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Monitoring pitting corrosion of AISI 316L austenitic stainless steel by acoustic emission technique: Choice of representative acoustic parameters

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This experimental work was aimed at investigating the monitoring of pitting corrosion by the acoustic emission (AE) technique, for pits developed by potentiostatic or galvanostatic polarization on two types of 316L austenitic stainless steels, in a 3% NaCl solution acidified to pH 2. The study of the evolution of AE global activity during the test showed the existence of a time delay before pits became emissive. This time delay and the AE events number rate measured during the propagation step of the pits are closely correlated with the sensitivity of the material towards pitting and with the polarization procedure. Moreover, the evolution of cumulative % of AE signals number versus selected acoustic parameters shows that rise time and counts number of signals appear to be discriminating acoustic parameters for monitoring pitting corrosion of austenitic stainless steels by acoustic emission technique in our experimental conditions, whatever the polarization procedure and the type of tested steel.

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1. Introduction

The acoustic emission (AE) technique, based on the rapid release of energy within a material generating a transient elastic wave propagation, is widely used as a non-destructive technique (NDT) for testing vessels on-site. Many microscopic deformation or fracture processes have also been studied with this technique in laboratory experiments, but most of them concerned stress corrosion cracking investigations [1]. Some published papers also deal with abrasion or erosion corrosion studies [2], and only a few attempts have been made to study purely electrochemical corrosion types such as uniform corrosion [3–5] or pitting corrosion [5–12]. In the latter case, the studies mainly concern aluminium alloys [7, 8, 10] and stainless steels [7, 9, 11, 12], in the presence of chloride ions. Moreover, different polarization procedures have been studied: evolution of the corrosion potential (in the presence of an oxidizing agent) [3, 7–10], galvanostatic polarization [7, 12] or potentiostatic polarization [11]. AE activity (number of events) has been correlated to the corrosion rate, which was estimated in terms of weight-loss, applied current density or hydrogen evolution rate. A direct quantitative correlation was even established between the number of AE events and the number of pits or the pitted area [12]. The most quoted mechanism responsible for the emission of AE signals is the evolution of hydrogen bubbles [5, 7, 8, 12]. Stress changes on metal surface [9], or rupture of an oxide or salt cap covering the pits [11] are also mentioned. Yet, to our knowledge, no comparative study of the acoustic parameters recorded during pitting, whatever the polarization procedure or the sensitivity of the material towards pitting has ever been made.

In that context, the aim of this work is to validate the use of acoustic emission technique for monitoring pitting corrosion on 316L austenitic stainless steels with different sensitivity towards pitting and different polarization procedures, and to evidence discriminating acoustic parameters.

2. Experimental method

2.1. Material and specimen preparation

AISI 316L austenitic stainless steel was used for this study. Two kinds of specimens, whose chemical

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TABLE I Composition of the studied materials

wt %	C	Si	Mn	Ni	Cr	Mo	S	P
316L sheet	0.02	0.45	1.78	11.76	17.20	2.40	0.006	0.027
316L bar	0.02	0.36	1.34	10.06	16.66	2.02	0.027	0.029

compositions are given in Table I, were studied: specimens were either sliced from an annealed bar and machined to a 65 mm diameter, 4 mm thick cylindrical plate (exposed area = 27.3 cm^2), or were cut out from a 2 mm thick rolled sheet (exposed area = 2.8 cm^2). Both types of specimens were wet ground up to 1200 grit silicon carbide paper. After a passivation treatment of 30 min in 20% HNO_3 at 60°C, the specimens were rinsed with de-ionised water then acetone, dried in a stream of cool air, and were stored overnight in a desiccator. That procedure leads to more reproducible data for the subsequent pitting behaviour.

2.2. Electrochemical environment

All the studies reported here were conducted at room temperature in 3% NaCl solution with the initial pH adjusted to 2 with HCl addition. For polarization tests, the electrochemically applied current or potential was controlled with an EG&G 273A potentiostat, the sample being the working electrode, a platinum mesh as the counter-electrode and a saturated calomel electrode as a reference. In order to avoid acquisition of acoustic noise induced by hydrogen evolution from the counter-electrode during anodic polarization of the specimen, the platinum mesh had to be placed in a near-by annex cell connected to the corrosion cell via a salt bridge (Fig. 1).

2.3. Acoustic Emission (AE) monitoring

AE instrumentation consisted of a transducer, a preamplifier and an acquisition device (MISTRAS from Physical Acoustic Corp.) (Fig. 1). The transducers were resonant R15D type from PAC (piezo-electric disks). They have been selected because of their high sensitivity around the value of 150 kHz. The acquisition system was completely computer controlled. The waveforms and the classical acoustic parameters (events number, amplitude, risetime, counts number) were stored on a hard disk as soon as detected, and were available for treatment under the form of ASCII files, as well as the electrochemical parameters.

