. [25]. However, other studies, such as Huang *et al*. [26] showed that the lower the diffusivity of hydrogen in the microstructure of the steel, the higher would be the trapping efficiency and the steel would be more vulnerable to HIC. Studying the effect of heat input on the parameters diffusivity, the permeability and solubility of API 5L X80 steel welded joints, Han *et al*. [18] found that the use of a greater heat input resulted in a decrease in the effective diffusivity in different zones of the welded joint, and can result in a higher solubility of hydrogen.

Therefore, given the nature of the steel in question and considering the T1.0 and E1.0 welded joint, which were executed with the same consumables, it is likely that the use of a higher current and higher welding speed, keeping practically the heat input value at the

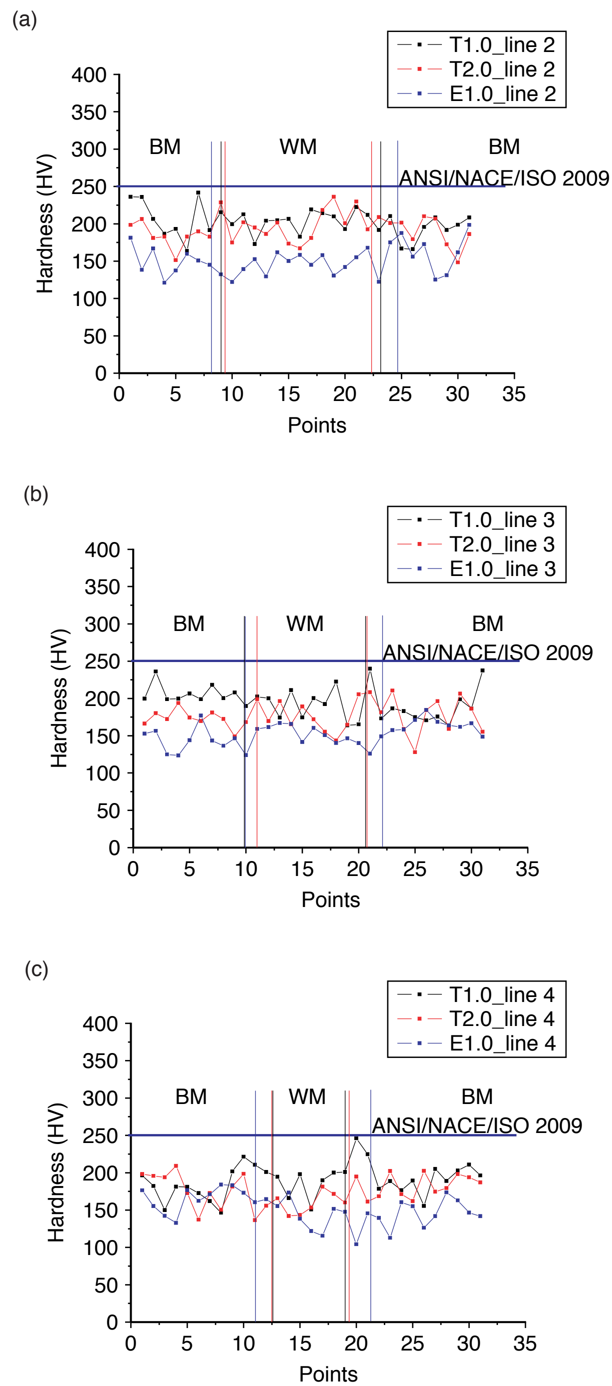

Figure 5 Hardness values for T1.0, T2.0 e E1.0 joints: (a) line 2, (b) line 3 and (c) line 4.

execution of E1.0 welded joint had caused a reduction in the diffusivity of hydrogen in the microstructure of the joint, reducing in this way the susceptibility to embrittlement, as noted in papers presented by Chang *et al*. [24] and Hardie *et al*. [25].