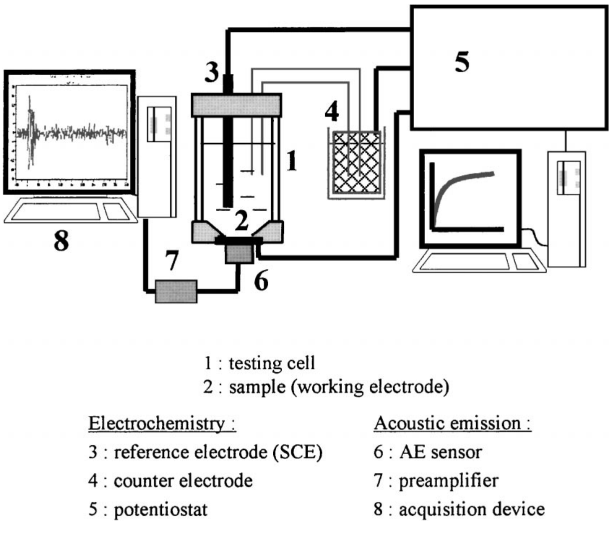

Figure 1 Experimental device.

2.4. Experimental procedure

Some potentiostatic tests were conducted on both 316L sheet and 316L bar specimens. Applied potential was chosen just above the pitting potential, which was previously determined through a 0.4 mV/s potentiodynamic test. 316L sheet specimens were then polarized at +600 mV/SCE, whereas 316L bar specimens were polarized at +200 mV/SCE.

In order to compare the present results to those presented in a previous paper [12], the same kind of tests was also carried out: after a 15 h stabilization step at open circuit potential, 316L bar specimens were anodically polarized by application of a constant current density of 0.25 mA/cm^2.

Except for one galvanostatic test, the duration of the polarization stage was chosen in order that the total number of AE signals recorded was the same (about 1500 signals), which allows to make statistical comparisons between the tests.

3. Experimental results

Experimental results are gathered in Table II.

For the specimens tested by potentiostatic polarization, it is first noticeable that two different kinetics of global AE activity are recorded (Figs 2a and 3a). Indeed, a time delay is existing before recording significant AE activity. During a time ranged from 1000 to 2500 seconds after the beginning of the test for 316L sheet, and during the first 1500 seconds for 316L bar specimens, AE events number rate is very low. After this time delay, AE activity increases sharply and AE events number rates reach the values from 0.3 to 0.4 events/s for 316L sheet and ranges from 0.5 to 1.6 for 316L bar.

It is therefore worth noting that the time delay tends to be longer for a material with a low pitting rate and shorter for a material with a high pitting rate. Indeed, the high number of inclusions such as MnS and the lower Mo content for the 316L bar specimens (Table I) makes

TABLE II Experimental results

Testing procedure	Material	AE results	Pits number and size
Potentiostatic	316L sheet	Time delay = [1000–2500 s] AE rate = [0.3–0.4 events/s]	3 or 4 large pits ($\Phi = 1.5–2$ mm)
Potentiostatic	316L bar	Time delay = 1500 s AE rate = [0.5–1.6 events/s]	Hundreds of small pits ($\Phi < 500\ \mu m$)
Galvanostatic	316L bar	Time delay = [1800–3500 s] AE rate = [0.03–0.3 events/s]	5 to 20 pits ($\Phi = 500–800\ \mu m$)

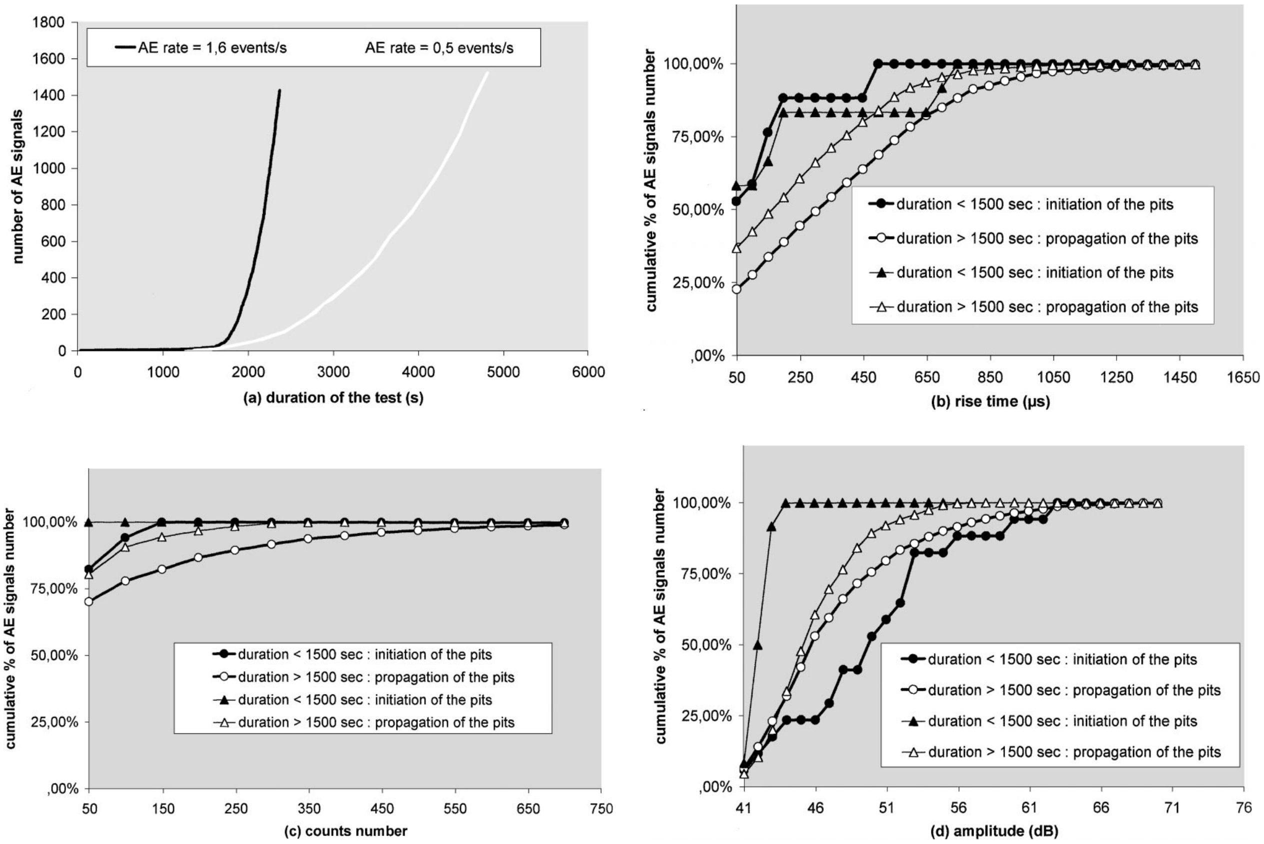

Figure 2 Evolution of the global AE rate (a) and of the cumulative % of AE signals number vs (b) rise time, (c) counts number and (d) amplitude, for 316L bar tested by potentiostatic polarisation, for initiation (black dots) and propagation (white dots) steps for two different tests.