Evaluating the test results from the results of hardness, according to the standard NACE MR0175 [27], for typical industrial applications, steels having Rockwell hardness greater than 22 HRC (248 HV; equivalent to $\sigma_{YE} = 550$ MPa), are considered to have high susceptibility to SSC. For welded joints studied were not found hardness values greater than 248 HV, but it was observed values close to these values (between 215 and 240 HV). The T1.0 welded joint showed a larger area of high hardness. Comparing, for example, with the E1.0 welded joint which was performed with same consumables, it was found that a slight increase in heat input, favored by the use of a higher current, there was obtained a reduction of approximately 17% in their susceptibility to hydrogen embrittlement.

Although all welded joints have exhibited a ductile behavior when tested in air, analysis of the fracture surface of the welded joints tested in solution with sodium thiosulfate showed regions of quasi-cleavage fracture, which gives an indication of susceptibility to SSC as can be seen in Figure 6. The observed reduction in ductility can be understood in this case as a reduction in toughness of the material. This loss of ductility can be related also to the presence of secondary cracking near the fracture surface and in the more remote regions of the fracture surface of the specimens tested in solution.

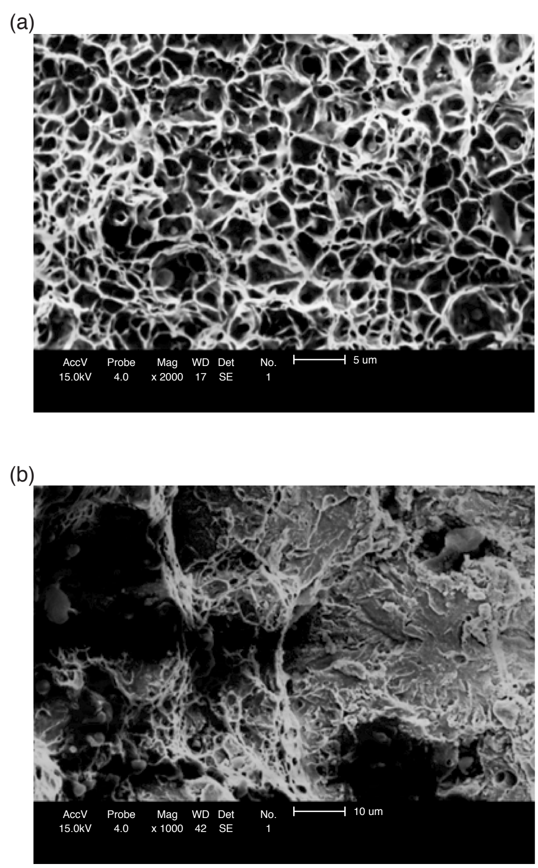

Figure 6 Frature surface of welded joints (a) T1.0 welded joint in air (2000X); (b) E1.0 welded joint in solution (1000X).

The secondary cracking commonly associated with the mechanism of recombination of hydrogen atom is promoted by the presence of non-metallic inclusions [15]. Secondary cracking was found in all joints tested in solution. Studies of susceptibility to hydrogen embrittlement in API 5L X80 steel welded joints also were executed by Ballesteros *et al*. [6] and Martins [28]. In all these works the welded joints of API grade steels exhibited susceptibility to hydrogen embrittlement when tested in sodium thiosulfate.

4. CONCLUSIONS

In the present study the susceptibility to SSC of three different welded joints was evaluated. The Embrittlement Hydrogen tests were quite effective, confirming the efficiency of replacement bubbling H_2S by additions of sodium thiosulfate. Based on these results we conclude that:

- All welded joints were susceptible to SSC, which has been verified for reasons of elongations obtained and the existence of quasi-cleavage regions of the fracture surfaces of samples tested in solution;
- The welded joints that had higher levels of hardness showed greater susceptibility to SSC;
- The welded joint with higher current intensity and welding speed for the same amount of heat input, showed a reduction in susceptibility to SSC.

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