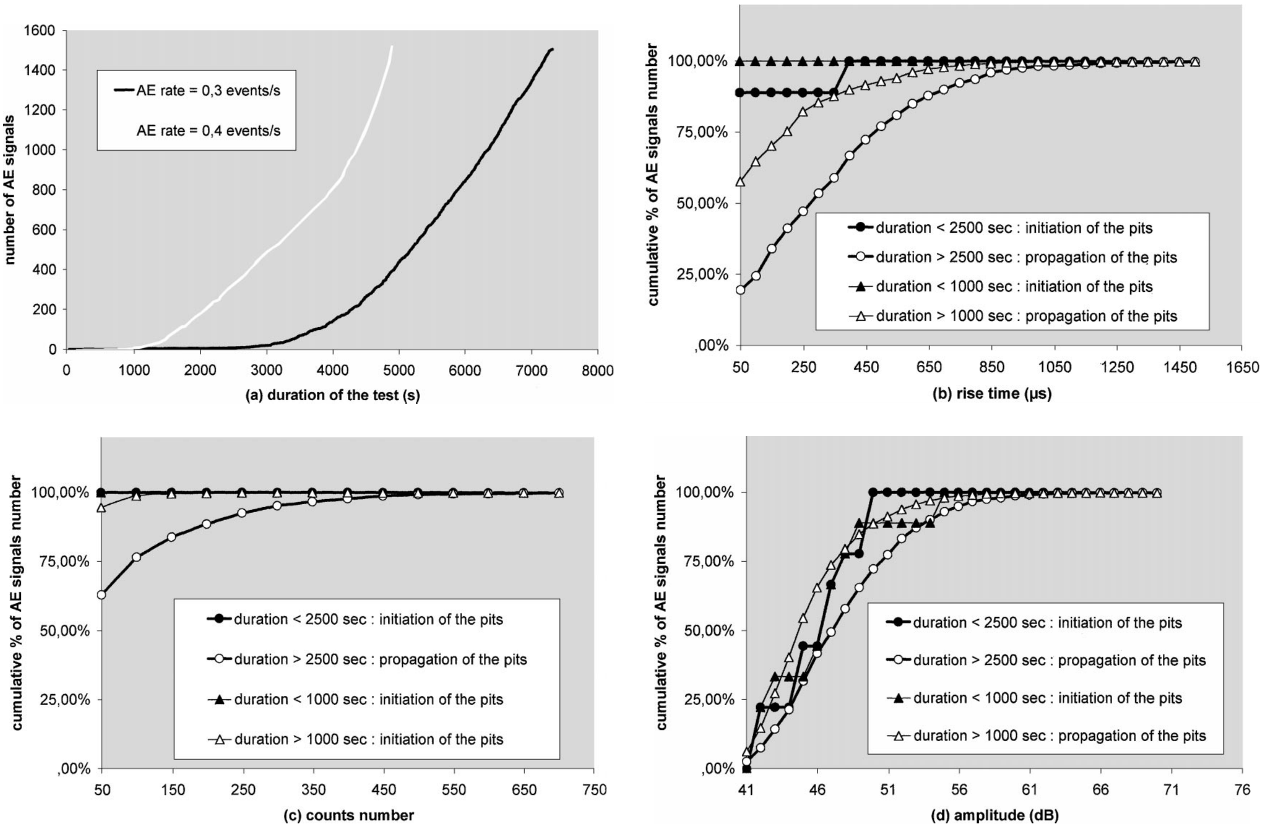

Figure 3 Evolution of the global AE rate (a) and of the cumulative % of AE signals number vs (b) rise time, (c) counts number and (d) amplitude, for 316L sheet tested by potentiostatic polarisation, for initiation (black dots) and propagation (white dots) steps for two different tests.

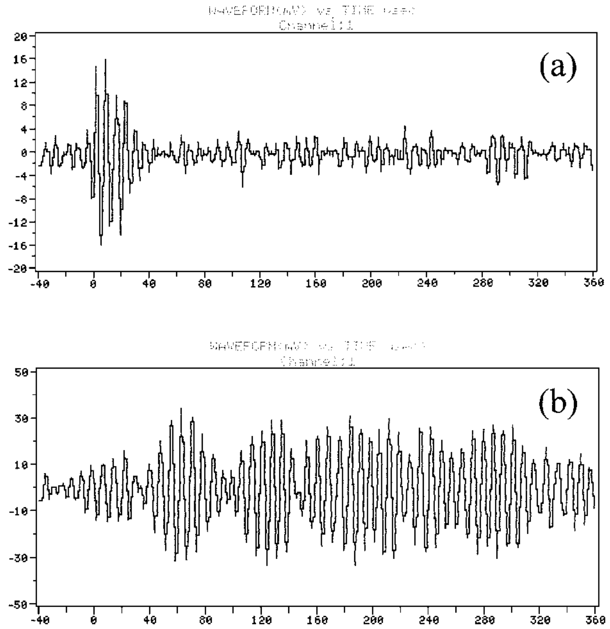

Figure 4 Waveforms of (a) short signals and (b) resonant signals.

the sensitivity of this alloy towards pitting much higher [13, 14]. The time delay is thus representative of the initiation step of the pits, which means that initiation step of pitting is not very emissive.

AE events number rate during development of the pits is also linked to the sensitivity of the material towards pitting as it can be four times higher for 316L bar specimens which are more sensitive.

As far as waveforms of AE signals are concerned, it is noticeable that, at the beginning of the test, only short signals are recorded (Fig. 4a), whereas signals become resonant when global activity increases (Fig. 4b).

This result is better illustrated by the evolution of the cumulative % of AE signals number versus acoustic parameters, i.e. the percentage of signals for which the parameter is lower than a pre-determined value. That evolution is presented on Figs 2b, c and 3b, c for initiation and propagation steps.

It is worth noting that rise time and counts number have different values whether they are recorded at the beginning of the test or at the end. For a 316L sheet specimen, during the initiation step, 90 to 100% of AE signals number have a rise time lower than 200 μs and 100% have a counts number lower than 100, whereas these numbers decrease to respectively 20% and 60% during the propagation step. The same tendency is observed for 316L bar (Fig. 3b and c), although it is less evidenced, especially for counts number.

Consequently, rise time and counts number increase during the test. No such a drastic effect is evidenced versus amplitude* (Figs 2d and 3d): the curves can be either very similar (316L sheet) or amplitudes can be either slightly higher or slightly lower at the beginning or at the end of the tests performed (316L bar).

The same kind of analysis was conducted on 316L bar specimens tested by galvanostatic polarization. Results are presented on Fig. 5.

As for the potentiostatic polarization, a time delay is observed before significant AE activity is recorded. This time delay varies from 1800 to 3500 seconds. After this period, AE events number rate reaches values ranged between 0.03 and 0.3 events/s.

At that point, a comparison can be made with 316L bar specimens polarized by potentiostatic testing. Indeed, it is noticeable that for a same material (316L bar), time delay is higher and AE events number rate is lower when the material is polarized by galvanostatic testing. This result is in good agreement with the fact that the development of the pits is completely controlled by the application of the current density, which limits it, whereas pits can develop freely and faster when potential is applied. AE events number rate is then well representative of the development step of the pits.

Moreover, the observation of the pits after potentiostatic testing show that pits developed on 316L bar specimens are much more numerous and smaller than those developed on 316L sheet specimens by potentiostatic polarization (Fig. 6). The influence of the pit morphology on their emissivity has been discussed elsewhere [15], but it can be assumed that the size of the pits is not a discriminating criteria for them to be emissive, and that an interaction phenomenon between signals produced by the simultaneous development of different pits should be considered.

This assessment is confirmed by the observation of the pits developed on specimens tested by galvanostatic polarization (Fig. 7). The specimen which presents the greatest AE events number rate and the lowest time delay, i.e. which is the most sensitive towards pitting, is affected by 5 pits that are very occluded and have developed very close together (Fig. 7a). The apparent pitted surface after manual removal of the metallic cap is then quite equivalent to that of one pit of a 316L sheet specimen tested by potentiostatic polarization (Fig. 6a), and which presents the same AE events number rate (0.3 events/s).

Moreover, the less sensitive specimen towards pitting (with high time delay and low AE number AE rate) presents 20 pits, but which are wide apart and not very developed (Fig. 7b).

In other respects, the evolution of cumulative % of AE signals number versus acoustic parameters is in very good agreement with the results presented just above for the potentiostatic polarization (Fig. 5b and c), as both rise time and counts number increase during the test. This effect is all the more pronounced when pitting is important. This result can be explained by the fact that the time delay for the less affected specimen is more difficult to determine precisely, as the increase of AE events number rate is less obvious (Fig. 5a).

Moreover, as for galvanostatic polarization, amplitude of AE signals does not seem to be a discriminating parameter for recording pitting by AE technique, as no discrimination can be made between the curves giving

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* Numerical results concerning amplitude are +20 dB shifted due to an acquisition software restriction.

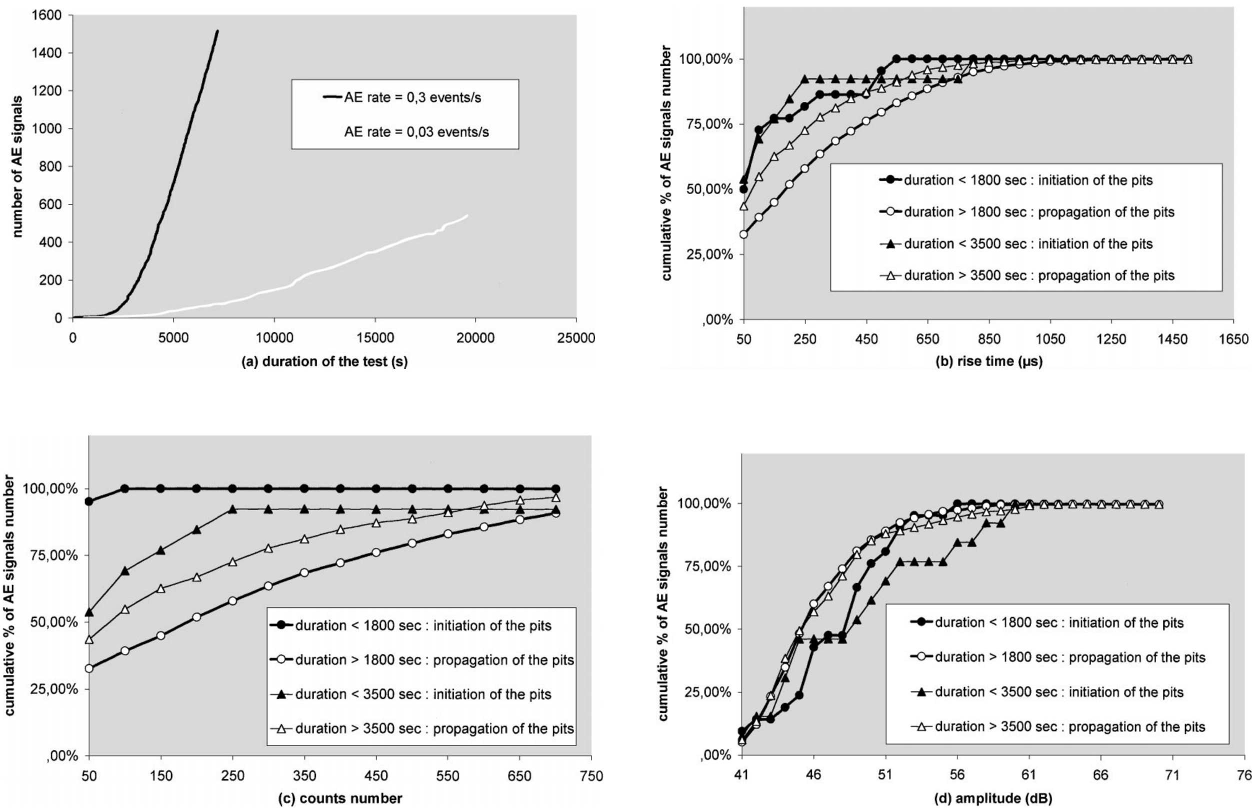


Figure 5 Evolution of the global AE rate (a) and of the cumulative % of AE signals number vs (b) rise time, (c) counts number and (d) amplitude, for 316L bar tested by galvanostatic polarisation, for initiation (black dots) and propagation (white dots) steps for two different tests.

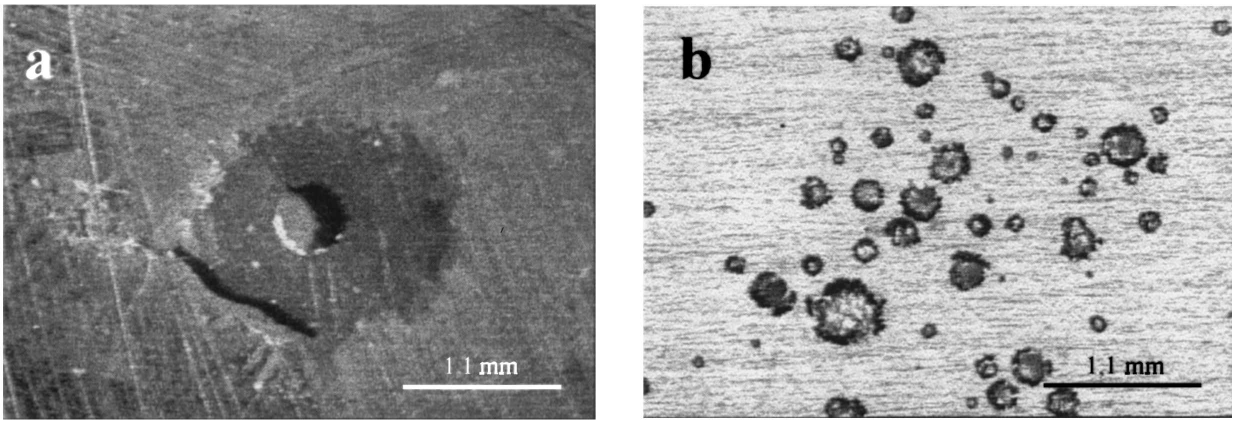
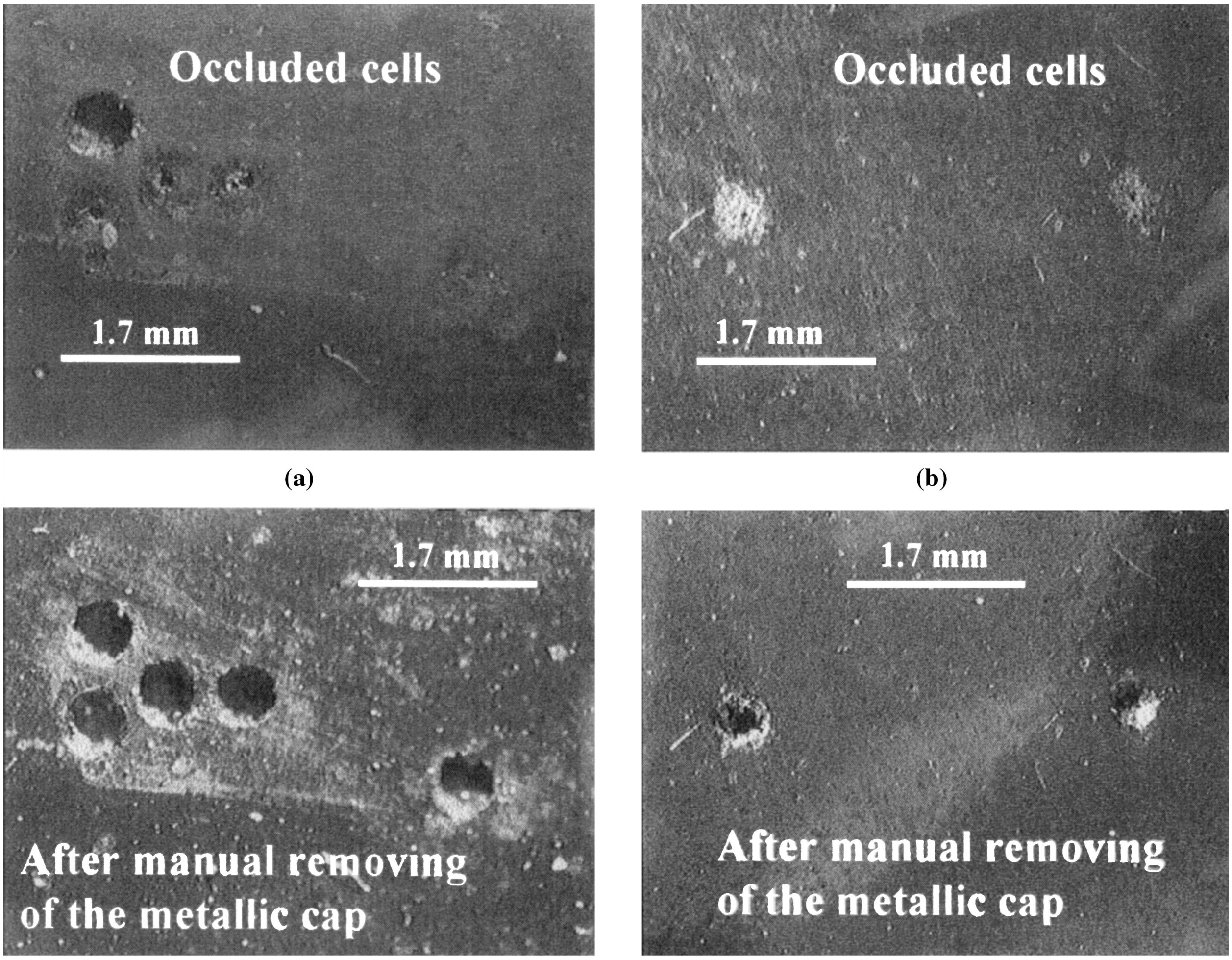


Figure 6 Morphology of the pits developed on 316L sheet (a) and 316L bar (b) tested by potentiostatic polarisation.

the evolution of cumulative % of AE signals number versus amplitude (Fig. 5d).

If we now compare the evolution of the acoustic parameters only during the propagation step of the pits, it can be evidenced that both rise time and amplitude of the signals are quite the same in all tested conditions (Fig. 8a and c). Counts number is also similar for different samples tested with the same polarization procedure (Fig. 8b). Yet, counts number of AE signals is different whether the polarization procedure: AE signals are more resonant (higher counts number) when constant current density is applied. If we assume that resonant signals are associated to the propagation step of the pits, whereas short signals are significant of initiation process, this result is in good agreement with the fact that, as soon as pits develop, less new pits initiate when constant current is applied than when constant potential is applied. The application of current density compels initiated pits to develop at a fixed rate, depending on the current density value.

Lastly, whereas no specific study of the physico-chemical source responsible for acoustic noise was carried out in the present work, it can be quoted that the results presented here are in good agreement with hydrogen bubbles evolution as acoustic source. The resonant character of signals recorded during pits propagation could be attributed to the "friction" of hydrogen bubbles along walls of the pits during their evolution.



(a) (b)

Figure 7 Morphology of the pits developed on 316L bar tested by galvanostatic polarisation. (a) time delay = 1800 seconds and AE events number rate = 0.3 events/s, (b) time delay = 3500 seconds and AE events number rate = 0.03 events/s.

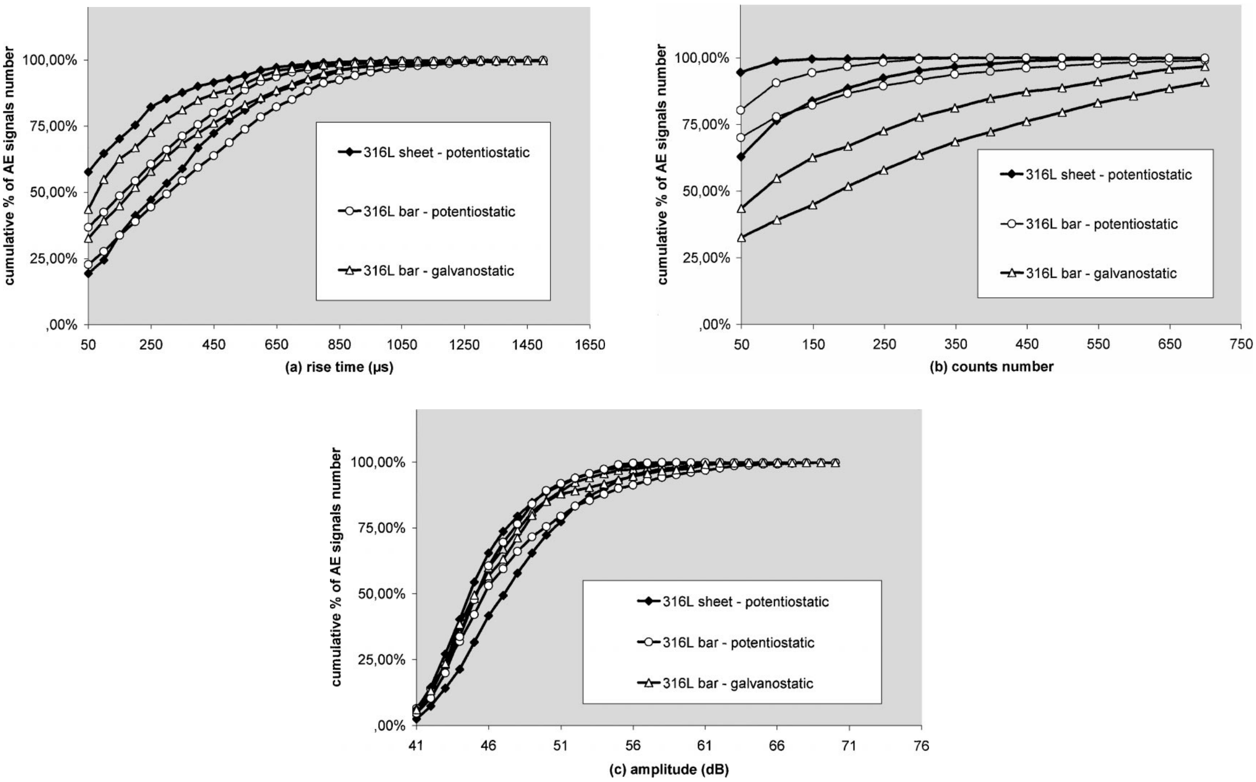


Figure 8 Comparative evolution of the cumulative % of AE signals number vs (a) rise time, (b) counts number and (c) amplitude, for 316L sheet and 316L bar tested by potentiostatic or galvanostatic polarisation.

4. Conclusions

From this experimental work aimed at investigating the monitoring of pitting corrosion by the acoustic emission technique, for pits developed by potentiostatic or galvanostatic polarization of two types of 316L austenitic stainless steel specimens, the following conclusions can be drawn:

– For all polarization procedures and the two types of tested specimens, the existence of a time delay has been evidenced before significant AE activity is recorded. It is then worth noting that during the initiation step of the pits, only few AE signals are recorded.

– Both time delay and AE events number rate are in very good agreement with the sensitivity of the material towards pitting and with the polarization procedure.

– AE activity becomes significant when pits propagate. The propagation of the pits is associated to the emission of signals with higher rise time and counts number, whereas amplitude remains the same whatever pits initiate or propagate.

– Therefore, rise time and counts number are discriminating acoustic parameters for monitoring pitting corrosion of austenitic stainless steels by acoustic emission technique, whatever the polarization procedure and the type of tested specimens.

– The present work shows the importance of studying separately initiation and propagation steps of pitting corrosion, as they are associated to the emission of different features of acoustic signals. Further experiments will be carried out next in that aim.

References

1. A. PROUST, P. FLEISCHMANN, H. MAZILLE and R. ROTHÉA, in “Corrosion Deformation Interaction CDI’92,” edited by T. Magnin and J. M. Gras (Les Editions de Physique, Les Ulis (F), 1993) p. 829.
2. F. FERRER, H. IDRISSI, H. MAZILLE, P. FLEISCHMANN and P. LABEEUW, *Wear* **231** (1999) 108.
3. K. H. W. SEAH, K. B. LIM, C. H. CHEW and S. H. TEOH, *Corros. Sci.* **34** (1993) 1707.
4. R. J. GROSSMAN, *IEEE Trans. Sonics and Ultrasonics* **SU-31** (1984) 25.
5. T. W. RETTING and M. J. FELSEN, *Corrosion* **32** (1976) 121.
6. D. L. PARRY, in Proceedings of 14th Conf. on Non-Destructive Evaluation, San Antonio, USA, 1983, p. 388.
7. F. MANSFELD and P. J. STOCKER, *Corrosion* **35** (1979) 541.
8. S. MAGAINO, *Boshoku Gijutsu* **34** (1985) 657.
9. S. MAGAINO, A. KAWAGUCHI, A. HIRATA and T. OSAKA, *J. Electrochem. Soc.* **134** (1987) 2993.
10. A. ARORA, *Corrosion* **40** (1984) 459.
11. R. H. JONES and M. A. FRIESEL, *ibid.* **48** (1992) 751.
12. H. MAZILLE, R. ROTHÉA and C. TRONEL, *Corros. Sci.* **37** (1995) 1365.
13. K. SUGIMOTO and Y. SAWADA, *Corrosion* **32** (1976) 347.
14. J. E. CASTLE and RUORU KE, *Corros. Sci.* **30** (1990) 409.
15. M. FREGONESE, H. IDRISSI, H. MAZILLE, L. RENAUD and Y. CÈTRE, in Proceedings of EUROCORR’99, Aachen (D) 1999, to be published in “Marine Corrosion of Stainless Steels” (EFC series, Institute of Materials in London, UK).

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